Environmental Impact of Radioactive Releases

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VIENNA, 8-12 MAY 1995
ENVIRONMENTAL IMPACT
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ENVIRONMENTAL IMPACT OF RADIOACTIVE RELEASES

PROCEEDINGS OF AN INTERNATIONAL SYMPOSIUM ON ENVIRONMENTAL IMPACT OF RADIOACTIVE RELEASES ORGANIZED BY THE INTERNATIONAL ATOMIC ENERGY AGENCY AND HELD IN VIENNA, 8–12 MAY 1995

INTERNATIONAL ATOMIC ENERGY AGENCY VIENNA, 1995
The early years of nuclear technology development, with their focus on production, caused contamination of the environment with radionuclides. In several countries, the health and environmental consequences of these early releases are now being evaluated. Examples include the radiation dose assessment and reconstruction programmes in the environments of nuclear facilities, weapons testing areas and marine waste dumping sites. Much of this renewed endeavour has been made possible by the improvements in relations between countries that have occurred in recent years.

Methods used for performing impact assessments have also been developed and improved by making use of the results from the extensive national monitoring programmes implemented following the Chernobyl accident. International projects such as the IAEA/CEC Validation of Environmental Model Predictions (VAMP) programme and the international Biospheric Model Validation Study (BIOMOVS) took advantage of the availability of the data as a means of testing the validity of environmental assessment model predictions.

The results of impact assessment studies are used as an input to decisions on remedial measures in the contaminated environments. The subject of environmental remediation, including criteria for aiding decision making and technical measures for environmental cleanup, is one of increasing importance.

The main purposes of the Symposium were to review recent developments and studies in the subject areas described above, as well as new information on the environmental transfer of radionuclides, and also to provide a forum for technical information exchange. In addition, the Symposium was intended to provide an occasion for presenting the main results of two major IAEA programmes: VAMP and IASAP (International Arctic Seas Assessment Project).

The Symposium was attended by 222 experts from 39 countries and five international organizations. There were 51 oral presentations and 90 poster presentations. The Symposium also included, in its final session, discussions on protection of the environment and the precautionary principle — topics on which there is much debate at the present time.

These Proceedings contain the text of all oral presentations and the extended synopses of the poster presentations.
EDITORIAL NOTE

The Proceedings have been edited by the editorial staff of the IAEA to the extent considered necessary for the reader's assistance. The views expressed remain, however, the responsibility of the named authors or participants. In addition, the views are not necessarily those of the governments of the nominating Member States or of the nominating organizations.

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*Global impact assessments*

(Session 1a)

Chairman

W. WHICKER

United States of America
EXPOSURES FROM WORLDWIDE RELEASES OF RADIONUCLIDES

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United Nations Scientific Committee on the Effects of Atomic Radiation, Vienna

Abstract

EXPOSURES FROM WORLDWIDE RELEASES OF RADIONUCLIDES.

Radiation exposures of individuals worldwide are caused both by natural radionuclide occurrence and processes and by man-made practices. The natural background of radiation has been extensively studied, as this forms the baseline upon which the man-made exposures occur. The man-made releases of radionuclides have included atmospheric nuclear weapons testing, routine operation of nuclear fuel cycle installations and accidents. These have given experience in tracing the movement through the environment of important radionuclides and in formulating models used for dose evaluation. The paper presents a summary of assessments performed by UNSCEAR of exposures from worldwide releases of radionuclides. As knowledge and experience increase, more accurate assessments can be made of past and future releases.

1. INTRODUCTION

A great deal of experience has been gained in the study of radionuclide behaviour in the environment and in the evaluation of the consequent doses to man. There are two main categories of radiation sources: natural and man-made. The natural radiation sources have always existed on Earth, and all living organisms are continually exposed. The doses to human beings from the natural radiation background are important, because they constitute the largest component of the collective dose received by the world's population. Doses from man-made sources are often compared with the reference level formed by exposure to the natural background sources.

Radionuclides have been released to the environment by several activities of man. By far the largest releases have occurred in the testing of nuclear weapons in the atmosphere. The most active periods of weapons testing were from 1952 to 1958 and in 1961 and 1962. No further atmospheric testing has taken place since 1980.

Several accidents at nuclear installations have released radionuclides into the atmosphere, most notably the accident at Chernobyl in 1986. The routine releases of radionuclides in normal operation of reactors and other nuclear fuel cycle installations have, in comparison to weapons fallout or the releases from accidents, been quite insignificant.
It has been noted that the specific environmental conditions at the times of the releases are factors in governing the dispersion and subsequent behaviour of radionuclides in the environment. The measurements of radionuclide concentrations in environmental media have allowed their movement and fate to be traced with accuracy and in considerable detail. The radionuclides released have served as tracers of the physical and biological transfers of material that have taken place. A variety of transfer models have been formulated, and these and the general understanding of the processes involved have been of quite general applicability.

In the following, the various sources of radionuclides in the environment are briefly considered, together with the main features of variability or transfer to man. The radiation doses per unit amounts of radionuclides released have been estimated. The knowledge of radiation doses received or of dose anticipated in specific circumstances can be used to plan and manage radionuclide releases and radioactive waste disposal programmes.

2. NATURAL RADIATION SOURCES

The sources of natural radiation include those of extra-terrestrial origin (i.e. cosmic radiation) and of terrestrial origin (i.e. radionuclides in the Earth’s crust, in building materials and in air, water and foods). These sources cause both external exposures and internal exposures, when the radionuclides are inhaled or ingested. Some of these exposures are relatively constant and uniform for all individuals throughout the world, for example the dose from ingestion in foods of $^{40}$K, an element that is homeostatically controlled in the body, and from cosmogenic radionuclides that are relatively homogeneously distributed over the surface of the Earth. Other exposures vary widely, depending on location, for example greater cosmic ray intensity at higher altitudes and elevated concentrations of uranium and thorium in soils in localized areas. Exposures can also vary because of human activities and practices. In particular, the building materials of houses and design and ventilation systems strongly influence the indoor levels of radon and thoron and their decay products, which can contribute significantly to internal doses through inhalation.

A number of naturally occurring radionuclides give rise to external and internal absorbed doses in the body. A summary of the various contributions is presented in Table I. The average dose to the world’s population from natural radiation sources has been estimated to be 2.4 mSv, of which one third is external exposure and two thirds internal exposure [1]. The highest component of the annual effective dose (1200 $\mu$Sv) comes from inhalation of radon and its short lived decay products. Of next importance is the dose from external irradiation (840 $\mu$Sv), arising approximately equally from cosmic radiation and terrestrial sources. Less significant contributors to dose are ingestion of $^{40}$K (170 $\mu$Sv), inhalation of $^{220}$Ra and its decay
TABLE I. ANNUAL DOSE FROM NATURAL RADIATION SOURCES IN AREAS OF NORMAL BACKGROUND

<table>
<thead>
<tr>
<th>Source</th>
<th>Annual effective dose (μSv)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>External</td>
</tr>
<tr>
<td>Cosmic rays</td>
<td>380</td>
</tr>
<tr>
<td>Cosmogenic radionuclides</td>
<td></td>
</tr>
<tr>
<td>Terrestrial radionuclides</td>
<td></td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>130</td>
</tr>
<tr>
<td>$^{238}$U series:</td>
<td></td>
</tr>
<tr>
<td>$^{238}$U - $^{234}$U - $^{230}$Th</td>
<td>140</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td></td>
</tr>
<tr>
<td>$^{222}$Rn - $^{218}$Po</td>
<td></td>
</tr>
<tr>
<td>$^{210}$Pb - $^{210}$Po</td>
<td></td>
</tr>
<tr>
<td>$^{232}$Th series</td>
<td>190</td>
</tr>
<tr>
<td>Total (rounded)</td>
<td>840</td>
</tr>
</tbody>
</table>

Terrestrial radionuclides contribute very little. Variations about the mean values by factors of 5 to 10 are not unusual for many of the radionuclides. The greatest variation occurs for indoor radon concentrations, which span over four orders of magnitude. Continued surveys of natural background radiation levels will provide more representative average values and better define the high background areas to which many populations are exposed.

3. NUCLEAR EXPLOSIONS

The use and testing of nuclear weapons in the atmosphere, which occurred from 1945 to 1980, has caused the greatest man-made release of radioactive materials to the environment. A single large explosion of 1 Mt fission yield produces 4200 PBq of $^{131}$I, 3.9 PBq of $^{90}$Sr and 5.9 PBq of $^{137}$Cs.
TABLE II. RADIONUCLIDE RELEASES TO THE ENVIRONMENT FROM MAN-MADE SOURCES

<table>
<thead>
<tr>
<th>Activity or event</th>
<th>Released amount (PBq)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{90}\text{Sr}$</td>
<td>$^{131}\text{I}$</td>
</tr>
<tr>
<td>Atmospheric nuclear testing</td>
<td>600</td>
<td>650 000</td>
</tr>
<tr>
<td>Nuclear power production</td>
<td>5.8$^a$</td>
<td>0.048</td>
</tr>
<tr>
<td>Chernobyl accident</td>
<td>8.1</td>
<td>1670</td>
</tr>
<tr>
<td>Windscale accident</td>
<td>–</td>
<td>0.74</td>
</tr>
<tr>
<td>Kyshtym accident</td>
<td>4.0</td>
<td>–</td>
</tr>
<tr>
<td>Three Mile Island accident</td>
<td>–</td>
<td>0.00055</td>
</tr>
</tbody>
</table>

$^a$ Release in liquid effluents from fuel reprocessing plants.

For estimating the total amounts of radionuclides released in nuclear testing, a total fission yield of 155 Mt is assumed. This was derived from measurements of $^{90}\text{Sr}$ depositing worldwide after injection of debris into the stratosphere. This does not account for some additional amounts of fission yield and the radionuclide amounts that have been produced as tropospheric and local fallout. Examples of the amounts of radionuclides produced in nuclear testing and receiving global distribution are 600 PBq of $^{90}\text{Sr}$, 650 000 PBq of $^{131}\text{I}$ and 910 PBq of $^{137}\text{Cs}$ [1]. These values are included in Table II for comparisons with other sources.

Large explosions in the atmosphere carry most of the radioactive material into the stratosphere, where it remains for some time, the mean residence times being estimated at from less than one year to about five years, depending on the altitude and latitude. Fallout can therefore occur years after an explosion has injected material into the atmosphere. Smaller explosions carry the radioactive material only into the troposphere, and fallout occurs within days or weeks.

A great deal of attention has been paid to monitoring fallout radionuclides in the environment, in foods and in man. From the monitoring records accumulated over tens of years, the relationships of radionuclide levels in the transfer chains from sources to man have been obtained. The time integrated concentrations give a measure of both the concentrations and the periods of residence. Transfer factors are then derived as the ratios of integrated concentrations in successive environmental compartments. Details of the measurement experience and derivations of the transfer factors are to be found in the UNSCEAR Reports [1–4]. These empirical relations
TABLE III. COLLECTIVE EFFECTIVE DOSE PER UNIT AMOUNT OF RADIONUCLIDES RELEASED TO THE ENVIRONMENT

<table>
<thead>
<tr>
<th>Source</th>
<th>Normalized collective effective dose (man·Sv per PBq)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{90}\text{Sr}$</td>
<td>$^{131}\text{I}$</td>
</tr>
<tr>
<td>Atmospheric nuclear testing</td>
<td>700</td>
<td>0.3</td>
</tr>
<tr>
<td>Nuclear power production</td>
<td>12$^a$</td>
<td>500</td>
</tr>
<tr>
<td>Chernobyl accident</td>
<td>—</td>
<td>20</td>
</tr>
<tr>
<td>Windscale accident</td>
<td>—</td>
<td>1000</td>
</tr>
</tbody>
</table>

$^a$ Release in liquid effluents to the ocean.

derived at various locations indicate the usual behaviour and also the extremes of behaviour under specific local conditions.

The radiation doses from fallout radionuclides are due mostly to the ingestion of radionuclides that have become incorporated in foods and to external irradiation from ground deposition. The most significant contributors to dose in decreasing order of importance are $^{14}\text{C}$, $^{137}\text{Cs}$, $^{95}\text{Zr}$, $^{90}\text{Sr}$, $^{106}\text{Ru}$, $^{144}\text{Ce}$ and $^{3}\text{H}$. Only residual irradiation from $^{14}\text{C}$, $^{137}\text{Cs}$, $^{90}\text{Sr}$ and $^{3}\text{H}$ remains to be received by the present and future world population. The collective effective dose commitment is estimated to be $3 \times 10^7$ man·Sv. This is equivalent to 2.4 years of exposure to natural sources of the present world population. Some estimates of the doses per unit amount released for important fallout radionuclides are listed in Table III.

The practice of atmospheric nuclear testing has been replaced by underground nuclear testing, which, because of lower yields and mostly effective containment, results in comparatively low and infrequent exposures of populations. There have been some 1400 underground tests conducted mostly since 1962. At present, a moratorium is being held. UNSCEAR has estimated the total collective effective dose from underground nuclear testing to have been 200 man·Sv.

Some additional exposures have been delivered to local and regional populations by the production of weapons material and the fabrication of the weapons. Some estimates of doses have been made for releases from the Hanford plant in the United States of America and for releases into the Techa River from the Chelyabinsk plant in the former Soviet Union. The releases were less controlled in the early period (prior to 1960). Both the production amounts and the releases have diminished in recent years.
4. NUCLEAR POWER PRODUCTION

During routine operation of nuclear reactors and other installations associated with the nuclear fuel cycle, small amounts of radioactive materials are released in airborne and liquid effluents. Even for all of the reactors of the world over the entire periods of operation, the estimated total releases of the radionuclides $^{90}$Sr, $^{131}$I and $^{137}$Cs are negligible compared to amounts released in nuclear testing. More significant releases occur from fuel reprocessing plants, but, until now, only 4–5% of utilized nuclear fuel has been reprocessed. The estimations of doses from radionuclide releases associated with nuclear power production are made with much reliance on environmental models. Measurements of the concentrations are usually not feasible beyond the immediate vicinity of the discharge points.

From mining and milling operations at the first stage of the nuclear fuel cycle the main radionuclide discharged is $^{222}$Rn. Limited data on radon releases from installations in a few countries are available from which production weighted average values have been calculated. Radon releases from the mill tailings can vary by orders of magnitude, depending on the treatment options exercised. UNSCEAR has estimated that the average exhalation rate of $^{222}$Rn from an uncovered tailings pile would be expected to be 20 Bq·m$^{-2}$·s$^{-1}$, and from a nominally treated abandoned tailings pile a release rate of 3 Bq·m$^{-2}$·s$^{-1}$ is assumed that is maintained over thousands of years because of the long radioactive half-life of the precursor radionuclide $^{230}$Th [1]. UNSCEAR has limited the assessment period to 10,000 years, beyond which substantial erosion or major geological processes would be expected to occur and significantly alter the assumed conditions.

UNSCEAR has assumed that the uranium mines and mills are located in semi-arid, rural locations. The population densities were assumed to be 3 km$^{-2}$ at 0–100 km and 25 km$^{-2}$ at 100–2000 km. An atmospheric dispersion model was used to estimate the collective dose. The derivation of the dose factors is described in the UNSCEAR 1993 Report [1]. The collective doses related to nuclear power production through 1989 were estimated to be 2700 man·Sv from the operating mines and mills and 277,000 man·Sv from the long term release of $^{222}$Rn from abandoned tailings piles [1].

The data on releases of radionuclides from nuclear reactors are particularly comprehensive and complete. There is some dependence of release rates on the specific reactor type and the waste treatment systems, but the variability is great, depending on particular conditions or maintenance operations. UNSCEAR has averaged releases over five-year periods to assess collective doses. The normalized releases (activity amounts per unit electrical energy generated) have decreased with time, reflecting improvements in the quality of nuclear fuel as well as in the performance and standards of reactors in operation. The estimates of normalized releases of radionuclides from reactors have been combined with the electrical energy generated to obtain estimates of the total releases from all reactor operations in the world.
From 1956, when the practice began, until the end of 1989, which was the latest assessment period of UNSCEAR, the total electrical energy generated worldwide was 1844 GW·a. Because of the relatively low release rates, it has been necessary to rely on dispersion models to obtain estimates of the collective doses. For the entire period of reactor operation the collective dose is estimated to be 3700 man·Sv, of which over 50% is due to the long term exposure from $^{14}$C.

Nuclear fuel reprocessing plants are operated in France, Japan and the United Kingdom. The effluents are largely released to the sea, and the doses associated with operations of these installations have been relatively low. Nevertheless, stricter controls on release amounts have been imposed since the early 1980s, and this is reflected in the release trends. The collective dose to local and regional populations from the reprocessing practice is estimated to be 4600 man·Sv. The main components of the dose (over 90%) are $^{137}$Cs and $^{106}$Ru in liquid effluents. A further contribution to collective dose delivered to the global population over 10 000 years is 123 000 man·Sv from $^{14}$C.

The total collective dose from nuclear power production is thus 11 000 man·Sv to local and regional populations from releases at the mines, mills, reactors and fuel reprocessing plants. A further long term component of 400 000 man·Sv is contributed by the continued release of $^{222}$Rn from tailings piles and by the long term residence of $^{14}$C in the global environment.

5. ACCIDENTS

There are a number of ways in which accidents may result in releases of radioactive materials to the environment. Some involve only local contamination and exposures of a few people, such as the Goiânia accident involving a discarded $^{137}$Cs medical source. Other accidents at nuclear installations have been of much greater significance. These include the accidents at Chernobyl, Three Mile Island, Kyshtym and Windscale.

The Chernobyl accident was the most serious that could occur at a reactor. The reactor was destroyed and substantial fractions of the core inventories of radionuclides were released and widely dispersed. The total estimated releases of the principal radionuclides (Table II) were 1670 PBq of $^{131}$I, 85 PBq of $^{137}$Cs, 43 PBq of $^{134}$Cs and 8 PBq of $^{90}$Sr [5].

Many measurements were made following the Chernobyl accident throughout the northern hemisphere, and especially in Europe. These have been useful to rediscover the characteristics of caesium behaviour in the environment or to point out more clearly the dependence of transfer on particular soil characteristics and the enhanced accumulations in, for example, mushrooms, berries, lake fish, lichens, reindeer and game animals.
The methodology for estimating doses from such an accidental release has been improved. Computer codes may now take account of seasonal differences in transfers. The UNSCEAR assessment of exposures of the population of the northern hemisphere took account of measurement results in countries during the first year following the accident and used the fallout experience to make projections of future doses. The doses per unit release (Table III) are for the specific conditions of the accident, i.e. surface release in the early growing season.

The accident at Kyshtym in the southern Urals in the USSR in 1957 was caused by a chemical explosion in a radioactive waste storage tank. The principal radionuclides released were $^{144}$Ce (49 PBq), $^{95}$Zr + $^{95}$Nb (19 PBq), $^{90}$Sr (4.0 PBq) and $^{106}$Ru (2.7 PBq). The release of $^{137}$Cs was much less (0.027 PBq). The measurement results have been particularly informative with regard to the behaviour of $^{90}$Sr [6, 7].

The accident at Windscale, UK, in 1957 involved a fire in the graphite core of a gas cooled reactor. Radionuclides released included 0.74 PBq of $^{131}$I, 0.022 PBq of $^{137}$Cs, 0.003 PBq of $^{106}$Ru, 1.2 PBq of $^{133}$Xe and 0.0088 PBq of $^{210}$Po [8]. The contamination of pastures with $^{131}$I was recognized as being of most concern, and prompt imposition of a ban on milk supplies reduced the intake of $^{131}$I via the pasture-cow-milk pathway. Doses to the thyroid and collective dose have been estimated. Of the total collective dose of 2000 man-Sv, 37% was estimated to be due to $^{131}$I, 37% to $^{210}$Po and 15% to $^{137}$Cs [8].

The accident at the Three Mile Island reactor in 1979 also caused serious destruction of the reactor core, but containment largely prevented releases of radioactive materials to the environment. The estimated release amounts were 370 PBq of noble gases, mainly $^{133}$Xe, and 0.55 TBq of $^{131}$I. The collective dose from the accident was estimated to be 40 man-Sv [9], about half of which was received by residents within 80 km of the plant and the remainder was due to $^{133}$Xe exposures at greater distances.

Several additional accidents have occurred for which the collective doses have been estimated. These include crashes of airplanes carrying nuclear weapons and re-entry of satellites into the atmosphere. The airplane crashes at Palomares, Spain, in 1966, and at Thule, Greenland, in 1968 caused $^{239}$Pu and $^{240}$Pu to become dispersed locally to the environment. The collective doses were minimal in both instances. The satellite re-entries were the SNAP-9A in 1964 and the Cosmos 954 in 1978. Fallout experience provided the dose factors for $^{238}$Pu and the fission products released on burnup in the atmosphere. The collective dose estimates were 2100 man-Sv from the SNAP-9A incident and 16 man-Sv from the Cosmos re-entry [1].

Several industrial and medical sources that were lost or improperly disposed of have caused exposures of members of the public. The collective doses have not been large, but the doses to individuals have sometimes been quite high and even fatal. A summary of dose estimates has been made by UNSCEAR [1].
TABLE IV. LONG TERM COMMITTED DOSES FROM MAN-MADE SOURCES

<table>
<thead>
<tr>
<th>Source</th>
<th>Main radionuclides</th>
<th>Collective effective dose (man • Sv)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atmospheric nuclear testing</td>
<td>$^{14}$C, $^{137}$Cs, $^{90}$Sr, $^{95}$Zr</td>
<td>30 000 000</td>
</tr>
<tr>
<td>Chernobyl accident</td>
<td>$^{137}$Cs, $^{134}$Cs, $^{131}$I</td>
<td>600 000</td>
</tr>
<tr>
<td>Nuclear power production</td>
<td>$^{14}$C, $^{222}$Rn</td>
<td>400 000</td>
</tr>
<tr>
<td>Radioisotope production and use</td>
<td>$^{14}$C</td>
<td>80 000</td>
</tr>
<tr>
<td>Nuclear weapons fabrication</td>
<td>$^{137}$Cs, $^{106}$Ru, $^{95}$Zr</td>
<td>60 000</td>
</tr>
<tr>
<td>Kyshtym accident</td>
<td>$^{144}$Ce, $^{95}$Zr, $^{90}$Sr</td>
<td>2 500</td>
</tr>
<tr>
<td>Satellite re-entries</td>
<td>$^{238}$Pu, $^{239}$Pu, $^{137}$Cs</td>
<td>2 100</td>
</tr>
<tr>
<td>Windscale accident</td>
<td>$^{131}$I, $^{210}$Po, $^{137}$Cs</td>
<td>2 000</td>
</tr>
<tr>
<td>Other accidents</td>
<td>$^{137}$Cs, $^{133}$Xe, $^{60}$Co, $^{192}$Ir</td>
<td>300</td>
</tr>
<tr>
<td>Underground nuclear testing</td>
<td>$^{131}$I</td>
<td>200</td>
</tr>
</tbody>
</table>

6. CONCLUSIONS

Exposures to radiation occur from both natural and man-made sources. The natural radiation background forms the baseline upon which the additional exposures from man-made sources occur. The pathways of transfer of radionuclides through the environment, the points of accumulation and the residence times have been studied in detail. The measurement experience is substantial, particularly for atmospheric releases such as occurred during atmospheric nuclear testing, and following accidents at nuclear installations. The dose estimation procedures have been well developed.

A summary of collective doses from man-made radiation sources is given in Table IV. It is evident that the most significant release of radionuclides to the environment thus far has been from atmospheric nuclear testing. Next in importance was the Chernobyl accident, followed by the long term exposures from $^{14}$C and $^{222}$Rn associated with nuclear power production. A significant portion of the collective dose from atmospheric nuclear testing (86%) is due to the long term exposure from $^{14}$C.

It is the intention of UNSCEAR to refine these various estimates with improved environmental transfer and dose models and to evaluate also annual doses from the various sources. The doses from natural sources cannot be neglected in further assessment in view of the fact that 13 000 000 man-Sv (2.4 mSv × $\left(5.6 \times 10^9\right)$ persons) is delivered each year to the world population from this source.
REFERENCES


GLOBAL RADIOECOLOGICAL IMPACT OF NUCLEAR ACTIVITIES IN THE FORMER SOVIET UNION

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Abstract

GLOBAL RADIOECOLOGICAL IMPACT OF NUCLEAR ACTIVITIES IN THE FORMER SOVIET UNION.

The political changes in the former Soviet Union (FSU) as well as the Chernobyl accident in 1986 have drawn attention to radioactive contamination problems in the FSU. Information that was earlier classified has recently been disclosed. This fact, as well as the rapidly growing scientific co-operation between Russia and industrialized countries, have made it possible to obtain a picture of nuclear activities in the FSU. However, information is still incomplete because of the fact that many of the nuclear activities in the FSU were connected with military activities. The paper describes the main nuclear activities in the FSU which are of global environmental significance. As a measure of the importance of contamination from $^{90}\text{Sr}$ and $^{137}\text{Cs}$, their global environmental inventories together with the doses received by the world population from these radionuclides have been used. To some extent, information on their input has been available from the FSU, but environmental measurements performed outside the FSU have also been applied in the assessment of the inventories and doses. There is a notable discrepancy between the estimates by the United Nations Scientific Committee on the Effects of Atomic Radiation and data from the FSU for global fallout from nuclear weapons testing at Novaya Zemlya. Nuclear weapons testing in the atmosphere at this site is, however, the main source of global radioactive contamination from nuclear activities in the FSU. Next in importance is the Chernobyl accident. Compared with these two sources of radioactive contamination, the impact of discharges of radioactive waste into the marine environment is unimportant.

1. INTRODUCTION

The Chernobyl accident in 1986 has drawn the attention of the world to environmental problems arising from radioactive releases in the former Soviet Union (FSU). The Soviet Union had until then provided very little information about environmental nuclear matters, but the Chernobyl accident gradually changed this situation. It became clear that this accident had not been the only nuclear event with environmental consequences in the FSU. Many of the incidents revealed since then had only been of environmental concern within the borders of the FSU. However,
in a number of cases the radioactive releases resulted in global contamination. This paper focuses on these cases. It discusses not only actual environmental contamination but also potential sources in the FSU which may result in future global contamination.

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) in its reports to the General Assembly has dealt with the global environmental contamination from radioactive releases. The present paper refers mainly to these UNSCEAR reports [1–5]. Another central reference is a recent report of the European Commission on Sources to Environmental Radioactive Contamination in the Former USSR [6].

In the present context, global radioecological impact means environmental contamination, with the two radioecologically most important fission products $^{90}$Sr and $^{137}$Cs measurable outside the borders of the FSU (of the order of 1000 km from the source). The paper considers separately two pathways to global contamination: atmospheric and aquatic pathways.

2. ATMOSPHERIC PATHWAYS

The major part of global contamination from radioactive releases was disseminated as airborne debris. This pathway predominated for nuclear test explosions as well as for nuclear accidents. Thermonuclear test explosions with yields corresponding to millions of tonnes trinitrotoluene (TNT) inject most of the radioactive debris into the stratosphere, and the fallout from such tests becomes globally distributed. Debris from nuclear weapons (kilotonne range) tests and from reactor accidents (e.g. Chernobyl) usually remain below the tropopause and the fallout is deposited mainly in a band around the latitude of the injection.

2.1. Nuclear weapons testing in the atmosphere

The major source of global radioactive contamination was nuclear weapons testing in the atmosphere. This testing occurred from 1945 to 1980. In the FSU, nuclear weapons testing in the atmosphere took place from 1949 to 1962, when the FSU acceded to the moratorium for atmospheric nuclear weapons testing together with the United States of America and the United Kingdom. The major sites for nuclear weapons testing in the FSU were Novaya Zemlya and Semipalatinsk.

2.1.1. Semipalatinsk

On 29 August 1949, the first explosion of a nuclear device in the FSU marked the beginning of nuclear weapons tests at the Semipalatinsk test site in Kazakhstan (50°N, 80°E) (Fig. 1). Other tests at Semipalatinsk followed, including the first
Soviet H-bomb test on 12 August 1953. Over the period 1949–1962, 124 explosions were conducted (8 explosions at high altitude, 91 atmospheric explosions and 25 underground explosions), amounting to 6.4 Mt TNT. The Semipalatinsk test site was the main site for conducting atmospheric nuclear explosions. These explosions resulted in radioactive contamination of the surrounding territories and irradiation of the population of these territories.

The atmospheric tests at Semipalatinsk produced 6.6 PBq $^{137}$Cs and 3.5 PBq $^{90}$Sr; 10–25% of the radioactive products were deposited as 'close-in fallout' at a distance of 100–300 km from the explosion [7]. Hence, about 5.5 PBq $^{137}$Cs and 3.0 PBq $^{90}$Sr were dispersed globally from the Semipalatinsk test site.

2.1.2. Novaya Zemlya

From 1955 until the moratorium on atmospheric nuclear weapons tests in 1962, 87 atmospheric test explosions were carried out at the arctic island Novaya Zemlya (see Fig. 1). The tests were performed on three technological platforms situated in the area of Gulf Chornaya (zone A), around the Matochkin Shar Sound (zone B), and in the area of Gulf Sulmenova (zone C) [8]. The energy release from these atmospheric tests amounted to 235 Mt TNT. The amounts of $^{137}$Cs and $^{90}$Sr produced in these test were 0.155 EBq and 0.1 EBq, respectively [8]. According to
UNSCEAR [9], the estimated total yield of all Soviet atmospheric nuclear tests was 357.5 Mt TNT, and the fission yield was 110.9 Mt TNT, corresponding to 0.43 EBq \(^{90}\text{Sr}\) and 0.65 EBq \(^{137}\text{Cs}\). Since Semipalatinsk and Novaya Zemlya are the only atmospheric test sites of real importance in the FSU, it is evident that the UNSCEAR estimates of \(^{90}\text{Sr}\) and \(^{137}\text{Cs}\) production do not agree with Russian information, which gives production yields of both radionuclides that are four times less than the UNSCEAR estimates. The total yields of nuclear weapons testing also disagree: the UNSCEAR estimates are 1.5 times higher than the Russian data. The UNSCEAR estimates of total \(^{90}\text{Sr}\) and \(^{137}\text{Cs}\) production from nuclear weapons testing are compatible with the worldwide deposition of these radionuclides measured globally. Hence, unless the yields of other nuclear activities have been vastly underestimated, the Russian data seem biased to the low side.

From the UNSCEAR report [9] it appears that about 90% of the activity produced in the FSU nuclear tests went to the stratosphere and thus became globally distributed; this corresponds to 0.4 EBq \(^{90}\text{Sr}\) and 0.6 EBq \(^{137}\text{Cs}\). These figures are used as the best estimate of the global input of fallout from atmospheric nuclear test explosions in the FSU dissipated through the atmospheric pathway.

### 2.2. Underground nuclear explosions

After the agreement on a stop of atmospheric nuclear tests in 1962, neither the FSU nor the USA and the UK have performed nuclear explosions in the atmosphere. However, underground explosions have been carried out for peaceful as well as military purposes. Some of these explosions have vented radioactive substances to the atmosphere.

#### 2.2.1. Peaceful nuclear explosions

In the FSU, 115 nuclear explosions have been conducted for peaceful purposes [10, 11]. About 70% of these explosions have been carried out on the Russian territory. The yields were from several tens of tonnes to 35 kt TNT, and their total yield did not exceed 800 kt TNT. The explosions were conducted during the period 1971–1988, predominantly in the Russian part of the FSU. It is difficult to estimate the extent of radioactive contamination as a result of peaceful nuclear explosions because there is still no sufficient information on the circumstances of the explosions. The short lived radioactive inert gases which are often released by these explosions are transformed by decay to \(^{90}\text{Sr}\) and \(^{137}\text{Cs}\). These radionuclides, together with long lived neutron activation products, e.g. \(^{60}\text{Co}\) and tritium, determine the long term radioactive contamination of the environment from peaceful nuclear explosions. It is believed that these explosions did not lead to any significant contamination outside the borders of the FSU.
2.2.2. Military underground nuclear tests

The underground nuclear tests in the FSU took place at Semipalatinsk and at Novaya Zemlya. From 1963 to 1989, 343 underground explosions (in boreholes and mine adits) were conducted at the Semipalatinsk test site. The total yield of the underground nuclear explosions amounted to approximately 10.1 Mt TNT. In 13 cases, so-called ‘emergency’ situations arose: the radiation situation did not correspond to the predicted one because of venting of some radioactive fumes and gaseous products. As a result of seepage of radioactive inert gases through ground cracks, 1.3 PBq $^{137}$Cs and about 1.4 PBq $^{90}$Sr were formed in the atmosphere [12]. The first underground nuclear explosion was carried out at the Novaya Zemlya test site in 1964.

The explosions conducted at Novaya Zemlya may be summarized as follows [10]: 15 explosions (36%) in which radioactive debris was practically totally contained; 25 explosions (60%) which were accompanied by seepage of radioactive inert gases into the atmosphere without residual contamination; 2 explosions (4%) which were accompanied by venting of radioactive fumes and gases.

The total yield (energy release) as a result of underground nuclear explosions at Novaya Zemlya did not exceed 25 Mt TNT; assuming that this corresponds to a fission yield of 10 Mt TNT, the production of $^{90}$Sr was 37 PBq and that of $^{137}$Cs was 56 PBq.

Although radioactive contamination that could be attributed to Soviet underground nuclear tests was occasionally measured, e.g. in the Nordic countries [13], such contamination was transient. Hence, underground nuclear explosions in the FSU did not contribute significantly to the global fallout.

2.3. The Chernobyl accident

On 26 April 1986, a serious accident happened at the Chernobyl nuclear power plant (Fig. 1). The circumstances of the accident have been described elsewhere [4]. Suffice it to say that the Chernobyl accident was a tropospheric event, since the debris was injected into the lower part of the atmosphere. Table I shows that the countries outside the FSU received more radiocaesium from the accident than the FSU itself. In the case of the other long lived radionuclides, e.g. $^{90}$Sr and the transuranics, most of the activity remained within the borders of the FSU. The Chernobyl accident thus had a global impact, although most of the activity was deposited in northern, south-eastern and central Europe within a distance of about 2000 km from Chernobyl (see Fig. 2).

Compared with fallout from Soviet nuclear weapons tests, the global $^{137}$Cs fallout from the Chernobyl accident was an order of magnitude less (UNSCEAR data) [4, 9]. However, the radioecological impact of the $^{137}$Cs fallout from Chernobyl was 30% of that of the $^{137}$Cs fallout from Soviet nuclear weapons
TABLE I. ESTIMATED RELEASES AND DEPOSITION OF LONG LIVED RADIONUCLIDES FROM THE CHERNOBYL NUCLEAR ACCIDENT

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Total released radioactivity (PBq)</th>
<th>Radioactivity deposited in the European part of the FSU (PBq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs-137</td>
<td>100</td>
<td>30</td>
</tr>
<tr>
<td>Cs-134</td>
<td>50</td>
<td>15</td>
</tr>
<tr>
<td>Sr-90</td>
<td>8</td>
<td>7</td>
</tr>
<tr>
<td>Ru-106</td>
<td>35</td>
<td>25</td>
</tr>
<tr>
<td>Ce-144</td>
<td>90</td>
<td>75</td>
</tr>
<tr>
<td>Ag-110m</td>
<td>1.5</td>
<td>0.5</td>
</tr>
<tr>
<td>Sb-125</td>
<td>3</td>
<td>2</td>
</tr>
<tr>
<td>Pu-239,240</td>
<td>0.055</td>
<td>0.05</td>
</tr>
<tr>
<td>Pu-238</td>
<td>0.025</td>
<td>0.02</td>
</tr>
<tr>
<td>Pu-241</td>
<td>5</td>
<td>4</td>
</tr>
<tr>
<td>Am-241</td>
<td>0.006</td>
<td>0.005</td>
</tr>
<tr>
<td>Cm-242</td>
<td>0.6</td>
<td>0.55</td>
</tr>
<tr>
<td>Cm-243,244</td>
<td>0.006</td>
<td>0.005</td>
</tr>
</tbody>
</table>

* All activities are corrected for decay up to 26 April 1986 and are approximate [14].

The testing [15, 5]. This was because the Chernobyl debris was deposited mainly in densely populated areas with a relatively high agricultural production, whereas the global fallout from nuclear weapons tests was distributed worldwide and a significant amount went to the world's oceans.

2.4. Other atmospheric releases

Although the nuclear test explosions in the atmosphere and the Chernobyl accident were the dominating sources of global fallout from nuclear activities in the FSU, there were other atmospheric releases of concern.

2.4.1. The Kyshtym accident

On 29 September 1957, an accident occurred at Chelyabinsk 40 (Mayak site) (Fig. 1), the so-called Kyshtym accident [16], caused by the explosion of dry nitrate and acetate salts in a tank containing high level radioactive waste. A failure in the
cooling system had resulted in self-heating of the explosive salts in the tank. The explosion dispersed about 74 PBq of nuclear fission products, of which $^{144}$Ce and $^{95}$Zr accounted for 91%. The amount of $^{90}$Sr was 2 PBq, while $^{137}$Cs amounted only to 26 TBq.

Most of the activity was deposited within an area of 300 km x 50 km. Measurements outside the FSU were not able to trace debris from the Kyshtym accident. Nevertheless, some $^{90}$Sr must have been deposited globally from this accident, but the amounts were insignificant compared with depositions from nuclear weapons testing.

2.4.2. Dispersion of radioactivity from Lake Karachay

Since 1951, Lake Karachay was used for disposal of medium level radioactive waste from the Mayak plant. The lake has an area of 0.45 km$^2$ and is situated on the Mayak site, 15 km east of Kyshtym in the Southern Urals. Wastes with an activity of 3.6 EBq $^{137}$Cs and 0.74 EBq $^{90}$Sr were discharged into the lake. Five per cent of the activity was in the aqueous phase [17]; the concentration in this phase was about 0.5 TBq/m$^3$. In 1967, as a result of extreme drought, the lake-side was stripped and an activity of about 22 TBq was dispersed over an area of about 1800 km$^2$ by strong winds. This contamination overlapped the south-eastern part of the radioactive contamination from the above mentioned Kyshtym accident. Any measurable global impact of this wind dispersion is unlikely.
2.4.3. Satellite burnups

The Soviet space programme also comprised the use of nuclear sources. A few accidents have been reported, but the amounts of radioactivity involved were relatively modest.

A Soviet satellite, Cosmos 954, powered by a nuclear reactor, re-entered the atmosphere over the Canadian Northwest Territories in January 1978. Radioactive debris were spread over a 1000 km path [18]. It is estimated that at the launch of the satellite the reactor core contained 3 TBq $^{90}$Sr, 0.2 PBq $^{131}$I and 3 TBq $^{137}$Cs. Seventy-five per cent of the original material is estimated to have remained in the upper atmosphere and thus to have been globally distributed.

Another Cosmos satellite (No. 1402) re-entered the atmosphere in 1982 [19] over the Indian Ocean and the South Atlantic, but no information on radioactive contamination from that event is available.

2.4.4. Miscellaneous releases

Besides the Chernobyl accident in 1986, a number of other (less severe) accidents in RBMK-type reactors have been reported, e.g. at Leningrad in 1975, at Chernobyl in 1982 and at St. Petersburg in 1992 [20]. Radioactivity from the last accident was measured in Finland, but the accidents had no global impact.

Besides the production of nuclear materials by the Mayak enterprise in Chelyabinsk, nuclear materials have been produced at six other sites in the FSU. The two most important ones for production of nuclear weapons plutonium are Tomsk-7, 15 km north-west of Tomsk on the Tom river, which is connected to the Ob river system; and Dodonovo on the shores of the Yenisey river, 50 km north-east of Krasnoyarsk (Fig. 1).

In Tomsk-7, an accident occurred on 6 April 1993 [21]. The facility was heavily damaged. The amount of radioactivity released is estimated at 1.5 TBq. The activity consisted mostly of $\beta\gamma$ emitters, first of all $^{95}$Nb and $^{106}$Ru; of the order of 50 GBq $^{239}$Pu may have been released. About ten days after the accident, radioactivity from Tomsk-7 was detected in the air over northern Sweden [22], but the levels were low (0.3 $\mu$Bq $^{95}$Nb/m$^3$ and 3 $\mu$Bq $^{106}$Ru/m$^3$).

3. AQUATIC PATHWAYS

Although most of the anthropogenic radioactivity in the aquatic environment comes via the atmosphere, radionuclides may also be transferred to water without mediation of the atmosphere. This is the case for liquid discharges into a water recipient, for run-off from land and for leakage from dumped objects.
3.1. Discharges into rivers

Any radioactive discharge into a river which runs to the open sea will in principle be of global concern. However, not all radionuclides will remain in the water phase. Some radionuclides (e.g. radiocaesium and plutonium) are retained by sediments and biota, and the river water becomes depleted of such radionuclides compared to the composition in the discharge.

3.1.1. Techa river

During 1949–1952, medium and low level liquid radioactive waste was discharged directly into the Techa river [23–27], which belongs to the river system Iset–Tobol–Irtysh–Ob. In the upper reaches it passes the area of the nuclear enterprise Mayak. The liquid radioactive waste was released into the open river system, 6 km from the source of the Techa. During that period, \(77 \times 10^6\) m\(^3\) of liquid radioactive waste was discharged, with a total activity of 100 PBq [28]. Most of the activity (~95%) was discharged in 1950 and 1951. The contributions of \(^{90}\)Sr and \(^{137}\)Cs were 11.6% and 12.2%, respectively. As of 1995, the environmental contamination from these discharges had decayed to 4.0 PBq \(^{90}\)Sr and 4.2 PBq \(^{137}\)Cs (Fig. 3).

![Graph showing mean annual \(^{90}\)Sr concentrations in the Techa river water collected at two settlements (Muslyumovo, 78 km from the point of discharge; and Pershinskoye, 213 km from the point of discharge) from 1949 to 1990 [25].](image-url)
In September 1951, the discharge of polluted wastes was discontinued and a cascade of reservoirs and bypass canals was established. This permitted removal of a substantial part of the activity from the open drainage network. However, before this was accomplished, large parts of the flood plain and the bottom of the river, especially upstream, were polluted. According to Nikipelov et al. [17], the major portion of radionuclides (about 99%) was deposited upstream of the village Muslymovo, i.e. in the first 80 km of the river flood land and the river bed. The Techa river was no longer used for economic purposes and the population of several villages was evacuated. At present, the source of permanent contamination of the Techa river is the Asanov swamps in the upper reaches of the river. Moreover, migration of radionuclides from the cascade of reservoirs and canals has been revealed. This system contains 7.1 PBq $^{90}$Sr and $^{137}$Cs [28]. It has been estimated that there is at least 1 PBq ($^{90}$Sr + $^{137}$Cs) outside the Techa river [29]. Some of this activity has undoubtedly entered the Arctic Ocean via the Kara Sea and is thus of global concern. Ongoing Russian–Nordic research is trying to improve this estimate of the global impact from the discharges into the Techa river.

3.1.2. Yenisey river

Information on the contamination of the Yenisey river is given in Ref. [30]: 0.5 km from the point of discharge from the Krasnoyarsk Mining and Chemical Industrial Complex (see Section 2.4.4), the river water contained (in 1991) 970 Bq $^{24}$Na/L, 40 Bq $^{51}$Cr/L, 0.07 Bq $^{60}$Co/L, 0.0015 Bq $^{90}$Sr/L and 0.07 Bq $^{137}$Cs/L. The concentrations of $^{90}$Sr in the Yenisey river are two to three orders of magnitude less than those measured in the Techa river 50 km from the discharge point and thus seem to be of no global concern. No information is presently available on previous discharges into the Yenisey river from the Krasnoyarsk plant.

3.1.3. Tomsk river

The third major production site of nuclear weapons plutonium in the FSU, Tomsk-7, may have discharged liquid radioactive waste into the Tomsk river, which is part of the Ob river system. Information on such discharges is, however, not available in the open literature.

3.2. Run-off

Radioactivity deposited in the terrestrial environment may be transported to groundwater, lakes and rivers, and later to the sea. Run-off may thus be considered as a secondary transfer of atmospheric radioactivity to the sea.
3.2.1. **Global fallout**

The land area of the FSU is \(22.4 \times 10^6 \text{ km}^2\). The major part of the FSU is situated between 40°N and 70°N. The integrated density of global fallout from nuclear weapons testing within this latitude band is 2.7 kBq \(^{90}\text{Sr}/\text{m}^2\) and 4.4 kBq \(^{137}\text{Cs}/\text{m}^2\) \[5\]. Hence the total inventories deposited within the FSU are 60 PBq \(^{90}\text{Sr}\) and 97 PBq \(^{137}\text{Cs}\). About 9% of the \(^{90}\text{Sr}\) inventory or 5 PBq will be removed by run-off \[31\] and thus reach the ocean. The run-off percentage of \(^{137}\text{Cs}\) is about one fifth of that of \(^{90}\text{Sr}\) \[32\], i.e. 1.5 PBq \(^{137}\text{Cs}\) may enter the ocean through run-off from the territory of the FSU.

3.2.2. **Fallout from the Chernobyl accident**

The land area of the FSU received about 30 PBq \(^{137}\text{Cs}\) and 7 PBq \(^{90}\text{Sr}\) from the Chernobyl accident (Table I). If the same run-off percentages as used above for global fallout are applied to the Chernobyl fallout, about 0.5 PBq \(^{137}\text{Cs}\) and 0.6 PBq \(^{90}\text{Sr}\) will enter the ocean through run-off from the FSU.

3.2.3. **Contamination in the Southern Urals**

The accumulated land contamination from the Techa river discharges (due to flooding) \[29\], from the Kyshtym accident \[16\] and from the Karachay wind dispersal \[17\] amounts to approximately 1 PBq \(^{90}\text{Sr}\) and 0.2 PBq \(^{137}\text{Cs}\). The activity in the run-off from the contaminated areas is estimated at 0.1 PBq \(^{90}\text{Sr}\) and 4 TBq \(^{137}\text{Cs}\).

3.2.4. **Other sources**

Some of the peaceful nuclear explosions in the FSU involved the construction of canals for river systems \[33\]. This may have led to run-off to the ocean. It is difficult to quantify this run-off, since adequate information is missing, but the amount is probably low compared with that from the above mentioned sources.

3.3. **Marine releases**

Over the years, the FSU has disposed liquid and solid radioactive waste in the ocean. This is documented in the so-called ‘White Book’ No. 3 \[34\].

3.3.1. **Discharges of liquid waste**

Since 1960, the FSU has discharged liquid radioactive waste into the ocean. The activities are distributed as follows: Barents Sea — 450 TBq, White Sea —
3.7 TBq, Kara Sea — 315 TBq and Far Eastern Seas (Vladivostok) — 456 TBq. The Murmansk Sea shipping lines stopped discharges of liquid radioactive waste in 1984, but the Russian navy continues this practice, although at a lower level than previously. In 1989, liquid waste with an activity of 74 TBq was discharged accidentally from a nuclear submarine at Murmansk. The composition of the liquid waste is not known, but it is unlikely that the contribution of $^{90}$Sr and $^{137}$Cs from this activity should have exceeded 1 PBq.

3.3.2. Dumped solid waste

Solid radioactive waste has been dumped by the FSU since 1964. As a rule, the waste was placed in metal containers before burial in one of the northern seas. Large size radioactive waste was dumped individually or encapsulated in a special vessel before dumping. The total medium and low level solid radioactive waste dumped into the Kara Sea amounts to about 570 TBq. The Barents Sea has received 1.5 TBq and the Far Eastern Seas 248 TBq.

The composition of this liquid waste is unknown. However, for dumping into the Kara Sea the activity was given as "$^{90}$Sr equivalents", which probably means that the total activity of this type of waste does not exceed 0.5 PBq ($^{90}$Sr + $^{137}$Cs). The solid waste has so far not contaminated the marine environment to any appreciable extent.

3.3.3. Dumped naval nuclear reactors

In the bays of Novaya Zemlya and in the Kara Sea, six submarine nuclear reactors with spent nuclear fuel have been disposed. Furthermore, one shield assembly with partially unloaded spent nuclear fuel from the Lenin ice-breaker has been dumped. Before disposal, the reactor compartments were filled with a solidifying mixture based on furfural, which prevents interaction with sea water for several hundred years (up to 500 years). Moreover, in the bays of Novaya Zemlya and the Kara Sea, ten reactors without nuclear fuel have been dumped. The total activity inventory at the time of disposal corresponded to 85 PBq. The content of $^{90}$Sr and $^{137}$Cs in 1995 is probably of the order of 10 PBq [35, 36]. Norwegian-Russian expeditions [37, 38] to the dumping sites in the Kara Sea have shown "that, in 1992, the influence of the dumped radioactive wastes on the general level of radioactive contamination in the Kara Sea was insignificant".

3.3.4. Submarine accidents

The FSU (and now Russia) [34] has 235 nuclear powered vessels and ships, including 228 belonging to the navy of the Russian Ministry of Defence and seven belonging to the Russian Ministry of Transportation (394 nuclear reactors on naval
ships and 13 on ice-breakers — 60% of the world total). Every year, the operation of nuclear submarines and nuclear powered ships produces up to 20,000 m$^3$ of liquid radioactive waste and about 6000 t of solid waste. According to available information [39], four Soviet ‘nuclear’ submarines were lost in the North Atlantic Ocean and two in the Pacific Ocean; however, two of these have probably been raised and one was not nuclear powered but was carrying nuclear weapons. The ‘White Book’ No. 3 [34] mentions the loss of only three nuclear submarines in the world’s oceans and estimates the maximum activity in these wrecks at 24 PBq. The last loss of a nuclear submarine was that of Komsomolets, which sank at a depth of 1700 m at Bear Island in the eastern part of the Norwegian Sea (Fig. 1). The activity content (1995) in the wreck is estimated at 2.8 PBq $^{90}$Sr and 3 PBq $^{137}$Cs, and nuclear warheads may contain 16 TBq $^{239,240}$Pu [38].

4. POTENTIAL SOURCES

In Sections 2 and 3, a number of nuclear installations are mentioned which contain large inventories of long lived radionuclides and which so far have not been of global environmental concern but may be in the future. For example, this is the case for Lake Karachay on the Mayak site (Section 2.4.2) and for the naval nuclear reactors (Section 3.3.3) dumped in the Arctic Ocean, including Komsomolets (Section 3.3.4). However, a number of other nuclear installations might be mentioned in this context; first of all, the nuclear reactors in operation in the FSU, civilian (34 GW(e)) as well as military (Section 3.3.4). The total inventory of $^{90}$Sr and $^{137}$Cs in the installations is of the order of 10 EBq [6] of each radionuclide. The plutonium content is of the order of 0.1 EBq. To these inventories should be added the radioactive waste, mainly spent fuel elements produced in civilian and military nuclear reactors. Some of this waste is stored at power plants and at military naval bases (e.g. Murmansk and Vladivostok), but the principal final storage places for high level radioactive waste in the FSU are Mayak and Krasnoyarsk. The total activity of radioactive waste stored at Mayak has been reported to be 38 EBq [40].

Another potential source of radioactive contamination is the various nuclear weapons, especially when they are transported by aircraft or ships. The exact stockpile of nuclear weapons in the FSU and thus the plutonium inventory is not known. From estimates of the production of weapons grade plutonium [41] the inventory of nuclear weapons may be about 100–150 t Pu (~200–300 PBq). At remote locations, e.g. along the Siberian coastline, lighthouses powered by $^{90}$Sr generators are used. Each generator contains of the order of 10 PBq $^{90}$Sr and is thus a potential source of environmental contamination. The sources are probably well contained. This was demonstrated by a helicopter crash off the east coast of Sakhalin Island in 1987, which did not result in any environmental contamination although the helicopter carried a 13 PBq $^{90}$Sr source [42].
5. DOSE ASSESSMENT

As stated in the Introduction, the principal radionuclides in this study are $^{90}$Sr and $^{137}$Cs. Consequently, a dose assessment of the global impact of nuclear activities in the FSU has been carried out for these two radionuclides. This assessment is not complete regarding the total global radioecological impact of FSU nuclear activities, since, for example, $^{14}$C has been omitted. On the other hand, if only the dose received by the present world population is considered, $^{90}$Sr and $^{137}$Cs will be the principal contributors to the total dose.

### TABLE II. GLOBAL INVENTORIES OF RADIONUCLIDES FROM NUCLEAR ACTIVITIES IN THE FSU$^a$

<table>
<thead>
<tr>
<th>Activity (PBq)</th>
<th>$^{90}$Sr</th>
<th>$^{137}$Cs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Semipalatinsk atmospheric test</td>
<td>3.0</td>
<td>5.5</td>
</tr>
<tr>
<td>Novaya Zemlya atmospheric test</td>
<td>400</td>
<td>600</td>
</tr>
<tr>
<td>Underground tests and peaceful nuclear explosions</td>
<td>(0.1)</td>
<td>(0.2)</td>
</tr>
<tr>
<td>Chernobyl accident</td>
<td>1</td>
<td>70</td>
</tr>
<tr>
<td>Kyshtym accident</td>
<td>(0.01)</td>
<td>(0)</td>
</tr>
<tr>
<td>Karachay accident</td>
<td>(0)</td>
<td>(0)</td>
</tr>
<tr>
<td>Satellite failures</td>
<td>0.003</td>
<td>0.003</td>
</tr>
<tr>
<td>Tomsk-7</td>
<td>0</td>
<td>(0)</td>
</tr>
<tr>
<td><strong>Total (atmospheric pathway)</strong></td>
<td>400</td>
<td>700</td>
</tr>
<tr>
<td>Techa river discharges</td>
<td>(1)</td>
<td>(0.2)</td>
</tr>
<tr>
<td>Tomsk and Krasnoyarsk discharges</td>
<td>~0?</td>
<td>~0?</td>
</tr>
<tr>
<td>Global fallout run-off</td>
<td>5</td>
<td>1.5</td>
</tr>
<tr>
<td>Chernobyl run-off</td>
<td>0.6</td>
<td>0.5</td>
</tr>
<tr>
<td>Run-off from South-Urals events</td>
<td>0.1</td>
<td>0.004</td>
</tr>
<tr>
<td>Discharges of liquid waste into the sea</td>
<td>(0.05)</td>
<td>(0.06)</td>
</tr>
<tr>
<td>Dumping activities</td>
<td>~0</td>
<td>~0</td>
</tr>
<tr>
<td><strong>Total (marine pathway)</strong></td>
<td>7</td>
<td>2</td>
</tr>
</tbody>
</table>

$^a$ Not corrected for decay. Values in brackets are uncertain estimates.
### TABLE III. GLOBAL DOSE ASSESSMENT FOR NUCLEAR ACTIVITIES IN THE FSU

#### TERRESTRIAL PATHWAYS

<table>
<thead>
<tr>
<th></th>
<th>$^{90}$Sr ingestion</th>
<th>$^{137}$Cs ingestion</th>
<th>$^{137}$Cs external</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Environmental dose factor (man·Sv/PBq)</td>
<td>Dose (man·Sv)</td>
<td>Environmental dose factor (man·Sv/PBq)</td>
</tr>
<tr>
<td>Semipalatinsk atmospheric tests</td>
<td>$6.7 \times 10^2$</td>
<td>$2 \times 10^3$</td>
<td>$7.4 \times 10^2$</td>
</tr>
<tr>
<td>Novaya Zemlya atmospheric tests</td>
<td>$6.7 \times 10^2$</td>
<td>$3 \times 10^5$</td>
<td>$7.4 \times 10^2$</td>
</tr>
<tr>
<td>Underground explosions</td>
<td>$6.7 \times 10^2$</td>
<td>$7 \times 10^1$</td>
<td>$7.4 \times 10^2$</td>
</tr>
<tr>
<td>Chernobyl accident</td>
<td>$6.7 \times 10^2$</td>
<td>$7 \times 10^2$</td>
<td>$2.2 \times 10^3$</td>
</tr>
<tr>
<td>Kyshtym accident</td>
<td>$6.7 \times 10^2$</td>
<td>$7 \times 10^9$</td>
<td>$7.4 \times 10^2$</td>
</tr>
<tr>
<td>Satellite failures</td>
<td>$6.7 \times 10^2$</td>
<td>$2 \times 10^0$</td>
<td>$7.4 \times 10^2$</td>
</tr>
<tr>
<td><strong>Total (terrestrial)</strong></td>
<td></td>
<td>$3 \times 10^5$</td>
<td></td>
</tr>
</tbody>
</table>
### MARINE PATHWAYS

<table>
<thead>
<tr>
<th>Discharge into the Techa river</th>
<th>$^{90}$Sr ingestion</th>
<th>$^{137}$Cs ingestion</th>
<th>$^{137}$Cs external</th>
</tr>
</thead>
<tbody>
<tr>
<td>Environmental dose factor</td>
<td>Environmental dose factor</td>
<td>Environmental dose factor</td>
<td></td>
</tr>
<tr>
<td>(man • Sv/PBq)</td>
<td>(man • Sv)</td>
<td>(man • Sv)</td>
<td>(man • Sv)</td>
</tr>
<tr>
<td>$2 \times 10^0$</td>
<td>$2 \times 10^0$</td>
<td>$5 \times 10^1$</td>
<td>$1 \times 10^1$</td>
</tr>
<tr>
<td>Run-off of global fallout</td>
<td>$2 \times 10^0$</td>
<td>$1 \times 10^1$</td>
<td>$5 \times 10^4$</td>
</tr>
<tr>
<td>Run-off of Chernobyl contamina</td>
<td>$2 \times 10^0$</td>
<td>$1 \times 10^0$</td>
<td>$5 \times 10^1$</td>
</tr>
<tr>
<td>Run-off of South-Urals contamina</td>
<td>$2 \times 10^0$</td>
<td>$2 \times 10^{-1}$</td>
<td>$5 \times 10^1$</td>
</tr>
<tr>
<td>Discharge of liquid waste into the sea</td>
<td>$2 \times 10^0$</td>
<td>$1 \times 10^{-1}$</td>
<td>$5 \times 10^1$</td>
</tr>
<tr>
<td>Subtotal</td>
<td>$1 \times 10^1$</td>
<td></td>
<td>$1 \times 10^2$</td>
</tr>
<tr>
<td>Atmospheric input into the sea</td>
<td>$1 \times 10^0$</td>
<td>$4 \times 10^2$</td>
<td>$2 \times 10^1$</td>
</tr>
<tr>
<td>Total (marine)</td>
<td>$4 \times 10^2$</td>
<td></td>
<td>$1 \times 10^4$</td>
</tr>
<tr>
<td>Grand total</td>
<td>$3 \times 10^3$</td>
<td>$6 \times 10^5$</td>
<td>$1 \times 10^5$</td>
</tr>
</tbody>
</table>

**Note:** The environmental dose factor is the collective dose received from an environmental input of 1 PBq. The dose factors for the terrestrial pathways were calculated from UNSCEAR data [5] and the marine dose factors are based upon information obtained through the IAEA MARDOS project [43]. The dose factors for atmospheric input into the sea are half of those for liquid discharges because the sea only receives about half of the atmospheric input and the rest is deposited on land.
Table II summarizes the global accumulated inventories (not corrected for decay) of $^{90}\text{Sr}$ and $^{137}\text{Cs}$ from the various nuclear activities in the FSU. The total atmospheric discharge of $^{90}\text{Sr}$ and $^{137}\text{Cs}$ is of the order of 1 EBq and the discharge into the ocean is of the order of 0.01 EBq; $^{137}\text{Cs}$ dominates the atmospheric releases and $^{90}\text{Sr}$ the aquatic releases.

From Table III it appears that the dose from ($^{90}\text{Sr} + ^{137}\text{Cs}$), from terrestrial pathways to the world population is about $2 \times 10^6$ man·Sv, whereas the dose from marine pathways is only $10^4$ man·Sv; if liquid discharges alone are considered, the dose is only $10^2$ man·Sv.

Table III shows that in the case of $^{90}\text{Sr}$ produced by nuclear activities in the FSU, the total global dose ($3 \times 10^5$ man·Sv) was due mainly to atmospheric tests at Novaya Zemlya. For $^{137}\text{Cs}$, 70% of the dose was due to tests at Novaya Zemlya and 30% to the Chernobyl accident.

If the annual individual dose received from natural background is 2.4 mSv [5] (including radon), the collective dose from this source to the world population is $2.4 \times 10^{-3} \times 5 \times 10^9$ man·Sv = $1.2 \times 10^7$ man·Sv. This figure is six times the collective ($^{90}\text{Sr} + ^{137}\text{Cs}$) dose from nuclear activities in the FSU, which thus corresponds to 2 months of extra background radiation of the world population.

6. CONCLUSION

The most important nuclear activity in the FSU with regard to global radioactive contamination was the atmospheric testing of nuclear weapons at Novaya Zemlya. There is, however, a notable discrepancy between UNSCEAR’s estimates and Russian data for the amounts of fission products produced in these tests.

Second in importance is the Chernobyl accident. In particular, the relative radioecological impact, i.e. the environmental radionuclide concentrations compared with the released activity, was high for Chernobyl compared with that of nuclear weapons fallout. All other nuclear sources in the FSU seem to have had nearly no global radioecological impact. However, information is still incomplete, e.g. on nuclear discharges to the great Siberian rivers running to the Arctic Ocean. Furthermore, inappropriate management (e.g. reactor operation) of the many potential nuclear sources of environmental radioactive contamination in the FSU may have a global impact in the future.

REFERENCES


[38] JOINT RUSSIAN-NORWEGIAN EXPERT GROUP, Radioactive Contamination at Dumping Sites for Nuclear Wastes in the Kara Sea. Results from the 1993 Expedition, Norwegian Radiation Protection Authority, Østerås (1994).


[42] NAOAKI, U., Russia won’t stop sea dumping but supplies details to Tokio, Nucleonics Week (20 May 1993) 17–18.

OVERVIEWS

New roles for environmental monitoring

(Session 1b)

Chairman

W. WHICKER

United States of America
APPLICATION OF ENVIRONMENTAL MONITORING TO IAEA SAFEGUARDS

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Abstract

APPLICATION OF ENVIRONMENTAL MONITORING TO IAEA SAFEGUARDS.

The IAEA, in co-operation with its Member States, has undertaken a series of field trials as part of its Programme 93+2 to evaluate environmental monitoring as a potential measure for the strengthening of safeguards. Emphasis in these sample collections, carried out in the vicinity of nuclear facilities in 11 countries, was on short range monitoring. Samples collected included hydrological samples (high volume water filters, grab water, biota, sediments), vegetation, soil and surface swipes. The sampling methods applied have proven to be effective in preventing cross-contamination of the samples. In addition, the analytical techniques available in Member State laboratories and within the IAEA have demonstrated their capability to carry out extremely low level radiochemical and isotopic measurements. From the results it can be concluded that environmental monitoring techniques can provide an effective tool for the detection of undeclared activities at declared sites. In particular, on-site swipe sampling, combined with particle analysis, is a powerful tool for providing unambiguous information about the full range of past and current nuclear activities. Signatures in terrestrial and hydrological samples can be seen at distances of up to some 10-20 km away from the facilities.

1. INTRODUCTION

In April 1993, the Standing Advisory Group on Safeguards Implementation (SAGSI) reported its recommendations for improving the cost effectiveness of safeguards to the Director General, who reported these recommendations to the Board of Governors in June 1993. The Board requested the Director General to submit concrete proposals for the assessment, development and testing of the measures proposed by SAGSI. At the December 1993 Board, the Secretariat’s development programme for a strengthened and more cost effective safeguards system (called Programme 93+2) was introduced; this programme provides for the evaluation of the technical, legal and financial implications of SAGSI’s recommendations.
One of SAGSI’s principal recommendations was to assess the effectiveness and costs of environmental monitoring for safeguards to detect undeclared activities at declared sites and undeclared activities at sites not declared. The corresponding activities are carried out under Task 3 of Programme 93+2 and are directed towards the following objectives:

- Identification and documentation of environmental signatures for uranium processing, enrichment, reactor and reprocessing operations;
- Development and documentation of sampling, data storage and analysis procedures;
- Extension of the Network of Analytical Laboratories for the analysis of environmental samples;
- Establishment of a clean room capability for the handling, screening and distribution of such samples at the IAEA Laboratories in Seibersdorf;
- Conduct of demonstration field trials, including quantification of cost factors.

Since September 1993, environmental sampling field trials have been carried out in Argentina, Australia, Hungary, Indonesia, Japan, Netherlands, the Republic of Korea, South Africa, Sweden, the United Kingdom and the United States of America. The emphasis in these field trials was on short range monitoring, in that all planned sample collections were in the vicinity of nuclear facilities.

In this paper, summaries of the sampling techniques and the associated analytical techniques, as well as overall conclusions drawn from the results of the field trials are given.

2. SAMPLING STRATEGIES

Any facility for the processing of nuclear material is expected to release small, but detectable, amounts of radioactive and non-radioactive isotopes to the immediate environment of the facility. These releases can be in the form of gases, particulates or aerosols, as well as solid or liquid waste streams which migrate through the environment and may be found at some distance downwind or downstream of the point of release. Modern analytical techniques have been developed which can measure extremely small amounts of these elemental or isotopic ‘signatures’, thus providing information about the process which created them. This is why environmental monitoring is seen as having promise for the detection of undeclared nuclear activities in States under IAEA safeguards.

The overall strategy of the field trials carried out in 1993–1994 was to study the sampling parameters (methods and media) and the associated analytical measurement techniques which could be used to detect the presence and nature of nuclear activities, such as reactor operation, uranium enrichment, spent fuel reprocessing, radioisotope production and fuel fabrication, at short range (within 10 km). An
essential part of this evaluation was the use of sampling techniques inside process
buildings and within the boundaries of nuclear sites in order to characterize facility
operations, historical and current.

Sampling plans were prepared in consultation with the local experts, taking
into account wind patterns around the sites, local topography, hydrography of the
region, effluent points (smoke-stacks, waste water discharge pipes, settling ponds),
site maps, floor plans of process buildings, as well as existing environmental
monitoring programmes and parameters of nuclear materials and processes present.

Individual field trials were carried out under conditions imposed by the host
organization and under the restraints of limited time and manpower. Therefore,
several field trials, in which only a few types of sampling were carried out, were
of a limited nature. Others were broader in nature and involved a combination of
terrestrial sampling (vegetation and soil), hydrological sampling (water, sediment
and biota) and swipe sampling inside and outside process buildings. The general
strategy for such sampling was to take terrestrial and hydrological samples first,
starting at the most remote locations, to establish background levels of the signatures
to be measured. Swipe sampling inside process buildings was done last, to minimize
the possibility of cross-contamination of the low level environmental samples by the
samplers.

3. SAMPLING TECHNIQUES

Sampling equipment, sample containers and sampling protocols were devel-
oped in collaboration with experts from the Member States. Member State labora-
tories provided certified clean sample bottles for grab water and biota sampling, high
volume water filters and pump systems, sediment tubes and swipe sampling kits. The
IAEA produced vegetation and soil sampling kits from commercially available
plastic bags. The individual techniques and protocols were designed in such a way
as to minimize the risk of cross-contamination.

3.1. Vegetation sampling

In general, vegetation was chosen for its ability to retain the signatures of
interest. Important considerations were long life (mosses and lichen), high surface
area for collection of wet and dry deposition (mosses and lichen), sticky surface for
collection of dry deposition (pine needles) and a growth pattern which accumulates
dry deposition (aloe, monkey tail). These attributes were chosen to maximize the
collection efficiency of the vegetation over a long time period (several years), rather
than to detect recent events. In cases where the most desirable types of vegetation
could not be found, grass was used because of its widespread availability. In one field
trial, it was possible to collect sheep faeces, which are known to concentrate radio-
nuclides such as $^{137}\text{Cs}$. As much as possible, vegetation from exposed areas was chosen, taking into account the prevailing wind direction from the site of origin of the effluents. Each vegetation sample was taken from the same approximate area and location. Samples were collected with gloved hands and placed into plastic mini-grip bags.

3.2. Soil sampling

Soil was taken from exposed areas where run-off may have collected, in order to maximize the collection of dry deposition. A trowel was used to loosen the top 1 cm of soil and place it in a mini-grip bag. The typical sample size was 500 g. Plastic or rubber gloves were worn during sampling and the individual subsamples were double bagged for shipment.

3.3. Water sampling

High volume water samples were pumped through a special filter cartridge incorporating a paper filter for particulates and a mixed ion exchange bed for collection of dissolved ions from solution. The pump unit was portable, operated on a 12 V battery, and included a flow meter to measure the total volume of water pumped through the filter cartridge. The normal volume sampled was 300 L. Replicate cartridges were collected in each location and double bagged for shipment. Figure 1 shows a pump unit operated on a river shore.

Grab water samples were collected in 500 mL acid-washed polyethylene bottles following one to two rinsings. The screw tops of the bottles were then taped and the bottles were double bagged. Plastic or rubber gloves were worn at all times.

3.4. Sediment sampling

Sediment was sampled by pushing a 45 mm diameter Lucite tube into the sediment to a depth of at least 10 cm. The tube was then removed, sealed with plastic end caps, taped and double bagged. The end of the tube corresponding to the top of the sediment was marked. Plastic or rubber gloves were worn at all times.

3.5. Biota sampling

Water plants, algae, seaweed, mussels, etc. were collected with gloved hands and placed in polyethylene bottles which were taped and double bagged. Approximately 100–200 g of material was collected. To the extent possible, these materials were stored in a frozen condition to avoid decomposition.
3.6. Swipe sampling

Swipe samples were taken following a special protocol designed to prevent cross-contamination, especially by the sampler. A two-person team was employed, with one person taking the samples and an assistant handling the clean sampling materials and double bagging the used swipes. The swipe materials utilized were prepared and certified by a Member State measurement laboratory to be free from significant amounts of U or Pu. Paper filters and cloth swipe media were employed.

Each swipe subsample was taken from the same approximate surface area and location, although it was not possible to ensure complete equivalence between subsamples. Areas sampled were between 100 and 500 cm² and were from locations such as door or window frames where environmental dust would collect. Sampling locations in process areas were chosen to give strong signatures of the radioisotopes present; these locations included the access doors to hot cells, UF₆ sampling points and valves in enrichment facilities, and waste handling equipment.

4. SAMPLE ANALYSIS

A number of laboratories in Member States participated in the analysis of environmental samples from the field trials. These laboratories were located in
Australia, Canada, the European Union, Finland, Hungary, the Russian Federation, the UK and the USA. In addition, the IAEA utilized its full analytical capabilities represented by the Marine Environment Laboratory in Monaco, the Isotope Hydrology Laboratory in Vienna, and the Safeguards Analytical Laboratory (SAL) and the Physics, Chemistry and Instrumentation Laboratory in Seibersdorf.

A protocol was established to code the samples so that the Member State analytical laboratories would not know the country or facility of origin. Preliminary screening of the swipe samples from inside process buildings was carried out with gamma spectrometry at the SAL to determine the level of radioactivity for shipment purposes.

A variety of analytical techniques was used in the analysis of the field trial samples. Short summaries of the main techniques and their expected performance are given below.

**Gamma spectrometry** was applied for samples of vegetation, soil, water filters, sediment, grab water, biota and swipes. These could be measured directly as a preliminary screening or reduced in volume by ashing or evaporation for a more accurate measurement. Isotopes of interest included activation products ($^{60}$Co), fission products ($^{134}$Cs, $^{137}$Cs, $^{106}$Ru) and actinides ($^{235}$U, $^{239}$Pu, $^{241}$Am). Detection limits for most isotopes of interest were less than 1 Bq/L in grab water samples or 1 Bq/kg (dry weight) in solid samples.

**Alpha spectrometry**, using semiconductor detectors, was used to measure isotope ratios of U and Pu, following chemical treatment of the sample to extract and purify the element of interest and to produce a thin alpha source. Gross alpha counting methods were used primarily to screen for the presence of actinide elements (U, Pu, Th). For measurement of the $^{234}$U/$^{238}$U ratio, approximately 1 µg U was required.

**Beta spectrometry** was carried out by liquid scintillation counting ($^{90}$Sr) or gas proportional counting ($^{3}$H) following chemical purification steps. Detection limits for $^{90}$Sr in vegetation samples were less than 0.1 Bq/kg and detection limits for $^{3}$H in grab water samples were less than 1 Bq/L.

**Delayed neutron counting** was used to screen for the presence of fissile isotopes, such as $^{235}$U or $^{239}$Pu, at nanogram levels in swipes, biota and vegetation samples. The samples were irradiated in a reactor for a short period, then removed, and the delayed fission neutrons were measured.

**Thermal ionization mass spectrometry** is the most accurate technique for the measurement of U and Pu isotopic composition. Sample preparation for vegetation, water filters, biota and swipe samples requires ashing, dissolution and chemical separation of the elements of interest by ion exchange or chromatographic methods. Soil and sediment samples may be totally dissolved with strong mineral acids or leached to remove the anthropogenic components in preference to the natural back-
ground (especially true for U). Typically, a spike isotope (\(^{233}\)U or \(^{244}\)Pu) is added to the sample before ashing or leaching to aid in sample recovery and quantification of the elemental and isotopic composition. Detection limits for U and Pu are approximately nanograms per litre or kilogram.

*Accelerator mass spectrometry* is the most sensitive technique for the measurement of rare radioisotopes which have long half-lives, such as \(^{36}\)Cl and \(^{129}\)I. Samples of vegetation, sheep faeces, biota and swipes were ashed and chemically treated to isolate the element of interest. Typical sample sizes are less than 10 g, and the detection limits for \(^{36}\)Cl and \(^{129}\)I are in the range of \(10^{-12}\) atoms of the rare isotope per atom of the natural isotope.

*Particle analysis* relies on the detection and analysis of individual particles in the size range 0.1–10 \(\mu\)m in diameter. The particles are removed from the substrate (swipe or vegetation) by ashing or ultrasonic vibration and placed on a clean substrate. They can then be measured by scanning electron microscopy, using an electron probe technique to detect the presence of heavy elements such as U or Pu with femtogram \((10^{-15}\) g) sensitivity. Ion microprobe mass spectrometry can also be employed to measure the isotopic composition of the particles. The fission track technique can be used to identify particles containing fissile isotopes (\(^{235}\)U, \(^{239}\)Pu), which may then be selected for thermal ionization mass spectrometric measurement of the isotopic composition with high precision.

Swipe sampling combined with particle analysis can reveal signatures of past and present activities in locations where nuclear material is handled. Figure 2 demonstrates the power of the technique. It displays the \(^{235}\)U versus \(^{234}\)U isotope abundances measured on swipe samples taken in the uranium laboratory of the SAL. The whole range of isotopics from depleted U to highly enriched U is seen. That this reflects the actual materials handled in this laboratory is shown by superimposing the isotope abundances of the samples analysed during 1994. The largest population of results is in the depleted to low enriched uranium fuel cycle. The important finding is that the picture displayed by the swipe results is practically identical with the actual work carried out. Results from the field trials were similar in that they revealed signatures of present and past activities, consistent with the declared activities for the facilities and for the locations sampled.

5. SUMMARY OF RESULTS

Detailed results of the individual field trials cannot be given in this paper, as this would require prior agreement of the States concerned. However, the results obtained can be summarized as follows:

- Power reactor operation in coastal areas could be detected by the presence of activation or fission products in the liquid wastes discharged at, and to a
distance of up to 20 km away from the discharge point. Power reactors along a river system showed different and less explicit signatures; therefore, this subject may need further study.

Research facilities release a variety of signature isotopes and elements to their immediate environment. Signatures such as fission products or $^3$H may be seen several kilometres from the site. Fuel fabrication facilities, using enriched U, provide an unambiguous signature in the environment up to several kilometres away.

Certain types of enrichment facilities can be detected at distances of up to a few kilometres, depending on the plant operating history, enrichment levels and throughput. Swipe sampling in common areas (e.g. change rooms or office areas) gives a nearly complete picture of the enrichment levels achieved in the facility.

The signature of a large reprocessing facility was detected at a distance of tens of kilometres by the measurement of the volatile fission product $^{129}$I. Swipe samples taken outside the main process buildings of the same facility gave a clear picture of the enrichment and burnup levels of materials under process. However, in a limited sampling campaign, using only high volume water filters, offshore of another large reprocessing facility, no signatures were found above background levels.
— The costs associated with sample taking, screening and distribution are small compared to the analytical costs, especially for the most powerful and sensitive techniques. Optimization of the system through screening methods in the IAEA Clean Laboratory should reduce the overall costs to the IAEA.

6. CONCLUSIONS

From these field trials, which were directed towards short range monitoring with sample collections generally up to 8-10 km away from the nuclear facilities, the following can be concluded:

— Environmental monitoring techniques can provide an effective tool for the detection of undeclared activities at declared sites.
— In particular, on-site swipe sampling, combined with particle analysis, is a powerful technique for providing unambiguous information about the full range of past and current nuclear activities.
— Signatures in terrestrial and hydrological samples can be seen at distances of up to some 10-20 km away from the facilities.

The results obtained in these field trials were entirely consistent with the activities and materials declared by the facility operators.
DETECTION OF ENVIRONMENTAL SIGNATURES FROM NUCLEAR FACILITIES

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Abstract
DETECTION OF ENVIRONMENTAL SIGNATURES FROM NUCLEAR FACILITIES.

Nuclear operations invariably result in the release of trace quantities of radionuclides into the environment. Undeclared activities include reactor operation, enrichment and reprocessing, and the detection of such activities is possible if suitable environmental samples are collected. The choice of environmental sample includes not only the sample type but also the sample location and should be optimized in order to maximize the signal to background ratio. Detectable releases from enrichment facilities are mostly associated with uranium. This might be detected either from enhanced background levels or in the form of enriched or depleted uranium. There is, however, a much larger range of materials which may be released from reprocessing activities and reactor operations. These include both gaseous and particulate species. The choice of radionuclide depends on the likely magnitude of the release, the ease of suitable sampling and the likely period since the release. Whilst it is inevitable that radionuclides are released during nuclear operations, the release is likely to be episodic, originating either at a fixed point in the process or from an accidental leakage. Techniques for detection of environmental signatures may involve air or rain sampling, the collection of soil and vegetation or sampling in the aquatic environment (rivers, lakes and seas). Each sample type has the potential for indicating the presence of nuclear activities, although the choice of sample is strongly dependent on the nature of the release, the surrounding environment (including topography and climatic/vegetation type) and the period since the release. Ultimately, detection will be limited by cost, although the chances of detection can be maximized by the adoption of an appropriate strategy for sample collection. The suitability of certain environmental media for collection is evaluated in the paper, along with observations which can be used in the design of a sampling programme.

1. INTRODUCTION

Nuclear operations invariably result in the release of at least trace quantities of radionuclides into the environment. Such releases might result from controlled discharges or from accidental leakages, following a failure of plant components or a defect in operating procedures. Most previous interest in the effects of radioactive discharges has involved an evaluation of the consequential dose, in order to provide
a risk assessment. However, releases from nuclear processes can provide a signature for the type of activity that is being undertaken. Such a signature can be useful in the identification of undeclared activities at either declared or undeclared sites. Environmental sampling and analysis, therefore, provide a potentially useful tool in the detection of these undeclared operations.

Activities which might be undeclared include uranium processing and enrichment, reactor operation and fuel reprocessing. Each of them is likely to result in the occurrence of a particular environmental signature and this is discussed in the following text. Identification of the signature in environmental media is likely to depend on the nature of the release and the time elapsed between release and sample collection. A strategy can therefore be developed to maximize the chances of detection. It is important to note that such sample collection and analysis are not intended to provide a representation of environmental levels, as in the case of dose assessment, but are intended to maximize the signal to background ratio.

Uranium processing and enrichment are most likely to be detected by measurement of released uranium. This might be detected either from enhanced background levels or, more probably, from displaced uranium ratios. The isotope ratios $^{235}\text{U}:^{238}\text{U}$ and $^{234}\text{U}:^{238}\text{U}$ can provide evidence of enrichment. The ratio $^{235}\text{U}:^{238}\text{U}$ is almost invariant in natural materials (the only exception being the Oklo uranite deposit, where $^{235}\text{U}$ deficiency is ascribed to a natural chain reaction) and any deviation from a ratio of 1 to 137.5 is indicative of enrichment activities. The $^{234}\text{U}:^{238}\text{U}$ ratio does, however, vary in nature.

Reactor operations can result in the release of a number of activation and fission products, as well as possibly actinides. It is difficult to identify precise signatures, since these depend on the reactor type and construction, and analysis for a range of gamma emitters might be useful. One difficulty with reactor operation is that excess heat has to be dissipated and this might be easier to identify than detecting an operation by systematic collection of environmental samples. Sampling from liquid discharge points is probably the most obvious way of detecting such operations.

Reprocessing activities can result in the release of a wide range of actinides and activation and fission products to the environment. A potential difficulty in detection is that there is a background level of radionuclides which results from nuclear weapons fallout and material from the Chernobyl accident or from natural radioactivity. However, there are certain criteria which might facilitate detection, including a low background concentration. An intermediate radioactive half-life may be desirable, since this will be sufficiently short to enable decay of nuclear weapons and Chernobyl fallout background whilst being sufficiently long that fuel storage would not allow substantial decay before the reprocessing operation and subsequent release. A list of likely release levels (assuming reprocessing of 8 kg of plutonium) and radioactive half-lives of the most important radionuclides is given in Table I. It must be noted, however, that technologies for restricting these releases
TABLE I. SOURCE TERM BASED ON THE REPROCESSING OF 8 kg OF PLUTONIUM

<table>
<thead>
<tr>
<th></th>
<th>Release level (Bq)</th>
<th>Half-life (days (d) or years (a))</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>As liquid effluent</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>H-3</td>
<td>$3 \times 10^{12}$</td>
<td>12.3 a</td>
</tr>
<tr>
<td>Sr-90</td>
<td>$1 \times 10^{10}$</td>
<td>29 a</td>
</tr>
<tr>
<td>Zr-95</td>
<td>$8 \times 10^{9}$</td>
<td>66 d</td>
</tr>
<tr>
<td>Nb-95</td>
<td>$1 \times 10^{10}$</td>
<td>35 d</td>
</tr>
<tr>
<td>Tc-99</td>
<td>$8 \times 10^{9}$</td>
<td>$2.14 \times 10^{5}$ a</td>
</tr>
<tr>
<td>Ru-106</td>
<td>$3 \times 10^{10}$</td>
<td>1.0 a</td>
</tr>
<tr>
<td>Ag-110m</td>
<td>$3 \times 10^{8}$</td>
<td>253 d</td>
</tr>
<tr>
<td>Sb-125</td>
<td>$3 \times 10^{10}$</td>
<td>2.7 a</td>
</tr>
<tr>
<td>I-129</td>
<td>$3 \times 10^{8}$</td>
<td>$1.57 \times 10^{7}$ a</td>
</tr>
<tr>
<td>Cs-134</td>
<td>$1 \times 10^{9}$</td>
<td>2.1 a</td>
</tr>
<tr>
<td>Cs-137</td>
<td>$3 \times 10^{10}$</td>
<td>30.1 a</td>
</tr>
<tr>
<td>Ce-144</td>
<td>$3 \times 10^{9}$</td>
<td>284 d</td>
</tr>
<tr>
<td>Pu-238,239,240 : Pu(α)</td>
<td>$1 \times 10^{9}$</td>
<td>$86/2.44 \times 10^{4}/6.58 \times 10^{3}$ a</td>
</tr>
<tr>
<td>Pu-241</td>
<td>$3 \times 10^{10}$</td>
<td>13 a</td>
</tr>
<tr>
<td>Am-241</td>
<td>$1 \times 10^{9}$</td>
<td>458 a</td>
</tr>
<tr>
<td>U-234</td>
<td>$3 \times 10^{4}$</td>
<td>$2.5 \times 10^{5}$ a</td>
</tr>
<tr>
<td>U-235</td>
<td>$1 \times 10^{6}$</td>
<td>$7.1 \times 10^{8}$ a</td>
</tr>
<tr>
<td>U-238</td>
<td>$3 \times 10^{7}$</td>
<td>$4.5 \times 10^{9}$ a</td>
</tr>
<tr>
<td><strong>As gases</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>H-3</td>
<td>$1 \times 10^{12}$</td>
<td>12.3 a</td>
</tr>
<tr>
<td>Kr-85</td>
<td>$1 \times 10^{14}$</td>
<td>10.7 a</td>
</tr>
<tr>
<td>I-129</td>
<td>$2 \times 10^{7}$</td>
<td>$1.57 \times 10^{7}$ a</td>
</tr>
<tr>
<td>I-131</td>
<td>$1 \times 10^{2}$</td>
<td>8.0 d</td>
</tr>
<tr>
<td><strong>With particulates</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sr-90</td>
<td>$1 \times 10^{5}$</td>
<td>29 a</td>
</tr>
<tr>
<td>Ru-106</td>
<td>$3 \times 10^{6}$</td>
<td>1.0 a</td>
</tr>
<tr>
<td>Cs-134</td>
<td>$1 \times 10^{5}$</td>
<td>2.1 a</td>
</tr>
<tr>
<td>Cs-137</td>
<td>$3 \times 10^{5}$</td>
<td>30.1 a</td>
</tr>
<tr>
<td>Pu-238,239,240 : Pu(α)</td>
<td>$3 \times 10^{5}$</td>
<td>$86/2.44 \times 10^{4}/6.58 \times 10^{3}$ a</td>
</tr>
<tr>
<td>Pu-241</td>
<td>$1 \times 10^{6}$</td>
<td>13 a</td>
</tr>
<tr>
<td>Am-241</td>
<td>$3 \times 10^{4}$</td>
<td>458 a</td>
</tr>
<tr>
<td>Cm-242</td>
<td>$3 \times 10^{4}$</td>
<td>162 d</td>
</tr>
<tr>
<td>U-234</td>
<td>$1 \times 10^{7}$</td>
<td>$2.5 \times 10^{5}$ a</td>
</tr>
<tr>
<td>U-235</td>
<td>$3 \times 10^{5}$</td>
<td>$7.1 \times 10^{8}$ a</td>
</tr>
<tr>
<td>U-238</td>
<td>$1 \times 10^{7}$</td>
<td>$4.5 \times 10^{9}$ a</td>
</tr>
</tbody>
</table>
could be applied, and detection may be likely to result from accidental emissions only. In this regard, the radionuclides listed in Table I are not all equally likely to be released in the prescribed amounts. Notably, $^{85}$Kr (and possibly other noble gases) might be the most difficult to contain and might provide optimum signatures in some instances. It is assumed that releases of uranium from enrichment operations could be at levels similar to those from reprocessing.

2. PHYSICAL NATURE OF RELEASED MATERIALS AND MEDIA FOR ENVIRONMENTAL SAMPLING

2.1. Atmospheric releases

As shown in Table I, radionuclides can be released to the atmosphere as gases or in association with particles. Direct atmospheric sampling has the disadvantage that, since the release is likely to be episodic, plume interception might be difficult. Detection on any scale will depend on the development of a specific sampling grid, which will ultimately be limited by cost.

As an alternative to direct atmospheric sampling, deposition samples can be collected from vegetation, or in soil and rain (i.e. bulk deposition). There are potential complicating factors, which include background levels resulting from nuclear weapons fallout and Chernobyl and natural radioactivity (see Table II). The collection of deposition (either on vegetation or in rain) may include contamination from resuspension and, in the case of plants, from root uptake.

The main radionuclides likely to be released from a reprocessing operation are summarized as follows.

(a) Tritium is most likely to be in the form of HTO. This form of tritium rapidly exchanges with HTO in vegetation so that vegetational levels are indicative of recent discharges. Organically bound tritium in vegetation can be used as an indicator of historic discharges, but it is much less likely to be detected above background levels than free tritium following a short term release. It is better to measure tritium in rainfall than tritium in soil because of the history of deposition of background tritium in soil.

(b) Krypton-85 is effectively non-depositing (although it can sometimes be detected in rain) and might be best used in the identification of releases at long distances.

(c) Iodine-129 and iodine-131 are both likely to be released as vapours, although they might have an associated particulate fraction. Iodine-131 has a short half-life and sampling would need to occur after a suspected release. Iodine-129 has a great potential in identifying reprocessing activities because of the relatively high amounts released, although little is known about background levels. Analysis of soil,
TABLE II. BACKGROUND ENVIRONMENTAL CONCENTRATIONS OF RADIONUCLIDES

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Typical background concentrations</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Air (Bq/m³)</td>
</tr>
<tr>
<td>H-3</td>
<td>1 × 10⁻²</td>
</tr>
<tr>
<td>Kr-85</td>
<td>1.0</td>
</tr>
<tr>
<td>Sr-89</td>
<td>1 × 10⁻⁷</td>
</tr>
<tr>
<td>Sr-90</td>
<td>3 × 10⁻⁶</td>
</tr>
<tr>
<td>Zr-95</td>
<td>1 × 10⁻¹²</td>
</tr>
<tr>
<td>Tc-99</td>
<td>1</td>
</tr>
<tr>
<td>Cs-134</td>
<td>1 × 10⁻⁷</td>
</tr>
<tr>
<td>Cs-137</td>
<td>1 × 10⁻⁶</td>
</tr>
<tr>
<td>Ce-144</td>
<td>1 × 10⁻⁹</td>
</tr>
<tr>
<td>Pu(α)</td>
<td>1 × 10⁻⁹</td>
</tr>
<tr>
<td>Pu-241</td>
<td>3 × 10⁻⁹</td>
</tr>
<tr>
<td>Am-241</td>
<td>3 × 10⁻¹⁰</td>
</tr>
<tr>
<td>U-234</td>
<td>3 × 10⁻⁷</td>
</tr>
<tr>
<td>U-235</td>
<td>2 × 10⁻⁸</td>
</tr>
<tr>
<td>U-238</td>
<td>3 × 10⁻⁷</td>
</tr>
</tbody>
</table>

vegetation and rainwater is feasible. Analysis of milk and thyroids has indicated high iodine levels, although such samples are not usually practical to collect for safeguards purposes and it can be difficult to ascertain the grazing habits of the animals.

(d) Caesium-137, plutonium and americium-241 all have relatively high background levels in soil from nuclear weapons testing (and possibly Chernobyl fallout, depending on location). Caesium-137 can have a relatively high level of root uptake, so that detection of deposition on vegetation is difficult. Plutonium isotopes and ²⁴¹Am are relatively insoluble, so that root uptake is small. All of these radioisotopes are released in particulate form, so that exposed fibrous vegetation (e.g. pine needles, or species with sticky/hairy leaves) are optimum collectors. High background levels in soil can make rainfall measurements difficult because of contamination via resuspension and, similarly, resuspension and soil splash can contaminate short vegetation.

Plutonium in herbivore faeces has been used to detect the presence of nuclear operations. In the case of Sellafield, plutonium originating from sea-to-land transfer has been detected in the characteristic $^{238}\text{Pu}:^{239}\text{Pu} + ^{240}\text{Pu}$ ratios found more than 100 km from the source [1]. Plutonium is poorly absorbed by the gut, and herbivore species effectively act as deposition integrators.
(e) Caesium-134 was not present to any extent in nuclear weapons fallout, although some material remains in the environment since the Chernobyl accident. The half-life of $^{134}$Cs of 2.1 years has enabled much of the Chernobyl derived material to decay, although predicted releases from undeclared facilities are small. Certain species of fungi, moss and lichens are known to accumulate $^{134}$Cs (and other radionuclides), and measured levels, especially if they are compared with $^{137}$Cs levels, can provide evidence of nuclear operations.

(f) Other fission products which are potentially useful in the detection of reprocessing activities include $^{106}$Ru and $^{90}$Sr.

It is worth while to note the deposition mechanisms that will result in contamination of the terrestrial environment. Close to the release point, dry deposition will dominate and the efficiency of a collection surface will depend on its aerodynamic properties (fibrous surfaces are the best collectors). At greater distances, material will become mixed within clouds and deposition in rain will ultimately dominate dry deposition (assuming the occurrence of precipitation). Thus, the use of rain collectors or the collection of vegetation, which effectively intercepts and retains rain, is preferred at greater distances.

Resuspension, soil splash and root uptake of deposited background material can act as sources of interference in the determination of dry deposition. Thus, rain collectors should be positioned above a surface and any vegetation sampled should preferably be one metre or more above the soil if resuspension or soil splash are potential problems. Certain vegetational species accumulate radionuclides. Fungi are intimately linked with the soil and high radionuclide concentrations in them are indicative of soil concentrations. Mosses and lichens, on the other hand, are more indicative of atmospheric concentrations. Lichens do, however, have the potential to take up radioactivity from the substrate on which they exist.

As previously noted, the environmental signature associated with enrichment facilities will be uranium in one form or another. Soil has high background levels of uranium and is, therefore, likely to be a poor sample type, although sorbed uranium can be solubilized by using a carbonate leaching technique, with the minimum removal of uranium associated with mineral matrices. Since released uranium will become associated with particles, the collection of vegetation samples might prove optimum, especially those which are efficient at collecting dry deposition (i.e. fibrous, sticky materials).

The above considerations are for off-site sampling only. On the site, the chances of detection increase dramatically and the use of swipe samples (i.e. tissue wipes) from work areas and dusty surfaces is likely to give the best chances of detection.
2.2. Aquatic releases

Aquatic releases of radionuclides can be detected either in water and sediment (deposited or suspended) or in aquatic biota. Generally, it is more difficult to detect discharges in the sea because of tides and strong currents, and because the sea is a large medium for dispersal. Nevertheless, given certain conditions (e.g. thermal stratification), detection of even small releases is at least theoretically possible at quite long distances from the discharge point. Freshwater sampling is desirable, since freshwater systems are effectively one-dimensional, with slow migration of material downstream. Not all facilities, however, may have access to freshwater systems. The success of water sampling (including suspended sediments) in the detection of discharges is likely to depend on the interception of the release at some point downstream (although this is not necessarily the case, since exchange with deposited sediments will continually take place, in terms of both dissolution and suspension of deposited sediments). Deposited sediments will provide a record of discharges, although the activity associated with them is dependent on the grain size (being determined by the surface area), the type of radionuclide and the chemical composition of the water (e.g. salinity). The ratio of concentration in the sediment to concentration in the aqueous phase is usually described by the distribution coefficient $K_d$, which normally relates to fine (suspendable) sediments. Some examples of $K_d$ are given in Table III. It is apparent that the probabilities of detection in either phase are determined by the values of $K_d$. To optimize the chances of detection in sediment, suspended material or fine deposited sediments should be analysed.

### Table III. Examples of the Distribution Coefficient $K_d$ for Elements in Marine and Freshwater Sediments [2]

<table>
<thead>
<tr>
<th>Element</th>
<th>$K_d$ (marine)</th>
<th>$K_d$ (freshwater)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag</td>
<td>$2 \times 10^3$</td>
<td>$4 \times 10^3$</td>
</tr>
<tr>
<td>Am</td>
<td>$4 \times 10^5$</td>
<td>$4 \times 10^5$</td>
</tr>
<tr>
<td>Ce</td>
<td>$1 \times 10^4$</td>
<td>$&lt;10^4$</td>
</tr>
<tr>
<td>Cm</td>
<td>$10^5$ to $10^6$</td>
<td>$10^4$ to $10^5$</td>
</tr>
<tr>
<td>Co</td>
<td>$1 \times 10^4$</td>
<td>$1 \times 10^3$</td>
</tr>
<tr>
<td>Cs</td>
<td>$2.5 \times 10^3$</td>
<td>$2 \times 10^4$</td>
</tr>
<tr>
<td>I</td>
<td>$1 \times 10^1$</td>
<td>$3 \times 10^2$</td>
</tr>
<tr>
<td>Nb</td>
<td>$1 \times 10^6$</td>
<td>$1 \times 10^6$</td>
</tr>
<tr>
<td>Np</td>
<td>$\sim 10^3$ to $10^4$</td>
<td>$1 \times 10^5$</td>
</tr>
<tr>
<td>Pu</td>
<td>$5 \times 10^4$</td>
<td>$1 \times 10^5$</td>
</tr>
<tr>
<td>Sb</td>
<td>$2 \times 10^3$</td>
<td>$2 \times 10^3$</td>
</tr>
<tr>
<td>Sr</td>
<td>$1 \times 10^2$</td>
<td>$1 \times 10^3$</td>
</tr>
</tbody>
</table>
TABLE IV. SAMPLE RATIOS OF RADIONUCLIDE CONCENTRATIONS IN BIOTA TO CONCENTRATIONS IN WATER [3]

<table>
<thead>
<tr>
<th>Element</th>
<th>Macrophytes</th>
<th>Invertebrates</th>
<th>Fish</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag</td>
<td>$2 \times 10^2$</td>
<td>$7.7 \times 10^2$</td>
<td>$2.3 \times 10^6$</td>
</tr>
<tr>
<td>Ce</td>
<td>$5 \times 10^3$</td>
<td>$1 \times 10^3$</td>
<td>$2.5 \times 10^1$</td>
</tr>
<tr>
<td>Co</td>
<td>$2 \times 10^2$</td>
<td>$2 \times 10^2$</td>
<td>$2 \times 10^1$</td>
</tr>
<tr>
<td>Cs</td>
<td>$8 \times 10^1$</td>
<td>$5 \times 10^1$</td>
<td>$4 \times 10^2$</td>
</tr>
<tr>
<td>I</td>
<td>$4 \times 10^1$</td>
<td>$5 \times 10^0$</td>
<td>$1.5 \times 10^1$</td>
</tr>
<tr>
<td>Nb</td>
<td>$8 \times 10^2$</td>
<td>$1 \times 10^2$</td>
<td>$3 \times 10^4$</td>
</tr>
<tr>
<td>Ru</td>
<td>$3.5 \times 10^2$</td>
<td>$1 \times 10^2$</td>
<td>$1 \times 10^1$</td>
</tr>
<tr>
<td>Sr</td>
<td>$2 \times 10^2$</td>
<td>$3 \times 10^2$</td>
<td>$1 \times 10^1$</td>
</tr>
<tr>
<td>U</td>
<td>$5 \times 10^2$</td>
<td>$1 \times 10^2$</td>
<td>$5 \times 10^9$</td>
</tr>
<tr>
<td>Zr</td>
<td>$1 \times 10^3$</td>
<td>$6.7 \times 10^0$</td>
<td>$3.3 \times 10^9$</td>
</tr>
</tbody>
</table>

Typical background concentrations of some of the major radionuclides are listed in Table II.

Aquatic plants are often accumulators of radionuclides. This is also true for invertebrates and fish (see Table IV), although the former can be difficult to locate and the latter tend to be nomadic. Often, aquatic sampling of biota will be restricted by the species available, although the collection of any vegetation is likely to be expedient.

It is important to note that an aquatic discharge might not necessarily come directly from a nuclear process but can result from the run-off of atmospherically deposited material. The water channel in this case can act as an integrator of aerial deposition.

3. SUMMARY

For the detection of radionuclide signatures released from undeclared nuclear processes, there is a range of environmental samples that can be collected and a number of radionuclides that can be analysed. For reprocessing and reactor operations, these will include actinides, fission products and activation products; for enrichment facilities, they will be restricted to uranium isotopes. The distances at which a nuclear process can be detected are dependent on the magnitude of the releases and the sensitivity of the analytical techniques deployed. This will ultimately be restricted by cost, although detection of radionuclide signatures over several tens or hundreds of kilometres is feasible, given an appropriate sampling programme.
REFERENCES


RADIONUCLIDE RELEASES TO THE ATMOSPHERE

(Session 2)

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F. FRY
United Kingdom
TRANSPORT OF RADIOACTIVE GASES AND PARTICLES FROM THE CHERNOBYL ACCIDENT

Comparison of environmental measurements and dispersion calculations

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Abstract

TRANSPORT OF RADIOACTIVE GASES AND PARTICLES FROM THE CHERNOBYL ACCIDENT: COMPARISON OF ENVIRONMENTAL MEASUREMENTS AND DISPERSION CALCULATIONS.

Transport and dispersion of gases and particles during the early stages of the Chernobyl accident are studied. Calculations have been performed using the long range atmospheric transport, dispersion and dose model TRADOS. The model uses high resolution weather data in calculating three-dimensional trajectories of air parcels and particles of different sizes. The trajectories show that the plume was segmented during the transport. The radioactive material deposited in Finland was transported at heights between 1400 and 3000 m. The largest particles transported to Finland had an aerodynamic diameter of 20 μm. Particle trajectories may explain some of the differences between observations and earlier calculations. Large particles originally in the upper air layer settled to lower air layers where wind conditions were different. To explain the deposition of large radioactive particles found in Sweden and Poland, it is necessary to assume convective updraft in cumulus clouds over Belarus during the daytime of 26 April 1986. The Finnish fallout map for $^{95}$Zr that was reconstructed in the simulations agrees well with the measurements.

1. INTRODUCTION

The Chernobyl accident in 1986 dispersed radioactive material all over Europe. The massive release following the explosion of Unit 4 (1986-04-25—21:23 UTC) contained large fuel fragments and other particles that were transported in a different way compared with gases and small particles. The fallout of some radionuclides, such as $^{137}$Cs, is well documented [1]. This contamination, however, is only part of the radiation hazard caused by the accident. The fallout pattern of some other nuclides is not well known in wide areas. Prompt nuclide specific measurements were performed only in very few locations.
The accident dispersed $2 \times 10^{18}$ Bq of condensable radioactive material, i.e. particles or gases that were later transformed to particulates [2]. About 25% of this material was released during the early stage of the accident in the energetic initiating events. The plume rose up to a height of a few kilometres. Later, the effective release height was considerably lower (less than about 400 m [3]).

The total mass of the radioactive particles released in the accident was about 6000–8000 kg [4]. More than half of the material was deposited near the plant (2–2.5% of the fuel mass [2]), but some particles (1–1.5% of the fuel mass) travelled thousands of kilometres. Generally, volatile elements, such as iodine and caesium, were transported far away (they were in gaseous form or in small particles). The non-volatile elements cerium and zirconium remained within the fragments of powderized fuel. The transport of these elements depends strongly on the size distribution of the carrier particles.

The dispersion conditions during the accident are well described [3, 5, 6]. At the beginning of the accident there was a stable boundary layer with an inversion at a level of 400–500 m over the Chernobyl area. Below this level the winds were very weak. At a level of about 1000–1500 m the air flow was towards the north-west, with wind speeds of about 10 m/s. Between these two layers there were much stronger winds. However, the thickness of this low level jet (maximum wind speed over 17 m/s) was only a few hundred metres.

The plume was transported initially towards the Baltic Sea. The plume was segmented into two parts over north-eastern Poland and Lithuania. Because of this separation the lower part of the plume (below 750–1000 m [3]) was transported towards Sweden, whereas the higher parts of the plume turned north towards Finland. This split plume hit the Nordic countries on 28 April, approximately two days after the explosion of the reactor [3, 5, 6]. After this first release stage the large scale weather pattern changed and the radioactive materials were transported to other parts of Europe.

The transport of gases and particles during the early phase of the accident is considered in the present paper. The release following the explosion of the reactor contained large amounts of highly radioactive particles ($d_a > 20 \mu m$) that were transported hundreds of kilometres away from the plant. The Finnish fallout maps for $^{137}$Cs and $^{95}$Zr [7] show that these nuclides were released, transported and deposited in different ways. Simulations are needed to explain these differences and other particle size dependent phenomena in long distance transport.

2. DISPERSION AND DOSE CALCULATIONS

A trajectory, dispersion and dose model, known as TRADOS, has been developed by the Finnish Meteorological Institute (FMI) and the Technical Research Centre of Finland [8]. TRADOS can perform real-time calculations for nuclear acci-
TRADOS is a Lagrangian-Gaussian trajectory model including a radiological dose module. The statistical version of the model is used for risk assessment and population dose estimation.

The trajectory module of TRADOS produces dispersion meteorology data for the dose module. Transport of the radioactive material is described by a set of three-dimensional trajectories. Each trajectory within a set can start either from a different elevation or at a different time. Trajectories can be calculated either forwards or backwards. Limited cluster analysis, for displacement of the starting points of trajectories 30–50 km from the release site, enables the reliability of the trajectories to be estimated.

TRADOS can be used for estimating the dispersion of particles. Because of gravitational settling, particles descend to lower air layers where different winds and transport conditions prevail [9, 10]. The resulting separation of particle and air parcel trajectories is usually most prominent around the areas of strong cyclonic curvature with strong pressure gradients. Contrary to air parcel trajectories, the particle trajectories are terminated when they reach the ground.

The lateral dispersion of radioactive material is simulated by trajectories starting at three-hour intervals. The total release is divided into segments of three hours. The plume width is calculated from the quadratic sum of the spread due to internal turbulence of the plume and from the external force due to meandering and wind veering. Horizontal dispersion, vertical concentration profile, mixing height, time integrated air concentration at ground level, and dry and wet deposition are calculated for each one-hour step at the trajectory.

The vertical concentration profile within the mixing layer is described by the gradient transfer approach using steady state $K_z$ profiles [8]. The dispersion is described using the concept of transport level. Only one transport level at a time is selected. Dry deposition is treated as a boundary condition for the vertical profile. At present, only one value is used for the dry deposition velocity (0.01 m/s). Material removal by precipitation is described by the scavenging coefficient approach. Contrary to dry deposition, wet deposition removes material from all layers of the cloud. The noble gases, however, are not affected by the removal processes.

TRADOS uses numerical weather data from the Finnish version of the Nordic weather prediction model HIRLAM (high resolution limited area model). Weather parameters from seven pressure levels, from ground surface up to 100 hPa (ca. 15 km), from each HIRLAM run, measured four times a day, are stored routinely in a special database. The spatial resolution of the model is 55 km.

The HIRLAM weather model was not operational in 1986. The Danish Meteorological Institute has now compiled a special HIRLAM database covering the period from 25 April to 10 May 1986. This database is used in the present study. The same weather data were used previously by the Swedish Meteorological Institute [11].
3. AIR PARCEL TRAJECTORIES

A prominent feature of the three-dimensional transport of radionuclides is the 'separation' of the plume when it approached Sweden and Finland [3, 5]. According to the present simulations, the trajectories originating above the level of 1200-1400 m moved towards Finland, whereas at lower heights the destination was Sweden (see insert in Fig. 1). Extensive trajectory calculations gave the following

*FIG. 1. Measured $^{95}$Zr fallout in Finland [7] compared with model calculations. The solid line, calculated with TRADOS, is an iso-curve of 2 kBq/m$^2$. The simulated fallout is integrated up to 1986-04-28—03:30 UTC. The transport height is 2000 m. Air trajectories, originating from Chernobyl at 1986-04-25—21:00 UTC, are shown in the upper left corner. The effective release heights are: A — 750 m, B — 1000 m, C — 1500 m and D — 2500 m.*
results for the early transport of gases and small particles (\(<5 \mu m\)) that follow air streams completely:

1) No trajectory originating below 400 m reached Sweden.
2) Trajectories originating between 400 and 600 m just reached the Swedish east coast.
3) The simulations cannot explain the low level jet that the synoptic data suggest. The anomaly was revealed by a direct analysis of the synoptic data.
4) Most trajectories between 500 and 1100 m from 1986-04-25—21:30 UTC to 1986-04-26—06:00 UTC arrived in Sweden.
5) No trajectory originating below 1000 m reached Finland.
6) No trajectory originating between 1000 and 1400 m reached Finland, unless their start was later than 1986-04-26—02:00 UTC.
7) Practically every trajectory originating between 1400 and 3000 m reached Finland (1986-04-25—21:30 UTC to 1986-04-26—08:00 UTC). The trajectories starting after 1986-04-26—08:00 UTC at 1000—1500 m did not reach Finland.
8) The trajectories starting before 1986-04-26—16:00 UTC and being higher than 3000 m arrived in Finland. The trajectories starting between 18:00 and 20:00 UTC arrived in Sweden. Later, the transport direction took a fast turn towards central Europe.
9) The altitude behaviour of the trajectories reaching Finland is as follows:

<table>
<thead>
<tr>
<th>Initial height (m)</th>
<th>Arrival height (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2500—4000</td>
<td>2000—2500</td>
</tr>
<tr>
<td>1600—2500</td>
<td>1500—1700</td>
</tr>
<tr>
<td>1500</td>
<td>1000—1300</td>
</tr>
<tr>
<td>1000—1400</td>
<td>800—1000</td>
</tr>
</tbody>
</table>

The dose rate measurements in southern Finland using aircraft showed that the maximum airborne activity on 28 April was at an altitude of 1500 m [12]. Similar measurements in Sweden over the Baltic Sea between Gotland and Stockholm showed that the centre of the plume was at an altitude of about 750 m [3]. These observations are supported by the present trajectory calculations.

The Soviet authorities issued no information on the accident before 28 April 1986. Consequently, only a few observations with high temporal resolution are avail-
TABLE I. TIME (UTC) OF PLUME ARRIVAL ACCORDING TO TRAJECTORY CALCULATIONS

<table>
<thead>
<tr>
<th>Release (m)</th>
<th>South-west Finland</th>
<th>Ahvenanmaa</th>
<th>Sweden (eastern Svealand)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2000</td>
<td>1986-04-27—12:00</td>
<td>1986-04-27—15:00</td>
<td>—</td>
</tr>
<tr>
<td>1500</td>
<td>1986-04-27—15:00</td>
<td>1986-04-27—16:00</td>
<td>1986-04-27—18:00 (800 m)</td>
</tr>
<tr>
<td>1300–1400</td>
<td>1986-08-28—00:00 to 03:00</td>
<td>1986-04-27—18:00 (1000 m)</td>
<td>1986-04-27—12:00 (500 m)</td>
</tr>
<tr>
<td>1000</td>
<td>—</td>
<td>—</td>
<td>1986-04-27—20:00 to 23:00 (400 m)</td>
</tr>
<tr>
<td>500–750</td>
<td>—</td>
<td>—</td>
<td></td>
</tr>
</tbody>
</table>

The height of the trajectory is given in parentheses.

Trajectories originating between 500 and 1000 m arrived in Gotland (1986-04-27—09:00 to 12:00 UTC at a height of 300–400 m).
able from the early phase, when the plume hit the Nordic countries. Later interpretation of the monitoring results gives site specific estimates for the arrival times of the plume. However, these results, as well as trajectory calculations (Table I), do not always allow unequivocal interpretation.

The monitoring data show that the plume first arrived over Gotland about 1986-04-27—12:00 UTC [3]. It reached the central and southern parts of the Swedish mainland on the evening of this day, some time before 18:00 UTC. During the night between 27 and 28 April, artificial radionuclides entered the surface air on the Finnish south-western coast. However, because of a strike of civil servants, the monitoring station in Ahvenanmaa between Finland and Sweden was unattended. Thus, the first alert went unnoticed.

Air sampling in southern Finland showed that large amounts of artificial radionuclides had arrived in Finland before 1986-04-28—06:00 UTC. The maximum concentration measured in Helsinki at 18:00 UTC was about 30 Bq/m$^3$ for $^{131}$I [12]. According to the trajectory calculations the radioactive material arrived in Helsinki via central Finland (not from the south) at 1986-04-28—08:00 to 12:00 UTC. During the following night at 00:00 UTC the measured nuclide concentration in air was reduced by a factor of ten and later in the morning, at 1986-04-29—09:00 UTC, by another factor of ten.

4. DEPOSITION OF $^{95}$Zr — MEASUREMENTS AND SIMULATIONS

The fallout pattern in Finland is highly nuclide specific [7]. The deposition of volatile fission products, $^{137}$Cs in particular, comes from the releases at different stages after the explosion. The deposition pattern of $^{103}$Ru is a combination of the fallout due to the initial explosion and the reactor burn. Ruthenium fallout is greatly affected by the later releases and the wet deposition that occurred between 10 and 12 May.

Zirconium and cerium are non-volatile elements. During the release they remained attached to small uranium fuel particles. The measured fallout of $^{95}$Zr is a direct marker of the plume transport during the early phase of the accident. The cloud, originating from the explosion and from subsequent prompt releases, passed over southern and middle Finland in twelve hours from 1986-04-27—12:00 UTC onwards. Zirconium-95 was also released at later stages, but its contribution to the fallout pattern in Finland was only secondary.

Dispersion calculations were performed with TRADOS. The total release of $^{95}$Zr, which was dispersed into small particles ($d_a \leq 20$ $\mu$m) and rose initially above 1500 m, is assumed to be about $9 \times 10^{15}$ Bq during the first nine hours of the accident (Fig. 1). A height of 2000 m was chosen for the transport level. The trajectories originating from 1500 m and 2500 m behave similarly. A transport level of 3000 m would move the deposition pattern towards north, giving better agreement
with the measurements (results not shown). The spatial coverage, however, would be much narrower and, thus, this transport level seems to be somewhat too high.

A typical feature of trajectory models is that the plume is often narrower than predicted by other models, e.g. models using the Eulerian approach. This behaviour is also seen in the present study. In addition, the model tends to create very steep concentration gradients near the boundaries of the plume. The measured fallout pattern is wider than predicted by the model (Fig. 1). However, TRADOS has well identified the areas at risk at a distance of 1500 km, and, moreover, the simulated absolute fallout is consistent with the measurements.

5. PARTICLE TRAJECTORIES

For emergency preparedness purposes it is essential to realize the existence of the radiological hazard caused by the highly radioactive particles. For example, the present skin dose limit of the International Commission on Radiological Protection [13] for the public (50 mSv averaged over 1 cm$^2$) is exceeded if a uranium fuel particle larger than 10 μm in physical diameter is deposited on the skin for one day [14]. The transport range of particles of different sizes formed in the initial explosion in Chernobyl is given in Table II.

Radioactive particles found in the environment after the Chernobyl accident are classified into monoelemental (or bielemental) particles and multielemental fuel fragments. Some particles were hundreds of micrometres in size and their activity was up to hundreds of kilobecquerels. The activity of monoelemental ruthenium particles found in Poland, 500–650 km away from Chernobyl, was on the average $19 \pm 3$ kBq, whereas that of uranium fuel fragments was $1.1 \pm 0.1$ kBq [15]. Ruthenium particles were up to 14 μm in physical diameter ($d_a \approx 50$ μm) [16]. Table II shows that even a release height of 3000 m does not explain these findings. Thus, either the plume rise was higher than estimated or, more likely, convective updraft in cumulus clouds over Belarus during the daytime of 26 April moved particles upwards [3, 17].

The largest fuel particle found in Finland has a diameter of 7 μm ($d_a \approx 20$ μm) [18]. Trajectory calculations show that particles up to 20 μm in aerodynamic diameter reached Finland, provided they were originally lifted to a height of 2500 m. Convective ascent is needed to explain the findings if the release height is below 2500 m.

Falk et al. [19] reported that some ruthenium particles found in Sweden, more than 1000 km away from Chernobyl, were rather large (up to $d_p = 12$ μm, $d_a \approx 37$ μm). This study shows that air parcel trajectories did not reach Sweden if they started at levels higher than about 1100 m. The maximum diameter of particles transported to Sweden within this lower layer is $d_a \approx 12$ μm (Table II). The large particles must initially have been in the upper air layer. During transport, gravita-
TABLE II. TRANSPORT RANGE OF PARTICLES OF DIFFERENT SIZES RELEASED IN CHERNOBYL AT THE TIME OF REACTOR EXPLOSION.a

<table>
<thead>
<tr>
<th>Particle size (µm)</th>
<th>Transport range (km) for release heights of 500-3000 m</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>500 m</td>
</tr>
<tr>
<td>10</td>
<td>3.1</td>
</tr>
<tr>
<td>20</td>
<td>6.2</td>
</tr>
<tr>
<td>30</td>
<td>9.3</td>
</tr>
<tr>
<td>40</td>
<td>12</td>
</tr>
<tr>
<td>50</td>
<td>15</td>
</tr>
<tr>
<td>60</td>
<td>19</td>
</tr>
<tr>
<td>70</td>
<td>22</td>
</tr>
</tbody>
</table>

a The length of the trajectories was calculated with TRADOS. The travel distances were calculated for particles of aerodynamic diameter \(d_a\). A conversion to physical diameter was made using a density of 10,500 kg/m³ for the particles \(d_p = d_a/3.2\).

6. DISCUSSION

Three-dimensional trajectory analysis takes into account the real meteorological conditions that are of particular importance for particle transport. Particles in the upper air layer settle to lower air layers where wind conditions are different. Because of gravitational settling, the particles may be transported to areas that are not reached by air trajectories.

The present simulations show that part of the radioactive particles at an initial height of 1500 m or at higher altitudes may have been transported to Sweden, although the air parcel trajectories from the same altitudes arrived in Finland. The fallout pattern of \(^{137}\)Cs in central Sweden has not been fully explained previously [3, 11]. The particle trajectories may explain part of the difference between observations and calculations.

Air concentration measurements were performed only in very few locations. In southern Finland (Nurmijärvi) the measured integrated air concentration of \(^{131}\)I was \(10^7\) Bq·s/m³ up to 30 April [12]. The preliminary transport and dispersion calculations give equal results. The simulations could be valuable for reconstructing the exposure to iodine from inhalation in areas where no measurements are available.
FIG. 2. Trajectories of an air parcel and a group of particles with different aerodynamic diameters ($\mu$m) released from Chernobyl at the time of explosion. The trajectories refer to an effective release height of 1500 m. The deposition sites are indicated by hatched circles. Small particles (5-10 $\mu$m) remain airborne because of small sedimentation velocity and large scale ascending motion of air at large distances. The transport distances of particles are much longer for release heights above 2000 m (see Table II).

The radiological hazard caused by short lived nuclides varies, depending on the time interval between the reactor explosion and the fallout on the target sites. Radioactive decay is of particular importance for thyroid exposure through inhalation. Iodine-131 causes the largest thyroid doses. However, the other isotopes of iodine ($^{132}$I to $^{135}$I) are also important during the first hours and days after the accident. Moreover, these isotopes emit beta particles that have high energies. The quality of radiation may be important in the induction of the thyroid cancers that have been observed in Belarus [20-22]. Further studies are needed to find out the nuclide specific integrated air concentrations at the trajectories of the initial plume.
ACKNOWLEDGEMENTS

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REFERENCES


VERITAL DISTRIBUTION
OF NATURAL AND ARTIFICIAL RADIONUCLIDES
IN THE ATMOSPHERE

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Abstract

VERTICAL DISTRIBUTION OF NATURAL AND ARTIFICIAL RADIONUCLIDES IN THE ATMOSPHERE.

During the past 22 years the high altitude monitoring programme (HAMP) of the Polish early warning system provided information on the vertical distribution of natural radionuclides and fission products in the troposphere and stratosphere between ground level and 15 km altitude. It was found that the stratospheric concentrations of $^{226}$Ra increased after large volcanic eruptions. The vertical concentration profile of $^{226}$Ra had a characteristic parabolic distribution, with the highest concentrations at ground level and in the stratosphere, and the lowest concentrations just below the tropopause. Concentrations of $^{210}$Pb, due to quiescent ascent of its gaseous $^{222}$Rn precursor from the ground, had a more homogeneous distribution. Vertical concentrations of fission products revealed different types of profile. Immediately after the Chernobyl accident, part of the radioactive debris entered the upper troposphere and the lower stratosphere. Because of resuspension, the stratospheric residence time of radiocaesium from the Chernobyl reactor was about three times longer than that of fallout from nuclear explosions. During the first few weeks after the accident, about 0.5% of the activity present near the ground level was uplifted into the stratosphere with ascending air currents. Between 1987 and 1994 the fraction of radiocaesium present at an altitude of 15 km reached 58% to > 100% of that at ground level, owing to resuspension of radioactive particles from the highly contaminated ground in the Chernobyl region.

1. INTRODUCTION

Several programmes for high altitude aircraft sampling of radionuclides have been carried out systematically since the 1950s in the troposphere and the stratosphere over Japan, Sweden, the former Soviet Union and the United Kingdom. Most of these programmes were short term studies and they were terminated in the 1970s. The largest, 27 years long, sampling project, covering a large part of the Earth, was performed by the US Defence Atomic Support Agency and was terminated in 1983 [1]. These studies contributed to our understanding of atmospheric mass transport and of the behaviour of radioactive and stable particles in the atmosphere. Their results were also used for projections of the public hazard of atmospheric emissions of radionuclides.
In most of these studies, complete short term vertical concentration profiles of radionuclides were not obtained. Half-day vertical profiles of fission products and natural radionuclides have been systematically recorded only over the north-eastern part of Poland, by the high altitude monitoring programme (HAMP). We present here a summary of the results of measurements carried out in the troposphere and the stratosphere during the last 22 years.

2. METHODS

Atmospheric particulates have been collected on chlorinated PVC filters with a stationary sampler near ground level, and with aircraft samplers at altitudes of 1, 3, 6, 9, 12 and 15 km. The particulates were collected during horizontal flights by means of ram pressure. Details of sampling, analytical and quality control procedures are given elsewhere [2-5].

Between 1973 and 1994, we collected 635 samples of radioactive aerosols, which were used to obtain 109 vertical concentration profiles of $^{90}$Sr, $^{134,137}$Cs, $^{210}$Pb and $^{226}$Ra. The samples for each concentration profile were collected during 5–6 h, under stable weather conditions from cloudless regions of the sky.

3. RESULTS AND DISCUSSION

During the 22 years of the study, several natural and man-made events have occurred which have influenced the levels of radionuclides at particular altitudes: the Chinese nuclear explosions in the atmosphere, the Chernobyl accident and numerous volcanic eruptions. Because of these events, the individual concentration profiles were changing by two or three orders of magnitude, depending on the location of the radionuclide sources and on the meteorological situation [2, 6-8]. The Chernobyl emissions occurred in the 'silent period' after the last Chinese explosion, in which the levels of radionuclides in the stratosphere and troposphere were much lower than in the three previous decades. This enabled us to observe effects which were hidden previously in the high background of stratospheric debris from the frequent nuclear tests.

3.1. Natural radionuclides

Dust particles in the atmosphere containing natural radionuclides have two types of sources: one type is a quiescent, continuous ascent from the ground level, and the other types are short term violent stratospheric sources, i.e. volcanic eruptions. The combined effect of stratospheric and ground level sources produces a characteristic parabolic vertical profile of the long term mean concentration of
The more uniform vertical distribution of $^{210}\text{Pb}$ is an effect of the quiescent ascent of its gaseous $^{222}\text{Rn}$ parent from the ground. The difference between the profiles of natural radionuclides and those of fission products from nuclear tests is due to the fact that the dominant source of the latter was the stratosphere, and the resuspension effect was below the detection limit. The particular concentration profiles of all nuclides were strongly dependent on changes in height of the tropopause, on short term changes of weather conditions and, to a lesser extent, on the season [7].

As demonstrated in Fig. 2, the $^{226}\text{Ra}$ content in the 1 m$^2$ column of air between the ground level and the lower stratosphere increases after large volcanic eruptions. Using mean concentrations for the period 1980–1993, we estimated tentatively that the $^{226}\text{Ra}$ content in the global atmosphere between the ground level and an altitude of 15 km was about $2 \times 10^{13}$ Bq. After eruption of the Fuego volcano...
Average contents of radium-226 in the global atmosphere:

- 1980 - 1987: $2.23 \times 10^{13}$ Bq
- 1988 - 1993: $2.09 \times 10^{13}$ Bq
- 1975 - 1979: $3.47 \times 10^{13}$ Bq

**FIG. 2.** Temporal changes of the content of $^{226}$Ra in the 1 m$^2$ column of air between the ground level and an altitude of 15 km.
in 1975 the concentration of $^{226}\text{Ra}$ in air increased rapidly, and during the period 1975–1979 the content of $^{226}\text{Ra}$ in the global atmosphere at altitudes between 0 and 12 km (about $4 \times 10^{13} \text{ Bq}$) was higher than that in other periods [7].

3.2. Fission products

3.2.1. Chinese nuclear tests

During the last twenty years we measured concentrations of fission products in three different situations: (1) in the period of intensive Chinese and French nuclear weapons tests in the atmosphere, which ended in October 1980; (2) in the 'silent period' of no atmospheric nuclear events; and (3) at the time of the Chernobyl accident and in the following period.

In the first period the vertical distribution of fission products in the troposphere increased steadily with altitude, and there was an abrupt increase in the stratosphere. All Chinese explosions in the megatonne range were easily detected over Poland. We obtained the most interesting results after the last Chinese 0.5 Mt test on 16 October 1980 [8]. Nine days after the explosions we collected aerosols from the radioactive cloud during its first circumnavigation of the Earth. The cloud was still contained in a limited parcel of air. In the sample collected on 25 October at an altitude of 15 km we found very high concentrations of fission products, and only slightly elevated concentrations at altitudes of 12, 10 and 8 km (Table I). Three days later, on 28 October, the concentrations of most of the radionuclides had decreased at 15 km by several orders of magnitude to pre-explosion levels.

During the next several months, the radioactivity of stratospheric air decreased steadily (Table I) and the maximum values of monthly concentrations of $^{137}\text{Cs}$ and $^{144}\text{Ce}$ descended successively from higher altitudes to lower ones. The rate of descent was in accordance with the theories of Machta [9] and Telegadas [10]. The mean stratospheric residence time of $^{137}\text{Cs}$ during the first three years after the last test was 240 days.

3.2.2. Chernobyl accident

Early in the morning of 29 April 1986, i.e. three days after the Chernobyl reactor accident, we started measurements of the vertical profiles of radioactivity concentrations in the troposphere and stratosphere over the north-eastern part of Poland. The measurements were carried out daily until 2 May, and later at longer intervals. In the first few weeks after the accident the results of these measurements (Fig. 3) were used for projections of the contamination of the air space over Poland.

On 29 April the elevated activity concentrations of radiocaesium were confined to the ground level air, with trace concentrations reaching the stratosphere. On 30 April, high activity was observed between 0 and 3 km; in the stratosphere at
TABLE I. VERTICAL DISTRIBUTION OF $^{90}$Sr, $^{137}$Cs AND $^{144}$Cs (mBq/m$^3$ STP) BEFORE AND AFTER THE NUCLEAR EXPLOSION IN THE ATMOSPHERE ON 16 OCTOBER 1980

<table>
<thead>
<tr>
<th>Date and altitude</th>
<th>$^{90}$Sr</th>
<th>$^{137}$Cs</th>
<th>$^{144}$Ce</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>17 July 1980</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6 km</td>
<td>0.06</td>
<td>0.09</td>
<td>0.16</td>
</tr>
<tr>
<td>9 km</td>
<td>0.44</td>
<td>0.75</td>
<td>1.16</td>
</tr>
<tr>
<td>12 km</td>
<td>0.97</td>
<td>1.22</td>
<td>2.52</td>
</tr>
<tr>
<td>15 km</td>
<td>0.35</td>
<td>0.56</td>
<td>4.19</td>
</tr>
<tr>
<td><strong>25 Oct. 1980</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8 km</td>
<td>&lt;0.09</td>
<td>0.05</td>
<td>0.29</td>
</tr>
<tr>
<td>10 km</td>
<td>0.07</td>
<td>0.09</td>
<td>2.81</td>
</tr>
<tr>
<td>12 km</td>
<td>1.92</td>
<td>2.69</td>
<td>73.68</td>
</tr>
<tr>
<td>15 km</td>
<td>321.40</td>
<td>542.50</td>
<td>15 533.20</td>
</tr>
<tr>
<td><strong>28 Oct. 1980</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6 km</td>
<td>&lt;0.07</td>
<td>&lt;0.06</td>
<td>0.13</td>
</tr>
<tr>
<td>8 km</td>
<td>&lt;0.09</td>
<td>&lt;0.08</td>
<td>0.36</td>
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<tr>
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<td>&lt;0.12</td>
<td>&lt;0.11</td>
<td>0.66</td>
</tr>
<tr>
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<td>0.15</td>
<td>0.23</td>
<td>1.58</td>
</tr>
<tr>
<td>15 km</td>
<td>0.33</td>
<td>0.52</td>
<td>42.08</td>
</tr>
<tr>
<td><strong>17 Dec. 1980</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6 km</td>
<td>0.04</td>
<td>0.07</td>
<td>1.79</td>
</tr>
<tr>
<td>9 km</td>
<td>0.29</td>
<td>0.44</td>
<td>10.43</td>
</tr>
<tr>
<td>12 km</td>
<td>1.79</td>
<td>2.85</td>
<td>61.45</td>
</tr>
<tr>
<td>15 km</td>
<td>10.50</td>
<td>16.61</td>
<td>395.30</td>
</tr>
<tr>
<td><strong>1981, 15 km altitude</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>27 Jan.</td>
<td>14.74</td>
<td>20.6</td>
<td>515.14</td>
</tr>
<tr>
<td>25 Feb.</td>
<td>5.15</td>
<td>7.72</td>
<td>159.70</td>
</tr>
<tr>
<td>31 March</td>
<td>4.26</td>
<td>5.76</td>
<td>91.04</td>
</tr>
<tr>
<td>21 May</td>
<td>1.92</td>
<td>3.01</td>
<td>43.70</td>
</tr>
<tr>
<td>28 June</td>
<td>0.74</td>
<td>1.34</td>
<td>19.80</td>
</tr>
<tr>
<td>4 Sep.</td>
<td>1.07</td>
<td>1.81</td>
<td>28.63</td>
</tr>
</tbody>
</table>
FIG. 3. Vertical distribution of $^{134,137}\text{Cs}\ (\text{mBq/m}^3\ \text{STP})$ after the Chernobyl accident over eastern Poland.
15 km, the activity concentration of caesium was eight times higher than the minimum value at 6 km. On 1 May, the radioactivity of air near the ground dropped by a factor of 40 compared with that on the previous day, but at an altitude of 1 km it was four times higher than at the ground level.

The Chernobyl reactor accident reversed the earlier vertical distribution of fission products in the atmosphere: the highest concentrations were found near the ground and the lowest ones in the stratosphere (Fig. 4). The large scale vertical transport of air masses at altitudes of 850 hPa (~1500 m) and 500 hPa (~5500 m) indicated that in the first eight days of May there was an upward transport in the troposphere. This was the cause of the elevated radioactivity concentrations observed by us in the upper troposphere and the stratosphere in this period. On 7, 8 and 11 May, we observed slightly increased concentrations in stratospheric air, indicating an upward transport through the tropopause. Such transport was observed earlier for natural radionuclides and stable lead [2, 7]. The stratospheric ascent of the Chernobyl debris enabled its transport with the stratospheric air currents into the Southern Hemisphere. As discussed by Philippot [11], the $^{137}$Cs concentration peak found in near-surface snow at the South Pole in summer 1987–1988 [12] was due to the Chernobyl accident. Atmospheric dust containing $^{134}$Cs and $^{137}$Cs from the Chernobyl reactor reached latitudes of 17°S (Tahiti) and 21°S (La Réunion) nine months after the accident [11].

In April and May 1986, when the air at the ground level was highly contaminated, about 0.5% of the activity present between 0 and 3 km entered the stratosphere [8]. Between 1987 and 1994, the fraction of $^{134},^{137}$Cs present in the
ambient air at 15 km reached 58% to >100% of that at the ground level. This indicates the existence of strong resuspension of radioactive particulates, probably mostly from the contaminated regions of Ukraine, Belarus and Russia. This suggests that also other species of natural and man-made particulate contaminants, e.g. sea-salt chlorine or particulate hydrocarbons, can enter the stratosphere because of non-violent atmospheric processes. The resuspension effects and the atmospheric mass transport in the boundary layer are probably also responsible for elevated activity concentrations at 1 km altitude, which we observed in 16 out of 20 vertical concentration profiles of radiocaesium measured between 1987 and 1994. This effect is also responsible for the stratospheric residence time of radiocaesium, which, after the Chernobyl accident, was about three times longer (up to 770 d) than after previous nuclear explosions [8].

REFERENCES

ESTIMATION OF RADIOACTIVE CONTAMINATION OF WATER RESERVOIR SURFACES NEAR THE CHERNOBYL NUCLEAR POWER PLANT AFTER A POSSIBLE ACCIDENT AT THE SARCOPHAGUS

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Ukrainian Academy of Agricultural Sciences,
Kiev, Ukraine

Abstract

ESTIMATION OF RADIOACTIVE CONTAMINATION OF WATER RESERVOIR SURFACES NEAR THE CHERNOBYL NUCLEAR POWER PLANT AFTER A POSSIBLE ACCIDENT AT THE SARCOPHAGUS.

The paper presents integrated conservative estimates of the radioactive contamination of the water cooling pond of the Chernobyl nuclear power plant (NPP), the river Pripyat, the river Dnieper near Kiev, and the Kiev and Kanev reservoirs, due to direct deposition of aerosol particles from the atmosphere on water after possible accidents at the Sarcophagus. Two scenarios of possible accidents resulting in significant radioactive aerosol release from the building are considered: (1) total or partial collapse of the Sarcophagus walls (scenario “A”), and (2) collapse of some constructions inside the Sarcophagus (scenario “B”). The total activity of released dust has been estimated for each scenario. Simulations of atmospheric transport of radioactive aerosols have been carried out with the regional model LEDI (Lagrangian–Eulerian Diffusional model) for various meteorological conditions. The maximum values of radioactive contamination of the Dnieper basin caused by such accidents were found to be: $1.1 \times 10^{15}$ Bq (total activity), $5.2 \times 10^{14}$ Bq ($^{137}$Cs), $1.4 \times 10^{13}$ Bq ($^{239}$Pu and $^{240}$Pu) for scenario “A”, and $5 \times 10^{13}$ Bq (total), $2.6 \times 10^{13}$ Bq (caesium), $3.1 \times 10^{12}$ Bq (plutonium) for scenario “B”. The largest parts of radioactive aerosols were deposited on the water cooling pond of the Chernobyl NPP (up to 93%) and on the Kiev reservoir (up to 80%), depending on the meteorological conditions. Extremely high increases in water contamination were obtained in the simulations for plutonium. For scenario “A”, the contamination of the cooling pond and the Kiev reservoir may increase 12.7-fold and 3.9-fold, respectively, and for scenario “B”, 2.2-fold and 2.6-fold, compared with the present contamination caused by the 1986 accident. The $^{137}$Cs contamination of the cooling pond and of the Kiev and Kanev reservoirs will increase 3.5-fold, 2.3-fold and 1.5-fold, respectively, for scenario “A”; for scenario “B”, the $^{137}$Cs contamination will increase only by 9%.
1. INTRODUCTION

The Sarcophagus was constructed to retain the nuclear fuel remaining in the destroyed power generating Unit 4 of the Chernobyl nuclear power plant (NPP). Its construction, completed in November 1986, helped to solve urgent problems in the first period after the accident. In particular, the necessary protection of the plant premises and the environmental territory from penetrating radiation and radioactive aerosol releases was ensured [1]. However, because of the present condition of both the Sarcophagus building itself and the nuclear fuel in it, the possibility of accidents of various kinds cannot be excluded [2]. The most probable occurrence is the release of radioactive dust from inside the Sarcophagus into the atmosphere. This would result in additional radioactive contamination of the air and the surface of the territory around Chernobyl to great distances.

The paper presents integrated conservative estimates of the radioactive contamination of the water cooling pond of the Chernobyl NPP, the river Pripyat, the river Dnieper near Kiev and reservoirs at Kiev and Kanev for two scenarios of a possible accident at the Sarcophagus. The following estimates have been made: (1) estimation of the amount and the characteristics of radioactive dust inside the Sarcophagus; (2) estimation of possible radioactivity releases from the Sarcophagus into the atmosphere for two accident scenarios; (3) calculations of aerosol particle transport to distances of 200 km from the source, for various meteorological conditions; and (4) integrated estimates of the contamination of the Dnieper water using calculated concentrations of radioactivity deposited on the water surface.

2. CHARACTERISTICS OF RADIOACTIVE DUST INSIDE THE SARCOPHAGUS

The most difficult task of the present work was the estimation of the quantity and the characteristics of radioactive dust inside the Sarcophagus because of the uncertainty of available data or because of their absence. It is difficult to estimate the total mass of high activity aerosol particles inside the Sarcophagus, for certain reasons. It was reported in Ref. [2] that there are dozens of tonnes of radioactive dust inside the Sarcophagus. Also, the physical and chemical processes occurring in the fuel lava, weighing several hundred tonnes, lead to its destruction and to an increase in the quantity of fuel dust [3]. For further calculations, the mass of the radioactive dust was chosen as \( M_0 = 6 \times 10^4 \) kg.

According to Ref. [4], 25% of the released activity is connected with large fuel particles, of more than 10-15 \( \mu \)m, and with an average density of \( 10^3 \) kg/m\(^3\). Measurement data of the specific activity of fuel and aerosol particles are presented in Table I.
TABLE I. MEASUREMENT DATA OF THE SPECIFIC ACTIVITY (MBq/g) OF LARGE FUEL PARTICLES ($A_i^L$) AND OF THE HEAVY FRACTION OF SMALL AEROSOL PARTICLES ($A_i^H$) INSIDE THE SARCOPHAGUS (November–December 1988)\(^a\)

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>$A_i^L$</th>
<th>$A_i^H$</th>
<th>Nuclide</th>
<th>$A_i^L$</th>
<th>$A_i^H$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co-60</td>
<td>0.38</td>
<td></td>
<td>Eu-155</td>
<td>38</td>
<td>5.3</td>
</tr>
<tr>
<td>Nb-95</td>
<td>4.8</td>
<td></td>
<td>Sr-90</td>
<td>880</td>
<td></td>
</tr>
<tr>
<td>Ru-106</td>
<td>1000</td>
<td>170</td>
<td>Pu-239</td>
<td>15</td>
<td>6.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Pu-240</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sb-125</td>
<td>65</td>
<td>5.9</td>
<td>Pu-238</td>
<td>6.9</td>
<td>3.1</td>
</tr>
<tr>
<td>Cs-134</td>
<td>230</td>
<td>6.2</td>
<td>Am-241</td>
<td>5.0</td>
<td>1.5</td>
</tr>
<tr>
<td>Cs-137</td>
<td>920</td>
<td>29</td>
<td>Cm-244</td>
<td>1.5</td>
<td>0.29</td>
</tr>
<tr>
<td>Ce-144</td>
<td>2500</td>
<td>620</td>
<td>Cm-242</td>
<td>3.8</td>
<td>1.1</td>
</tr>
</tbody>
</table>

\(^a\) From Ref. [4].

The air pollution inside the Sarcophagus is due mainly to light ($\rho < 4 \times 10^3$ kg/m\(^3\)) and heavy ($\rho = 8 \times 10^3$ kg/m\(^3\)) fractions of small aerosol particles of a size of a few micrometres. Data on the specific activities of the heavy fraction of aerosol particles, taken from Ref. [4], are presented in Table I. Unfortunately, we did not have any information about the specific activity of the light fraction of aerosol particles. Thus, it was assumed in subsequent calculations that, by the end of 1988, 25% of the activity was determined by the contribution of large (20 \(\mu\)m) particles and 75% by that of the heavy fraction of small (2 \(\mu\)m) particles. This condition is satisfied if the total masses of small and large particles are equal to $5.7 \times 10^4$ kg and $3 \times 10^3$ kg, respectively. By taking into account the total specific activities of small and large particles, equal to $8.5 \times 10^8$ Bq/g and $56.7 \times 10^8$ Bq/g, respectively (calculated with the data given in Table I), it is possible to estimate the values of the total radioactivity of each fraction as a value of the total radioactivity in the Sarcophagus. Using these data, the decrease in the activity of the dust caused by radioactive decay of some isotopes (mainly Ru, Ce, Eu) by the end of 1994 and 1999, was calculated (Table II). Similar estimations of the activities of $^{137}$Cs (in 1994) and $^{239,240}$Pu are also given in Table II.
TABLE II. CALCULATED VALUES OF THE ACTIVITY (10^{15} Bq) OF SMALL AND LARGE FUEL PARTICLES IN THE SARCOPHAGUS FOR A TOTAL MASS OF DUST OF 6 \times 10^4 kg

<table>
<thead>
<tr>
<th>Particle size</th>
<th>Total activity</th>
<th>$^{137}$Cs</th>
<th>$^{239,240}$Pu</th>
</tr>
</thead>
<tbody>
<tr>
<td>2 \mu m</td>
<td>49</td>
<td>2.5</td>
<td>1.9</td>
</tr>
<tr>
<td>20 \mu m</td>
<td>17</td>
<td>5.0</td>
<td>4.3</td>
</tr>
<tr>
<td>Total</td>
<td>66</td>
<td>7.5</td>
<td>6.2</td>
</tr>
</tbody>
</table>

3. SCENARIOS OF POSSIBLE ACCIDENTS AT THE SARCOPHAGUS

Two scenarios of possible accidents at the Sarcophagus that would result in significant radioactive aerosol release from the building are considered.

(1) Total or partial collapse of the Sarcophagus walls caused by natural ageing of the building on one hand and adverse external events (e.g. strong earthquake) on the other hand (scenario "A"). It is assumed that practically all radioactive dust inside the Sarcophagus may be involved in atmospheric transport.

(2) Collapse of some constructions inside the Sarcophagus (scenario "B"). In particular, the apprehension exists that the unstable ferroconcrete constructions of the top part of the destroyed unit and the top cover of the reactor, weighing over $2 \times 10^6$ kg [2], may collapse. This could result in a considerable increase (by several orders of magnitude) in the aerosol concentration in the air inside the Sarcophagus. Because of the non-airtightness of the Sarcophagus (the total area of holes in the roof and walls has been estimated at 1000 $m^2$ [2]) and the existence of a natural exhaust ventilation through the ventilation stack of the 3rd and 4th power generating units, a significant part of the radioactivity will be released into the atmosphere.

After an accident inside the Sarcophagus, aerosol particles will settle on the floor or will be carried by air flow into the atmosphere through the ventilation stack and through holes in the walls and roof. The change of the mass of airborne aerosols inside the Sarcophagus under the influence of these processes is described by the following equation:

$$\frac{dm}{dt} = - \frac{W}{L} m - \frac{Q}{V} m$$

(1)
where \( W \) is the particle sedimentation velocity, \( Q \) is the total air flow rate through the stack and holes, \( L = 60 \text{ m} \) is the height of the Sarcophagus and \( V \) is its volume. The Sarcophagus is assumed to be a cube, with 60 m long sides; thus, \( V = 2.2 \times 10^6 \text{ m}^3 \).

The total mass of aerosol particles carried by air flow into the atmosphere is

\[
M = M_0 (1 + t_s/t_R)^{-1}
\]

where \( t_s = L/W \) is the characteristic particle sedimentation time and \( t_R = V/Q \) is the characteristic time of aerosol release from the Sarcophagus.

According to Ref. [5], in 1989 the radioactivity release through the ventilation stack was \( 3.0 \times 10^9 \text{ Bq} \) and the total radioactivity release from the Sarcophagus was \( 7.4 \times 10^9 \text{ Bq} \), i.e. the radioactivity release from the Sarcophagus building was 1.5 times larger than that from the stack. The air flow rate through the ventilation stack is \( 3.9 \text{ m}^3/\text{s} \). Assuming that the air flow rate through holes in the walls is half as large, the total air flow rate is \( 9.8 \text{ m}^3/\text{s} \). The sedimentation velocities of 20 \( \mu \text{m} \) and 2 \( \mu \text{m} \) particles are 0.12 m/s and \( 9.6 \times 10^{-4} \text{ m/s} \), respectively. It follows from Eq. (2) that, owing to collapse of constructions inside the Sarcophagus and subsequent transport of dust into the air, 2.2% of the large particles and 73.8% of the small particles will be carried into the atmosphere. Taking into account their contributions to total activity, the absolute activity values obtained are \( 3.7 \times 10^{14} \text{ Bq} \) and \( 3.6 \times 10^{16} \text{ Bq} \) (in 1988), \( 1.1 \times 10^{14} \text{ Bq} \) and \( 1.8 \times 10^{15} \text{ Bq} \) (in 1994) and \( 9.5 \times 10^{13} \text{ Bq} \) and \( 1.4 \times 10^{15} \text{ Bq} \) (in 1999), respectively. Analogous values (1994) are \( 5.3 \times 10^{13} \text{ Bq} \) and \( 1.1 \times 10^{15} \text{ Bq} \) for \(^{137}\text{Cs} \), and \( 9.5 \times 10^{11} \text{ Bq} \) and \( 2.7 \times 10^{14} \text{ Bq} \) for plutonium. Thus, the possible total activity release from dust in the Sarcophagus decreases from 56\% to 26\% during the first six years and to 24\% during the subsequent five years. The share of large fuel particles contributing to the release activity increases from 1\% to 7\%.

4. MODELLING OF CONTAMINATION WITH THE AID OF THE LEDI MODEL

For modelling of atmospheric transport and deposition of radioactive aerosol particles, the regional diffusional model LEDI (Lagrangian–Eulerian Diffusional model) [6] has been used. The model is based on a combination of Lagrangian and Eulerian approaches and has been applied in the simulation of the pollutant dispersion in the atmosphere. The model makes it possible to calculate the transport of pollutants released from a time dependent source in inhomogeneous and time dependent meteorological conditions, for horizontal scales of the order of tens and hundreds of kilometres, and requires relatively little computation time. The model enables calculation of air pollutant concentration and surface deposits on non-
uniform underlying surfaces. The model takes into account turbulent diffusion of pollutants, gravitational sedimentation and dry deposition on underlying surfaces.

For the case of total destruction of the Sarcophagus (scenario "A") a point source of release is considered, with an effective height $h_{ef} = 60$ m, corresponding to the height of the Sarcophagus. The initial size of the aerosol cloud has been neglected.

The amount of activity deposition on the Dnieper and on reservoirs is determined by the principal direction of transport. The maximum contamination of these reservoirs will be in north-western wind conditions, i.e. in the direction of the river Pripyat, the Kiev and Kanew reservoirs and a part of the Dnieper near Kiev. Transport calculations have been carried out for three wind directions (Fig. 1):

- **Variant (a) — 308°**: transport is along the water cooling pond of the Chernobyl NPP and the river Pripyat;
- **Variant (b) — 319°**: transport is in the direction of the central part of the Kiev reservoir;
- **Variant (c) — 331°**: transport is in the direction of the southern part of the Kiev reservoir and the central part of the Kanew reservoir.

A similar set of input meteorological data was considered for scenario "B". It was assumed that 40% of the total release is carried through the ventilation stack and the rest through holes in the walls and roof of the Sarcophagus. In this case, the source of aerosol release was assumed to be a superposition of two point sources with effective heights of $h_{ef} = 60$ m and $h_{eff} = 120$ m (ventilation stack height).

To investigate the influence of the thermal regime of the atmospheric boundary layer on the value of the surface deposit for each of the six accident variants, calculations were carried out for three typical vertical profiles of wind and temperature, corresponding to neutral, stable and unstable boundary layers [7].

The value of the dry deposition velocity was set at 0.002 m/s on the ground and at $5 \times 10^{-4}$ m/s on water for 2 µm particles, and was taken to be equal to the sedimentation velocity of 0.12 m/s for 20 µm particles. The value of the surface roughness of the ground is 0.1 m; for water it was calculated according to Charnock's formula.

5. **CALCULATIONS OF DEPOSITION**

Integrated calculations of the contamination of objects in the Dnieper basin caused by direct deposition of aerosols on water were performed for each of the 18 variants of the accident and meteorological conditions. Estimated maximum values of radioactive contamination of the NPP cooling pond, the rivers Pripyat and Dnieper and the reservoirs are given in Table III. For variant (a), the deposit activity was $1.1 \times 10^{15}$ Bq in 1994 (including $5.2 \times 10^{14}$ Bq of $^{137}$Cs and $1.4 \times 10^{13}$ Bq
of Pu) and will decrease to $9.2 \times 10^{14}$ Bq by 1999. The cooling pond of the NPP was the most contaminated object in this case. The contribution of 20 $\mu$m particles to the pond contamination increased from 94.6% in 1988 to 99.1% in 1994. For variants (b) and (c), the most contaminated objects are the Kiev and Kanev reservoirs (up to 80% of the total deposit for the former and up to 11% for the latter). The deposited activity is determined mainly by large particles, except for Pu.

The variations in the atmospheric stability result in changes of the deposition value of up to one order of magnitude. The maximum deposition on the pond is under neutral stratification and that on the river Pripyat and on the reservoirs is under daytime unstable stratification.
TABLE III. ESTIMATED MAXIMUM VALUES OF RADIOACTIVE CONTAMINATION (10^{12} Bq) OF THE NPP COOLING POND, THE RIVERS PRIPYAT AND DNIEPER AND THE RESERVOIRS

<table>
<thead>
<tr>
<th>Scenario “A”</th>
<th></th>
<th></th>
<th></th>
<th></th>
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<th></th>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>1988</td>
<td>1994</td>
<td>1999</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Cooling pond</td>
<td>3100 (1)</td>
<td>880 (1)</td>
<td>760</td>
<td>(1)</td>
<td>430</td>
<td>(1)</td>
</tr>
<tr>
<td>Pripyat river</td>
<td>840 (2)</td>
<td>240 (2)</td>
<td>210</td>
<td>(1)</td>
<td>120</td>
<td>(2)</td>
</tr>
<tr>
<td>Kiev reservoir</td>
<td>970 (4)</td>
<td>280 (4)</td>
<td>240</td>
<td>(4)</td>
<td>130</td>
<td>(4)</td>
</tr>
<tr>
<td>Dnieper river</td>
<td>27 (6)</td>
<td>7.3 (6)</td>
<td>6.3</td>
<td>(6)</td>
<td>3.5</td>
<td>(6)</td>
</tr>
<tr>
<td>Kanev reservoir</td>
<td>58 (6)</td>
<td>14 (6)</td>
<td>12</td>
<td>(6)</td>
<td>7.0</td>
<td>(6)</td>
</tr>
<tr>
<td>Total</td>
<td>4100 (1)</td>
<td>1100 (1)</td>
<td>920</td>
<td>(1)</td>
<td>520</td>
<td>(1)</td>
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</table>

<table>
<thead>
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<th>Scenario “B”</th>
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<tr>
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<td>1988</td>
<td>1994</td>
<td>1999</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cooling pond</td>
<td>190 (1)</td>
<td>31 (1)</td>
<td>26</td>
<td>(1)</td>
<td>15</td>
<td>(1)</td>
</tr>
<tr>
<td>Pripyat river</td>
<td>170 (1)</td>
<td>13 (1)</td>
<td>10</td>
<td>(1)</td>
<td>7.0</td>
<td>(1)</td>
</tr>
<tr>
<td>Kiev reservoir</td>
<td>310 (3)</td>
<td>16 (3)</td>
<td>12</td>
<td>(3)</td>
<td>9.3</td>
<td>(3)</td>
</tr>
<tr>
<td>Dnieper river</td>
<td>11 (5)</td>
<td>0.58 (5)</td>
<td>0.44</td>
<td>(5)</td>
<td>0.34</td>
<td>(5)</td>
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<tr>
<td>Kanev reservoir</td>
<td>31 (5)</td>
<td>1.6 (5)</td>
<td>1.2</td>
<td>(5)</td>
<td>0.93</td>
<td>(5)</td>
</tr>
<tr>
<td>Total</td>
<td>480 (1)</td>
<td>50 (1)</td>
<td>41</td>
<td>(1)</td>
<td>26</td>
<td>(1)</td>
</tr>
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</table>

<table>
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<tr>
<th>Total activity a</th>
<th>137Cs</th>
<th>239,240Pu</th>
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<tr>
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<td>9.1</td>
</tr>
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<td>3.1</td>
</tr>
<tr>
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<td>130</td>
<td>4.4</td>
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<tr>
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<td>14</td>
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<td>Pripyat river</td>
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<tr>
<td>Dnieper river</td>
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<td>0.086</td>
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<tr>
<td>Kanev reservoir</td>
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<td>0.24</td>
</tr>
<tr>
<td>Total</td>
<td>26</td>
<td>3.1</td>
</tr>
</tbody>
</table>

* Meteorological conditions of transport:
1. - Wind direction 308°, neutral stratification;
2. - Wind direction 308°, unstable stratification;
3. - Wind direction 319°, stable stratification;
4. - Wind direction 319°, unstable stratification;
5. - Wind direction 332°, stable stratification;
6. - Wind direction 332°, unstable stratification.
For scenario "B", the deposition values decrease considerably (up to twentyfold) for both the total activity and the $^{137}$Cs activity compared with the previous values; this is due to the relatively small release of large fuel particles from the Sarcophagus. In this case, small particles generally determine the deposited activity. Because of the relatively high specific activity of small particles of $^{239,240}$Pu compared with large particles, Pu deposition decreases only four to five times in this case compared with that of scenario "A".

6. SUMMARY

Possible accidents at the Sarcophagus may result in the release of large amounts of radioactive dust into the atmosphere and lead to considerable contamination of water in the Dnieper basin. In the case of a total collapse of the Sarcophagus, the Cs contamination of the NPP cooling pond, the Kiev reservoir and the Kanev reservoir may increase 3.5-fold, 2.3-fold and 1.5-fold, respectively (at present, the total amount of $^{137}$Cs is $1.7 \times 10^{14}$ Bq in the NPP pond, $10^{14}$ Bq in the Kiev reservoir and $1.3 \times 10^{15}$ Bq in the Kanev reservoir [8]). Especially large increases in contamination have been obtained for Pu: the contamination of the NPP pond and of the Kiev reservoir will increase 12.7-fold and 3.9-fold, respectively.

For scenario "B", the Cs contamination of all mentioned objects will increase by only 8–9%, but there is still the possibility of high additional Pu contamination in this case. The contamination of the NPP pond and that of the Kiev reservoir will increase 2.2-fold and 2.6-fold, respectively.

If the contamination of the flood-plains of Pripyat and Dnieper and the wash-off of activity into the water would be considered, much higher estimated values would probably be obtained.

REFERENCES


INVESTIGATIONS INTO THE EMISSIONS OF POLONIUM-210 FROM A TIN SMELTER

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Abstract

INVESTIGATIONS INTO THE EMISSIONS OF POLONIUM-210 FROM A TIN SMELTER.

Polonium-210 was found to be separated during processing of mineral ore concentrates, metal refinery residues and scrap at a tin smelter. Up to 80,000 t of feed materials were processed per annum, with the annual input of $^{210}\text{Po}$ estimated at about 87 GBq. Substantial proportions of the $^{210}\text{Po}$ either decayed during processing or were isolated within a tellurium dross by-product. About 65 MBq $^{210}\text{Po}$ in process off-gases were discharged to the atmosphere per week. The concentrations of $^{210}\text{Po}$ in air, rainwater and school playground dust, measured at various distances from the smelter, were indistinguishable from reported background values for urban areas. The total annual radiation exposures for members of the public, using pessimistic worst case assumptions, were estimated to have been less than one microsievert.

1. INTRODUCTION

Processing of mineral ores containing elevated levels of naturally occurring radionuclides can result in technologically enhanced concentrations of these radionuclides being present in wastes from certain industrial facilities. Where these concentrations exceed the statutory exemption levels in the United Kingdom, the processes are subject to regulation. One such facility was a tin smelter on the east coast of England where the recovery of tin from ore concentrates, metal refinery residues and scrap resulted in $^{210}\text{Po}$ being separated during processing and being emitted to the atmosphere at concentrations marginally above the relevant exemption level of 0.22 mBq/g for gaseous wastes. The smelter closed down for commercial reasons in 1991.

It had been observed that in certain villages downwind of the smelter there was an apparent cluster of childhood leukaemias. Local and national media alleged a link between this apparent leukaemia cluster and atmospheric discharges from the smelter. In response to the anxiety of the local population arising from these allegations, the regulatory authority, Her Majesty’s Inspectorate of Pollution (HMIP), carried out investigations into the processes employed at the smelter, the levels of $^{210}\text{Po}$ in the environment and their radiological impact.
2. UK STATUTORY CONTROLS

The use of radioactive substances and the disposal of radioactive wastes are subject to the legal provisions of the Radioactive Substances Act and related Exemption Orders. This Act is concerned primarily with the protection of the general public from hazards which may arise from the disposal of radioactive wastes to the environment. The Act is administered and enforced in England and Wales by HMIP. Control is exercised by the granting of registration and authorization certificates, which place limits and conditions on the keeping and use of radioactive substances and on the storage and disposal of radioactive wastes, respectively. Visits are made to premises by inspectors to check that users are complying with the limits and conditions specified in their certificates.

The Radioactive Substances Act distinguishes between man-made radioactivity, such as occurs in the nuclear power industry, and radioactivity of natural origin. In recognition of the ubiquitous nature of natural radioactivity, solid and liquid materials containing certain specified elements, including uranium, thorium and polonium, at concentrations of less than 15 Bq/g are exempt from regulation. The concentrations at which gaseous materials are subject to the provisions of the Act vary, depending on the radioactive element. The limit for polonium corresponds to 0.22 mBq/g.

3. PROCESS INVESTIGATIONS

The processes employed for the recovery of tin, lead and other metals were extremely complex and involved considerable recycling of materials. The simplified flowsheet in Fig. 1 identifies the principal process stages and the waste and product arisings. Samples of feed materials, of solid and liquid wastes and of filter pads used to monitor emissions via the main stack to the atmosphere were analysed.

Up to 80 000 t of feed materials were processed per annum. They consisted of a varied mixture of mineral ores, secondary residues, concentrates and metal scrap. For many of these feed materials the average levels of uranium, thorium, radium and $^{210}$Po were well below the regulatory limit of 15 Bq/g. Only a few supplies exceeded this value. The annual input of $^{210}$Po into the tin smelter was estimated to be about 87 GBq.

The fate of $^{210}$Po during processing is illustrated in Fig. 2. Substantial proportions of the $^{210}$Po present in feed materials either decayed within the plant or were isolated within tellurium dross, which was stored until the $^{210}$Po had decayed to insignificant levels. Most of the decay within the plant occurred in fume trapped by the electrostatic precipitator and recycled to the sinter plant. About 19% of the $^{210}$Po in feed materials was discarded at low concentrations in large quantities of
FIG. 1. Simplified flowsheet.
slags arising from the blast furnaces. Polonium-210 was not detected in liquid wastes and was detected only at trace concentrations in product metals.

The $^{210}$Po present in molten unrefined lead and in lead/bismuth alloy at concentrations of up to 30 and 60 Bq/g, respectively, was effectively reduced by a process used to remove tellurium impurity. A by-product of tellurium dross was skimmed from the surface of molten metals and transferred to metal drums for storage. About 24 t of dross were produced annually, containing 27 GBq $^{210}$Po at typical concentrations of 1100 Bq/g.

About 500 000 m$^3$ of air were emitted to the atmosphere every hour via the main, 180 m tall, stack. This air contained off-gases from the main process stages. As illustrated in Fig. 3, the major contribution of $^{210}$Po was found to be a small proportion of fume from the sinter plant which was not recovered by the electrostatic precipitator. An average weekly emission of 65 MBq was calculated.
4. ENVIRONMENTAL MONITORING

Measurements were made of the concentrations of $^{210}$Po in air, rainwater and dust from school playgrounds, and of the deposition rates at various distances from the smelter. The results were compared with literature values for natural background as reported by Parfenov [1].

The concentrations of $^{210}$Po in air were indistinguishable from natural background levels at about 0.0001 Bq/m$^3$. The concentrations in rainwater at about 0.09 Bq/L were just above the reported literature values. The concentrations in playground dust were indistinguishable from natural background levels for soils of 0.008–0.22 Bq/g.

FIG. 3. Contributions of $^{210}$Po to the stack discharge.
5. RADILOGICAL IMPACT

The radiological significance of potential emissions of \(^{210}\text{Po}\) via the main stack to the atmosphere at the authorized limit of 592 MBq per week, taking into account the influence of the prevailing wind from the west, was assessed by considering four principal pathways.

5.1. Direct inhalation of the plume

A maximum integrated air concentration of 210 Bq·m\(^{-3}\)·s\(^{-1}\) per year was calculated using the methodology detailed in Ref. [2]. This integrated air concentration was equivalent to an average concentration of \(^{210}\text{Po}\) in air of \(6.7 \times 10^{-6}\) Bq/m\(^3\). This maximum concentration, assuming a uniform windrose, was at a distance of 3 km from the stack. On the assumption that a member of the public inhaled air at this maximum ground level concentration for 24 hours per day and 365 days per year at average breathing rates and allowing for the influence of the prevailing westerly wind, the annual radiation exposures for an adult, a ten year old child and an infant would have been 0.18, 0.27 and 0.21 Sv, respectively.

5.2. Deposition from the plume onto the ground

As the plume passed over the ground, \(^{210}\text{Po}\) associated with dust particles would have been deposited under both dry and wet conditions. The magnitude of this deposition was estimated, using the methodology detailed in Ref. [3], at a rate of \(3.6 \times 10^{-8}\) Bq·m\(^{-2}\)·s\(^{-1}\). Constant deposition would have tended to build up the surface deposit of \(^{210}\text{Po}\), but this would have been balanced by radioactive decay so that a dynamic equilibrium would have been established. The equilibrium surface deposit would have been 0.6 Bq/m\(^2\). Other environmental mechanisms, such as wash-out and migration into the soil sink, would have acted to reduce this level further. Because \(^{210}\text{Po}\) does not emit any substantial gamma or X rays, external radiation exposure of persons from deposited activity would have been negligible.

5.3. Inhalation of resuspended activity

Although deposited \(^{210}\text{Po}\) posed no external radiation hazard, it may have been transferred back into the air by resuspension. The concentration of \(^{210}\text{Po}\) in air as a result of such resuspension was calculated to have been \(6 \times 10^{-7}\) Bq/m\(^3\). Using the same assumptions as detailed in Section 5.1, the annual radiation exposures for an adult, a ten year old child and an infant would have been 0.02, 0.03 and 0.02 \(\mu\)Sv, respectively.
5.4. Ingestion of contaminated foodstuffs

Polonium-210 might have entered the food-chain as a result of deposition on the leaves of plants eaten by man or animals, or by root uptake from activity deposited in soil. The extent to which this was likely to have occurred was estimated in accordance with the methodology detailed in Ref. [4]. It was assumed that at least 24% of the food eaten by the public was grown locally; in order to evaluate the effects of this food, published data on consumption rates were used. The annual radiation exposures by this pathway, allowing for the prevailing westerly wind, for an adult, a ten year old child and an infant were estimated to have been 0.26, 0.46 and 0.53 μSv, respectively.

Consequently, the total annual radiation exposures for an adult, a ten year old child and an infant, using pessimistic worst case assumptions for both the inhalation and ingestion pathways, were estimated to have been 0.46, 0.76 and 0.76 μSv, respectively.

Radiation of this magnitude was put into context by comparing it with levels to which members of the public are exposed in their daily lives. Radiation from all natural background sources amounts to about 2500 pSv per year and, because 210Po is present naturally in diet, a direct comparison may be made with this source, which contributes about 25 μSv per year. Polonium-210 is also present at enhanced levels in tobacco and is inhaled into the lungs of smokers where it increases their natural radiation dose by up to 100 μSv per year [5].

6. CONCLUSION

The Inspectorate concluded that there was no evidence from environmental monitoring data, predicted dispersion patterns and radiological assessments to link the reported childhood leukaemia excesses with emissions of 210Po from the smelter to the atmosphere.

REFERENCES


ENVIRONMENTAL IMPACT ASSESSMENT OF COAL FIRED POWER PLANTS AND NUCLEAR POWER PLANTS IN INDIA

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Trombay, Bombay, India

Abstract

ENVIRONMENTAL IMPACT ASSESSMENT OF COAL FIRED POWER PLANTS AND NUCLEAR POWER PLANTS IN INDIA.

Coal plays an increasingly important role in meeting the energy needs of India. One of the environmental problems associated with the use of coal as a fuel for Indian thermal power plants is the generation of very large amounts of fly ash. Coal contains trace quantities of the naturally occurring primordial radionuclides and so coal burning is one of the sources of technologically enhanced exposure of man from natural radionuclides. The concentrations of these radionuclides are usually very low. However, when coal is burnt in thermal power plants the fly ash emitted through the stack becomes enriched in some radionuclides; hence, combustion of coal on a large scale for thermal power generation has an important impact on the environment. Coal combustion accounted for about 65% of the installed capacity for power generation in 1994. Many of the thermal power plants in India are situated in densely populated areas. The major coal fields in India are situated in the densely populated eastern parts of the country. The natural radioactivity content in samples from these coal fields is found to be higher than that from other coal fields. The Bhabha Atomic Research Centre has carried out extensive studies of coal and fly ash samples collected from more than 30 thermal power plants with an installed capacity of 10 000 MW(e), spread all over the country, to determine their natural radioactivity content. The radiation doses to the population residing within a radius of 88.5 km (50 miles) from such plants have also been computed. The collective effective dose commitments from these plants are 206 man-Sv/a for bones, lungs and thyroid and 73 man-Sv/a for the whole body. The dose commitments to the population living within a radius of 88.5 km from coal fired plants and nuclear power plants in India have also been computed and the estimated doses from these two types of energy production have been compared.

1. INTRODUCTION

Coal is the most abundant resource for electricity generation in India. At present, coal is used as fuel for about 65% of the total electricity generated in the country. However, Indian coal used for electricity generation is characterized by a very high ash content (up to 55%). This results in large amounts of residual fly ash and large emissions to the environment, not only of gaseous pollutants but also of
particulates. Several thermal power plants do not have fly ash control equipment, such as electrostatic precipitators, and even those which have such equipment are not able to control atmospheric emissions to the extent desirable because of the high ash content of the coal.

Since all kinds of coal contain natural radioactivity of primordial origin, which becomes concentrated in fly ash emitted from the stacks of power stations, thermal power plants utilizing coal have an adverse environmental impact due to radioactivity releases in addition to releases of gaseous and particulate pollutants. Extensive studies on the radioactivity content of coal and fly ash have been carried out in India, and the results of these studies are briefly summarized. Utilization of ash from coal burning is considered mainly for three applications: cement manufacture, production of bricks for building houses, and mixing of ash with asphalt for road construction. The largest possible application is for brick production. Because of the higher levels of natural radioactivity in fly ash, houses built with such bricks have higher indoor radon levels. However, this additional exposure is low — a fraction of the exposure from natural radioactivity — and it is possible to minimize it by the application of appropriate technology.

2. RADIONUCLIDE RELEASE

The concentration of radionuclides in Indian coal varies widely from seam to seam, as well as horizontally and vertically in a given seam. Samples of coal from several collieries from all over India and from different thermal power plants have been collected and analysed by gamma spectrometry for their natural radioactivity content. Coal (in pulverized form) and fly ash samples were stored in airtight cylindrical plastic containers (6.5 cm diameter and 7.5 cm height) for one month in order to ensure that the radionuclides $^{226}$Ra and $^{228}$Th attain radioactive equilibrium with their daughters. An analysis of the activity levels of $^{226}$Ra, $^{228}$Th and $^{40}$K was carried out, using the 1.76 MeV peak from $^{214}$Bi, the 2.62 MeV peak from $^{208}$Tl and the 1.46 MeV peak from $^{40}$K [1, 2]. All of these samples were counted for about 500–1000 minutes, depending on the concentrations, using a low background, 12.5 cm × 10.0 cm NaI(Tl), gamma spectrometer. The counting time depended on the radioactivity levels in the samples, and the results were reported only when they were above three standard deviations from the background. The estimated concentration of $^{238}$U varied from 0.3 to 14.1 ppm, with a mean of 3.1 ppm. The average value is within the range reported in Ref. [3]. Table I gives the estimated uranium, thorium, radium and potassium contents of Indian coal.

The principal radionuclide released during coal mining is radon. Radon is released when the coal seam is exposed and also when coal is crushed. However, the release of radon during mining does not constitute a significant environmental hazard [4, 5]. Coal also contains extraneous rock matter, which constitutes about
### TABLE I. ESTIMATED URANIUM, THORIUM, RADIUM AND POTASSIUM IN INDIAN COAL

<table>
<thead>
<tr>
<th>Region</th>
<th>(^{238}\text{U}) (ppm)</th>
<th>(^{232}\text{Th}) (ppm)</th>
<th>(^{226}\text{Ra}) (ppm)</th>
<th>(^{228}\text{Ra}) (Bq/kg)</th>
<th>(^{40}\text{K}) (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Coal deposits</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Western coal fields</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Madhya Pradesh</td>
<td>1.8</td>
<td>10.0</td>
<td>22.2</td>
<td>40.7</td>
<td>103.6</td>
</tr>
<tr>
<td>Maharashtra</td>
<td>1.2</td>
<td>6.4</td>
<td>14.8</td>
<td>70.0</td>
<td>62.8</td>
</tr>
<tr>
<td><strong>Central coal fields</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bihar</td>
<td>2.6</td>
<td>12.7</td>
<td>33.3</td>
<td>51.8</td>
<td>155.4</td>
</tr>
<tr>
<td>Madhya Pradesh</td>
<td>2.1</td>
<td>2.7</td>
<td>25.9</td>
<td>11.1</td>
<td>155.7</td>
</tr>
<tr>
<td>Uttar Pradesh</td>
<td>2.1</td>
<td>15.5</td>
<td>25.9</td>
<td>22.2</td>
<td>144.1</td>
</tr>
<tr>
<td>Orissa</td>
<td>2.1</td>
<td>2.7</td>
<td>25.9</td>
<td>18.5</td>
<td>122.0</td>
</tr>
<tr>
<td><strong>Eastern coal fields</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>West Bengal</td>
<td>3.2</td>
<td>12.0</td>
<td>40.7</td>
<td>51.8</td>
<td>180.0</td>
</tr>
<tr>
<td><strong>North-eastern coal fields</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Meghalaya</td>
<td>1.4</td>
<td>10.0</td>
<td>14.8</td>
<td>44.4</td>
<td>90.6</td>
</tr>
<tr>
<td><strong>Lignite deposits</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Tamil Nadu</td>
<td>6.5</td>
<td>10.9</td>
<td>88.4</td>
<td>44.4</td>
<td>170.0</td>
</tr>
<tr>
<td>Gujarat</td>
<td>6.0</td>
<td>10.8</td>
<td>81.4</td>
<td>55.5</td>
<td>190.0</td>
</tr>
</tbody>
</table>

10–15% of the raw coal, before it is delivered to a power plant. This discarded rock and mineral matter as well as other coal mining refuse are the major constituents of the dumps that are scattered over wide stretches of land in coal mining areas. Weathering and leaching from coal refuse dumps produce water contaminants, such as silt, acids and dissolved mineral matter.

It is estimated that about 2000 coal waste dumps discharge effluents to nearby streams or water bodies. Typically, 1–2 kg of acid and 0.5–0.7 kg of soluble iron per hectare of refuse dump are being discharged per day. Uranium, thorium, radium, lead and polonium are leached as easily as other non-radioactive minerals and the impact on surface water and groundwater could be significant. In one of the studies, the concentration has been found to be 3.1 ppm of uranium and \(^{226}\text{Ra}\) [3]. Depending upon the configuration of the piles, the background radiation levels may or may
not be significantly enhanced. Since no field data are available, this cannot be accurately assessed. After recovery of suspended solids from waste water, significant amounts of dissolved radionuclides may still be present in the processed water. Furthermore, the processing of mined coal before transport to a power plant also involves a wide variety of relatively new technologies, such as liquefaction and gasification. It has been reported in the literature that neither the products from gasification nor those from liquefaction contain significant concentrations of radionuclides [6].

Coal fired power plants maintain stockpiles of coal to ensure continuous supply. The amount of coal thus stored depends upon the generating capacity of the power plant, seasonal changes in power demand and uncertainties regarding future supplies. Weathering and leaching of radionuclides from on-site coal may lead to contamination of groundwater and surface water, and emanation of radon from coal may increase the concentration of radon in the atmosphere. This aspect has not been studied in detail in India.

Combustion of coal releases radionuclides, which may enter the environment through the stack of a power plant or remain in the bottom ash or slag. The noble gas $^{222}$Rn is emitted from the stack following combustion of coal, and the other radionuclides are partitioned between the bottom ash/fly ash and flue gases. Depending upon the collection efficiencies of emission control devices, fractions of fly ash of various sizes may be emitted by the stack.

Table II gives the estimated maximum releases for a model 1000 MW(e) power plant operating in India. The estimated doses are quite small compared to the natural background of about 2.4 mSv/a. For dose estimation, the methods outlined

<table>
<thead>
<tr>
<th>Dose commitments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Individual (mSv/a)</td>
</tr>
<tr>
<td>Whole body</td>
</tr>
<tr>
<td>Bone</td>
</tr>
<tr>
<td>Lung</td>
</tr>
<tr>
<td>Thyroid</td>
</tr>
<tr>
<td>Kidney</td>
</tr>
</tbody>
</table>
by McBride et al. [6] have been applied, and the necessary corrections for the population density around an operating power plant, the coal consumption per day per MW(e), the ash content of the coal used and the average plant load factor have been incorporated.

Another area of radiological concern is the potential contamination of groundwater and surface water by radionuclides leached from ash ponds. On the basis of values reported in the literature, it is found that, even though a pathway exists for transport of radionuclides to water, there is no significant transfer of radionuclides to underground water bodies. This is because the leachability of the ash is quite small. Further studies are needed to substantiate this observation. The low leaching rate for fly ash is attributed to its chemical composition.

Operation of coal based thermal power plants has resulted in changes of trace elements and of the radionuclide contents of the soil in the vicinity of these plants. The studies carried out by our laboratory have shown that there is a definite increase in the levels of $^{226}$Ra and $^{228}$Th in soils collected in the neighbourhood of thermal power plants compared to the range of these isotopes in soil samples collected in areas that are not affected by power plants. The concentrations of $^{226}$Ra and $^{228}$Th in soils near thermal power plants have been found to increase with (a) increasing plant age, (b) increasing installed plant capacity, (c) inadequacy of fly ash emission control methods, and (d) higher radioactivity content of the coal and fly ash fed to the power plant boilers [7]. An additional pathway for exposure of the public is incorporation of fly ash into various products for house building. Fly ash enriched in radioactivity leads to enhanced indoor exposures of the inhabitants of such buildings.

3. COAL FIRED POWER PLANTS VERSUS NUCLEAR POWER PLANTS

Comparative studies of coal fired power plants and nuclear power plants have been reported by many scientists [1, 6, 8]. Several authors have reported the results of studies dealing with radionuclides in either coal fired plants or nuclear power plants [9, 10]. The general tasks of these studies are: (a) to measure the concentration and volumes of radionuclides released to the environment from a given source, or to provide estimates of values on the basis of sound information; (b) to model the dispersion of radionuclides in the atmosphere and their deposition on the ground; (c) to model the uptake of radionuclides and the radiation doses; and (d) to evaluate the health risks from radiation or to compare the radiation doses with those from natural background radiation.

Several published reports on these studies, however, point out that there remain a number of queries regarding the quality of the data and the adequacy of the models. It is also important to emphasize the existing levels of uncertainty associated with predictions based on these approaches. Table III gives the reported
TABLE III. REPORTED LEVELS OF UNCERTAINTY ASSOCIATED WITH ESTIMATES OF RADIOLOGICAL IMPACTS OF COAL FIRED POWER PLANTS AND NUCLEAR POWER PLANTS

<table>
<thead>
<tr>
<th>Parameters considered</th>
<th>Order of magnitude of uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>Source terms</td>
<td>1</td>
</tr>
<tr>
<td>Dispersion and deposition</td>
<td>1</td>
</tr>
<tr>
<td>Dose conversion</td>
<td>3</td>
</tr>
<tr>
<td>Risk estimates per sievert</td>
<td>0.03</td>
</tr>
</tbody>
</table>

* The total uncertainty is estimated to be a factor of 1400–2000.

levels of uncertainty associated with estimates of radiological impacts of coal fired power plants and nuclear power plants in the literature.

For example, the uncertainty associated with data and with sample collection from the stack of a coal fired power plant is at least 30% and more often an order of magnitude. Atmospheric dispersion models currently being used predict the deposition of radionuclides to within about an order of magnitude of uncertainty. Dose conversion factors for ingested and inhaled radionuclides add another one to three orders of magnitude of uncertainty. The risk conversion factors also have uncertainties and are at least as uncertain as the dose conversion factors. The level of uncertainty associated with predictions of health and environmental risk from radionuclides in coal is the sum of all of the above uncertainties.

There is an additional flaw in the use of various models for comparing the radiological risks of coal fired power plants and nuclear power plants. The principal radionuclides released from nuclear power plants differ from those released from coal fired plants in their physiological and environmental pathways. It is very difficult to determine whether the apparent differences in the estimates of radiological risks from coal fired power plants and nuclear power plants are due to the bias in each case or whether they are real. The uncertainties in the quantities of radionuclides released from both types of power plant are, however, smaller than the uncertainties associated with health risk assessments [11].

Table IV gives the variations in dose estimates reported by various authors. Table V gives the estimated collective dose commitments from coal fired and nuclear power plants in India. It can be seen that the dose commitments are higher for coal fired power plants.

The US Environmental Protection Agency compared data (1979) for radiological impacts caused by atmospheric emissions from several types of power stations [13]. The mean dose commitments estimated for 25 BWRs are 0.11 mSv/a
TABLE IV. REPORTED VARIATIONS IN DOSE ESTIMATES

<table>
<thead>
<tr>
<th>Dose from thermal power plants (mSv/a)</th>
<th>Dose from nuclear power plants (mSv/a)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0004 (stack)</td>
<td>0.004–0.014</td>
<td>[12]</td>
</tr>
<tr>
<td>0.0004–0.0035</td>
<td>—</td>
<td>[9]</td>
</tr>
<tr>
<td>0.019</td>
<td>0.018 (PHWR)</td>
<td>[13]</td>
</tr>
<tr>
<td></td>
<td>0.046 (BWR)</td>
<td></td>
</tr>
<tr>
<td>0.067–0.15 (lungs)</td>
<td>0.008 (PHWR)</td>
<td>Old plants [13]</td>
</tr>
<tr>
<td>0.083–0.26 (bones)</td>
<td>0.02 (BWR)</td>
<td></td>
</tr>
<tr>
<td>0.008–0.021 (lungs)</td>
<td>—</td>
<td>New plants</td>
</tr>
<tr>
<td>0.016–0.16 (bones)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.02–0.201</td>
<td>0.021–0.054 (PHWR)</td>
<td>Present study</td>
</tr>
<tr>
<td></td>
<td>0.046–0.269 (BWR)</td>
<td></td>
</tr>
</tbody>
</table>

TABLE V. ESTIMATED COLLECTIVE DOSE COMMITMENTS (man-Sv/a) TO THE POPULATION RESIDING WITHIN AN AREA OF 88.5 km RADIUS OF A 1000 MW(e) POWER PLANT

<table>
<thead>
<tr>
<th></th>
<th>Coal fired power plant</th>
<th>Nuclear power plant (PHWR type)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Dose excluding ingestion pathway</td>
<td>Dose including ingestion pathway</td>
</tr>
<tr>
<td>Whole body dose</td>
<td>4.7</td>
<td>40.0</td>
</tr>
<tr>
<td>Organ doses (bones, lungs and thyroid)</td>
<td>73.0</td>
<td>206.0</td>
</tr>
</tbody>
</table>

for the thyroid and 0.02 mSv/a for the whole body; those for 44 BWRs are 0.01 mSv/a for the thyroid and 0.008 mSv/a for the whole body; for 250 existing coal fired power plants the estimates are in the range of 0.067–0.15 mSv/a for the lungs and 0.083–0.26 mSv/a for the bones. The estimated dose levels for Indian BWR type nuclear power stations are 0.269 mSv/a for the thyroid and 0.054 mSv/a
for the whole body; those for coal fired power plants are 0.02 mSv/a for the lungs and 0.201 mSv/a for the bones. The estimates for the worst type of thermal power plant exceed the estimate for the worst type of normally operating nuclear power plant, but for modern coal fired power plants the same doses as for nuclear plants are expected [13].

The environmental impact of nuclear power plants is only due to radioactive emissions. However, in the case of coal fired thermal power plants, a number of conventional air and water pollutants are also emitted into the atmosphere and into water bodies. To assess the overall impact, the present authors have attempted in two earlier publications [14, 15] to define the term environmental quality index (EQI). This is obtained by adding the ratios of the maximum observed/expected concentration of each pollutant to its ambient air/water standard. Even though this is not scientifically correct in view of the differences in the impacts of different types of pollutants, it does provide a quantity in terms of which different types of electrical plants can be compared.

It has been observed that, compared with chemical pollutants (SO$_2$, NO$_x$, CO, HC, Pb, particulates, etc.), the contribution of radioactivity to the EQI of thermal power plants is significant. The contribution of radioactivity to the EQI of PHWR nuclear power plants of nearly the same capacity is about ten times lower. The inhalation dose from thermal power plants becomes significant if the individual dose limit of 1 mSv/a for members of the public is used. In the case of nuclear power plants, the safety of population groups residing near the plant (critical groups) is ensured by the use of zoning concepts (exclusion zone and sterilization zone). Such a system is not normally considered when siting thermal power plants. Hence the total population dose due to the inhalation of radioactivity released from the stack of such a plant is considerable. Assessment of this dose may be desirable for coal fired power plants, as well as the use of a zoning concept as it is applied in nuclear power plants.

The present study for India shows that the exposure of the population living within a radius of 88.5 km of a power station of either type is only a small fraction of the exposure due to natural background radiation (about 0.135% in the case of thermal power plants and 0.117% in the case of nuclear power plants).

4. CONCLUSIONS

An accurate comparison of the radiological impact of coal fired power plants with that of nuclear power plants is quite difficult because of (a) insufficient data on both types of plant, (b) differences in the design and the operating conditions of coal fired plants and in their emission control systems, and (c) the relatively low concentrations of radionuclides to be measured. The radiation doses to the population from the operation of these types of plant are small compared with the doses from natural background radiation. Furthermore, the level of uncertainty in the estimation of the
radiological impact is so large that it is not possible at present to categorically state the degree of difference between the radiological risks from coal fired plants and from nuclear power plants. However, it has been shown that the impact of coal fired plants is at least as high as that of nuclear power plants; in the case of coal fired plants based on old technology, it is worse.

REFERENCES


INHALATION OF THE AEROSOL OF NUCLEAR FUEL PARTICLES FROM THE CHERNOBYL NUCLEAR POWER PLANT BY ADULT PERSONS FROM THE GOMEL REGION OF BELARUS

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Abstract

INHALATION OF THE AEROSOL OF NUCLEAR FUEL PARTICLES FROM THE CHERNOBYL NUCLEAR POWER PLANT BY ADULT PERSONS FROM THE GOMEL REGION OF BELARUS.

Under the conditions of the Chernobyl accident, plutonium isotopes were released only as part of nuclear fuel particles. To investigate the role of these particles in the exposure of Chernobyl victims, the results of post-mortem measurements of the plutonium body content of 125 adult persons living in the Gomel region were examined by officers of the Gomel Branch of the Institute for Radiation Medicine. Autopsy samples were investigated radiochemically for the total specific activity of alpha emitting plutonium isotopes. The isotopic composition of plutonium was determined by alpha spectrometry. The available data suggest that the distribution of the nuclear fuel particle aerosol inhaled by adults in the Gomel region is logarithmic-normal, with a median value of 11 Bq $^{239}\text{Pu} + ^{240}\text{Pu}$ and with a geometric standard deviation of 5.0. This inhalation level leads to a committed effective dose of 1.3 mSv. The contribution of $^{239}\text{Pu} + ^{240}\text{Pu}$ to this dose is only 6%.

1. INTRODUCTION

As a result of the destruction of the core of Unit 4 of the Chernobyl nuclear power plant, large amounts of airborne particles of dispersed nuclear fuel as well as radioactive vapours and gases formed an aerosol of condensed particles that was released to the atmosphere.
Condensed particles were formed by radioactive vapours of relatively volatile elements, e.g. $^{103}\text{Ru}$, $^{106}\text{Ru}$, $^{131}\text{I}$, $^{134}\text{Cs}$ or $^{137}\text{Cs}$ and, probably, $^{90}\text{Sr}$, subsequent to interaction with non-radioactive material. When the radionuclides, which are bound by such aerosol particles, enter the body, they are quite independent of each other and follow the well known biokinetic patterns of their respective oxides. These radionuclides are inhaled or enter the body through food-chains and through consumption of foodstuffs contaminated by surface deposition of nuclides (surface contaminated foodstuffs). The condensed particles are well known as part of the aerosol in the air of working places in the nuclear industry.

Aerosol particles of fragmented nuclear fuel contain fission products and transuranium radionuclides strongly bound in the uranium matrix of nuclear fuel particles [1, 2]. Nuclear fuel particles are a rare form of airborne radioactivity. Similar aerosol particles were found in the environment only after nuclear tests in the atmosphere; their radiological properties are not well known. The dominant pathway of intake of these aerosols is inhalation.

The fuel particles found in the vicinity of Chernobyl all had a similar isotopic composition, close to that of a representative nuclear fuel particle. The strong bond between the radionuclides and the matrix allows the contamination of man or the environment by nuclear fuel particles to be estimated by using the activity of one of the radionuclide tracers which, under the Chernobyl accident conditions, are only present in these nuclear fuel particles. The tracers include isotopes of relatively refractory elements, e.g. Zr and Ce, and transuranium isotopes, e.g. Pu. The activities of the main radionuclides in a representative nuclear fuel particle, normalized to the activity of the tracer, were used in the investigations reported in Ref. [1]. The specific activity of the sum of $^{239}\text{Pu} + ^{240}\text{Pu}$ in $\text{UO}_2$ fuel is essentially independent of the time since the accident and is equal to 14 MBq per gram of $\text{UO}_2$.

2. MATERIAL AND METHODS

In the period 1990–1991, the Gomel Branch of the Institute for Radiation Medicine performed a study of the autopsy material obtained from 125 residents of 17 districts of the Gomel region, aged from 20 to 86 years, who died of different causes.

The dominant pathway of nuclear fuel particle intake by these individuals was inhalation. Since in most cases the period from the accident to the death of these persons averaged 1700 days, this intake was assumed to be a single event. The value of this intake is equal to the sum of the short term intake of radioactive aerosols by inhalation during the passage of a radioactive plume and the intake with air for about six months due to resuspension. The Reference Man models can be expected to provide average values of the radionuclide biokinetics in these individuals.
Samples of the liver, skeleton (fragments of the fifth or sixth rib), lung and pulmonary lymph nodes were investigated for the total specific activity of the alpha emitting Pu. The stages of the radiochemical analysis of the Pu content were as follows: decomposition of organic substances by dry and wet ashing, anion exchange separation of radionuclides using ion exchange resins, and measurement of the material obtained in a thin layer solid scintillator with a low background radiometer. The radiation background in glassware, reagents, etc. was continuously monitored.

In some samples, the Pu isotopic composition was determined by alpha spectrometry. In this case, following anion exchange separation, Pu was applied to stainless steel discs by electrolysis, with subsequent measurements of the alpha spectra using a counting ionization chamber. The Pu yield was checked, with $^{242}$Pu added to the samples as tracer prior to mineralization. The lower detection limit was 1.6 and 1.3 mBq per sample for $^{238}$Pu and for $^{239}$Pu + $^{240}$Pu, respectively.

Comparison of the total alpha activity of $^{238}$Pu + $^{239}$Pu + $^{240}$Pu, measured by an alpha spectrometer, with the alpha radiometry data showed satisfactory convergence of the results.

3. RESULTS AND DISCUSSION

3.1. Plutonium isotopic composition of the autopsy material

Table I shows the Pu isotopic composition found in organs of a few residents of the Gomel region. Nuclear tests are known to produce no appreciable amounts of atmospheric $^{238}$Pu. As estimated by UNSCEAR, the 1945–1980 atmospheric tests released two Pu isotopes: $^{239}$Pu (6.5 PBq) and $^{240}$Pu (4.4 PBq); by 1986, the total surface density of $^{239}$Pu + $^{240}$Pu global fallout in the northern hemisphere at the latitude of Chernobyl averaged 60 Bq/m$^2$ [3]. According to the OECD [4], the major contributor of $^{238}$Pu was burnup of this isotope upon re-entry of satellites that used $^{238}$Pu as a power source. According to these estimates, up to 1986, about 0.6 PBq $^{238}$Pu entered the atmosphere. The data in Table I give the integral specific activity of global Pu and Chernobyl Pu. Long term global Pu contamination of the environment resulted in an equilibrium specific activity of Pu isotopes of primary and secondary deposition in human organs. The 1984 estimates for residents of western Europe at the altitude of the Gomel region (persons from Germany) evidenced that the lungs contained 2 ± 1 mBq, pulmonary lymph nodes 12 ± 10 mBq, liver 24 ± 4 mBq, skeleton 6 ± 3 mBq and the whole body 100 ± 25 mBq of $^{238}$Pu + $^{239}$Pu + $^{240}$Pu per kilogram [5].

Taking the pre-accident global background and the equilibrium global specific activity of Pu for the Gomel region to be equal to those for western Europe, we found that Pu exceeded the background level; the median ratio of $^{238}$Pu to $^{239}$Pu + $^{240}$Pu was 0.3, with the 25 and 75 percentiles being 0.25 and 0.35, respectively. The
TABLE I. ISOTOPIC COMPOSITION OF Pu IN EXAMINED ORGANS

<table>
<thead>
<tr>
<th>District of the region</th>
<th>Organ</th>
<th>Specific activity (mBq/kg)</th>
<th>239Pu + 240Pu</th>
<th>238Pu</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gomel</td>
<td>Liver</td>
<td>19.9 ± 1.5</td>
<td>3.97 ± 1.1</td>
<td></td>
</tr>
<tr>
<td>Gomel</td>
<td>Liver</td>
<td>21.7 ± 2.4</td>
<td>4.18 ± 2.0</td>
<td></td>
</tr>
<tr>
<td>Gomel</td>
<td>Liver</td>
<td>22.4 ± 1.7</td>
<td>3.73 ± 1.3</td>
<td></td>
</tr>
<tr>
<td>Gomel</td>
<td>Liver</td>
<td>50.6 ± 3.7</td>
<td>6.55 ± 3.2</td>
<td></td>
</tr>
<tr>
<td>Vetka</td>
<td>Liver</td>
<td>21.5 ± 6.3</td>
<td>10.6 ± 2.7</td>
<td></td>
</tr>
<tr>
<td>Vetka</td>
<td>Liver</td>
<td>22.7 ± 2.0</td>
<td>4.10 ± 1.8</td>
<td></td>
</tr>
<tr>
<td>Khoiniki</td>
<td>Liver</td>
<td>36.6 ± 2.5</td>
<td>3.50 ± 1.8</td>
<td></td>
</tr>
<tr>
<td>Rechitsa</td>
<td>Liver</td>
<td>80.7 ± 4.2</td>
<td>14.2 ± 2.8</td>
<td></td>
</tr>
<tr>
<td>Rechitsa</td>
<td>Liver</td>
<td>38.1 ± 5.0</td>
<td>5.0 ± 2.0</td>
<td></td>
</tr>
<tr>
<td>Svetlogorsk</td>
<td>Liver</td>
<td>18.5 ± 5.9</td>
<td>31.0 ± 0.5</td>
<td></td>
</tr>
<tr>
<td>Rogachev</td>
<td>Liver</td>
<td>18.0 ± 1.4</td>
<td>3.0 ± 1.1</td>
<td></td>
</tr>
<tr>
<td>Budo-Koshelev</td>
<td>Liver</td>
<td>43.1 ± 2.6</td>
<td>6.4 ± 1.9</td>
<td></td>
</tr>
<tr>
<td>Budo-Koshelev</td>
<td>Liver</td>
<td>39.3 ± 2.8</td>
<td>5.6 ± 2.1</td>
<td></td>
</tr>
<tr>
<td>Narovlya</td>
<td>Liver</td>
<td>54.9 ± 2.7</td>
<td>4.0 ± 1.6</td>
<td></td>
</tr>
<tr>
<td>Zhlobin</td>
<td>Liver</td>
<td>18.8 ± 1.6</td>
<td>3.6 ± 1.2</td>
<td></td>
</tr>
<tr>
<td>Zhitkovich</td>
<td>Liver</td>
<td>28.0 ± 3.8</td>
<td>10.1 ± 4.1</td>
<td></td>
</tr>
<tr>
<td>Gomel</td>
<td>Lungs</td>
<td>4.0 ± 1.8</td>
<td>7.5 ± 2.4</td>
<td></td>
</tr>
<tr>
<td>Vetka</td>
<td>Lungs</td>
<td>14.2 ± 2.4</td>
<td>4.5 ± 2.4</td>
<td></td>
</tr>
<tr>
<td>Gomel</td>
<td>Bones</td>
<td>17.9 ± 5.2</td>
<td>19.4 ± 6.5</td>
<td></td>
</tr>
<tr>
<td>Chechersk</td>
<td>Bones</td>
<td>8.4 ± 3.2</td>
<td>6.6 ± 3.3</td>
<td></td>
</tr>
</tbody>
</table>

correlation coefficient for the specific activity of 238Pu and 239Pu + 240Pu appeared to be 0.58. The data obtained are in good agreement with our assessment of the radionuclide inventory of fuel in Unit 4 at the time of the accident [1]. It follows that Pu isotopes found in residents of the Gomel region are of Chernobyl origin.

3.2. Plutonium in residents of the Gomel region affected by the Chernobyl accident

The Pu content in the liver of persons living in the Gomel region was determined in 125 (100%) individuals. For 65% of the cases, the total Pu specific activity
exceeded 28 Bq/kg, i.e. the average specific activity in the liver of persons in western Europe plus a standard deviation of unity. A detailed study of Pu distribution in the body was carried out for a small control group of about 30 persons, mostly those with a specific activity of Pu in the liver in excess of the background level. The Pu content of the lung was determined in 31 persons, that of the skeleton in 33 persons and that of pulmonary lymph nodes in 9 persons. Table II gives the specific activity values of Chernobyl Pu used for further analysis to obtain $R_{Pu}$; these values include the data for 21 residents of the Gomel region with a specific activity of Pu in the lung and the liver in excess of the western European average. These figures are the values obtained for the above group, minus the global Pu background in the organs and tissues of people in western Europe, thus indicating the specific activity of alpha emitting Pu isotopes from Chernobyl.

On the average, the lung-to-liver total activity ratio ($R_{Pu}$)

$$R_{Pu}(AMAD, t) = \frac{Q_{lungs}(AMAD, t)}{Q_{liver}(AMAD, t)}$$

is dependent on the aerosol particle size (the activity median aerodynamic diameter (AMAD) of nuclear fuel particle aerosol distribution) and the time (t) after a single inhalation [2]. The set of $R_{Pu}$ values obtained from the Gomel data is shown in Table II. Thus, the residents of the Gomel region affected by the accident have a characteristic mean $R_{Pu}$ value of 0.87.

The aerosol size dependence of the fractional Pu activities in the liver ($Q_{liver}$) and lungs ($Q_{lungs}$) of the adult Reference Man for $t = 1700$ d after a single inhalation of fuel particle aerosol was evaluated with the ‘R-MAN’ code [6]. These fractional activities vary according to the following equations:

$$Q_{liver}(AMAD) = I_0 (3.8 \times 10^{-3}) \exp(-0.042 \text{ AMAD})$$

$$Q_{lungs}(AMAD) = I_0 (1.3 \times 10^{-2}) \exp(-0.15 \text{ AMAD})$$

where AMAD is in micrometres and $I_0$ is the intake of fuel particle aerosol in becquerels.

According to the mean $R_{Pu}$ of 0.87, an AMAD of 12 ± 3 μm for the distribution of fuel particles is expected for inhabitants of the Gomel district who were affected by the accident.

Results of alive and post-mortem evaluation of the body content of staff of the Chernobyl nuclear power plant (CNPP staff) who witnessed the Chernobyl accident [1] and of inhabitants of the Gomel district who were affected by the accident were used for the estimation of AMAD values of nuclear fuel particles. The results of this estimate (mean AMAD ± standard deviation of mean) are as follows:
TABLE II. SPECIFIC ACTIVITY (SA) OF CHERNOBYL PLUTONIUM\(^a\) IN ORGANS OF THE EXAMINED GROUP OF GOMEL REGION INHABITANTS

<table>
<thead>
<tr>
<th>Date of death</th>
<th>Age (years)</th>
<th>District of the region</th>
<th>Settlement</th>
<th>Lungs</th>
<th>Liver</th>
<th>(R_{Pu})</th>
</tr>
</thead>
<tbody>
<tr>
<td>1990-11-15</td>
<td>62</td>
<td>Vetka</td>
<td>Khalch</td>
<td>38</td>
<td>80</td>
<td>0.27</td>
</tr>
<tr>
<td>1990-11-15</td>
<td>51</td>
<td>Gomel</td>
<td>Gomel</td>
<td>23</td>
<td>7.1</td>
<td>1.8</td>
</tr>
<tr>
<td>1990-11-17</td>
<td>68</td>
<td>Gomel</td>
<td>Gomel</td>
<td>23</td>
<td>10</td>
<td>1.3</td>
</tr>
<tr>
<td>1990-11-18</td>
<td>61</td>
<td>Vetka</td>
<td>Novoselki</td>
<td>30</td>
<td>6.0</td>
<td>2.8</td>
</tr>
<tr>
<td>1990-11-20</td>
<td>66</td>
<td>Gomel</td>
<td>Terenichi</td>
<td>17</td>
<td>6.0</td>
<td>1.5</td>
</tr>
<tr>
<td>1990-11-22</td>
<td>55</td>
<td>Vetka</td>
<td>Neglubka</td>
<td>29</td>
<td>52</td>
<td>0.31</td>
</tr>
<tr>
<td>1990-11-23</td>
<td>55</td>
<td>Gomel</td>
<td>Gomel</td>
<td>34</td>
<td>16</td>
<td>1.2</td>
</tr>
<tr>
<td>1990-11-24</td>
<td>66</td>
<td>Gomel</td>
<td>Gomel</td>
<td>40</td>
<td>62</td>
<td>0.36</td>
</tr>
<tr>
<td>1990-11-24</td>
<td>63</td>
<td>Bragin</td>
<td>Komarin</td>
<td>33</td>
<td>28</td>
<td>0.66</td>
</tr>
<tr>
<td>1990-11-24</td>
<td>60</td>
<td>Vetka</td>
<td>Stolbun</td>
<td>44</td>
<td>73</td>
<td>0.33</td>
</tr>
<tr>
<td>1990-11-26</td>
<td>65</td>
<td>Vetka</td>
<td>Svetilovichi</td>
<td>15</td>
<td>5.2</td>
<td>1.6</td>
</tr>
<tr>
<td>1990-11-29</td>
<td>63</td>
<td>Buda-Koshelev</td>
<td>Boevoj</td>
<td>47</td>
<td>34</td>
<td>0.77</td>
</tr>
<tr>
<td>1990-12-01</td>
<td>46</td>
<td>Bragin</td>
<td>Bragin</td>
<td>29</td>
<td>110</td>
<td>0.16</td>
</tr>
<tr>
<td>1990-12-04</td>
<td>51</td>
<td>Rogachev</td>
<td>Zagrebenie</td>
<td>54</td>
<td>53</td>
<td>0.57</td>
</tr>
<tr>
<td>1990-12-12</td>
<td>78</td>
<td>Vetka</td>
<td>Sivinka</td>
<td>18</td>
<td>5.6</td>
<td>1.8</td>
</tr>
<tr>
<td>1990-12-17</td>
<td>49</td>
<td>Dobrush</td>
<td>Ivanpolie</td>
<td>39</td>
<td>25</td>
<td>0.87</td>
</tr>
<tr>
<td>1990-12-21</td>
<td>61</td>
<td>Bragin</td>
<td>Bragin</td>
<td>27</td>
<td>74</td>
<td>0.20</td>
</tr>
<tr>
<td>1991-01-08</td>
<td>31</td>
<td>Gomel</td>
<td>Gomel</td>
<td>17</td>
<td>59</td>
<td>0.16</td>
</tr>
<tr>
<td>1991-02-13</td>
<td>70</td>
<td>Gomel</td>
<td>Gomel</td>
<td>14</td>
<td>10</td>
<td>0.75</td>
</tr>
<tr>
<td>1991-03-25</td>
<td>55</td>
<td>Gomel</td>
<td>Gomel</td>
<td>11</td>
<td>19</td>
<td>0.33</td>
</tr>
<tr>
<td>1991-06-05</td>
<td>80</td>
<td>Gomel</td>
<td>Gomel</td>
<td>13</td>
<td>13</td>
<td>0.55</td>
</tr>
</tbody>
</table>

\(\text{Chernobyl Pu} = \text{total Pu} - \text{pre-accident Pu}\).

— 16 ± 2 \(\mu\)m, from the data of 15 living CNPP staff who witnessed the accident, examined with a semi-conductor whole-body counter in 1986–1987 [1].
— 12 ± 2 \(\mu\)m, from autopsy data of 23 dead CNPP staff who witnessed the accident and died 90 days after the accident [1].

These values are close to those of the dimensions of uranium dioxide grains in fuel pellets of Chernobyl type reactors.

3.3. Intake of nuclear fuel particles by residents of the Gomel region

To assess the intake of fuel particles by residents of the contaminated areas of the Gomel region, we chose individuals with a total specific activity of the alpha emitting Pu in the liver in excess of the background level of 28 mBq/kg. The distribution of the levels of total Chernobyl alpha emitting Pu in persons, as found by autopsy, was characterized by a logarithmic-normal density function with a median value of 29 mBq and with a geometric standard deviation of about 5.0.

Relation (2) was used in the reconstruction of the primary intake of fuel particles by the residents of the Gomel region. According to earlier estimates, the AMAD of fuel particles was assumed to be 12 μm. Within 1700 days, the liver retained 0.2% of the Pu inhaled with this aerosol. The available data suggest that the Pu intake by 65% of the residents of the Gomel region affected by the Chernobyl accident shows a logarithmic-normal distribution, with a geometric standard deviation of 5.0 and with a median value of 15 Bq $^{238}$Pu + $^{239}$Pu + $^{240}$Pu (11 Bq $^{239}$Pu + $^{240}$Pu).

3.4. Radiation dose

A special procedure, based on the biokinetic model for fuel particle radionuclides, was used for the retrospective evaluation of internal exposure [1, 2].

For our estimates we used the available results of post-mortem measurements of the Pu body content of Gomel inhabitants who witnessed the accident and were examined in 1990–1991 by officers of the Gomel Branch of the Institute for Radiation Medicine.

Because of the matrix, the behaviour of fuel particle radionuclides in the body acquires a collective character. Bound by the matrix, the radionuclides do not become 'individual' in the body until the matrix is chemically destroyed in the barrier organs, e.g. the respiratory system or the gastrointestinal (GI) tract.

It stands to reason that the nuclear fuel particles are the crystals produced by fragmentation of fuel pellets. Their lattice forms a fuel matrix and strongly holds fission products and transuranium radionuclides. By analogy with the assignment of U oxides by the International Commission on Radiological Protection (ICRP) to inhalation class Y according to their transportability in the respiratory system, all radionuclides entering the respiratory system as part of fuel particles should be assigned to the same inhalation class.
Therefore, the following model for the behaviour of matrix bound radio-
uclides in the body is suggested:

(a) Because of the matrix, the behaviour of fuel particle radionuclides in the body
acquires a collective character. Matrix bound radionuclides penetrate into body
fluids only after chemical destruction of the fuel matrix.

(b) The matrix bound radionuclides acquire biokinetic properties of U oxides
which are normally unusual for them, so that they should be assigned to the
non-transportable compounds of inhalation class Y. When fuel particles enter
the GI tract, the fractional absorption of matrix bound radionuclides does not
exceed that of U oxides.

(c) The distribution and retention of radionuclides penetrating into body fluids
after chemical destruction of the fuel matrix have individual peculiarities and
are determined only by the biokinetic properties of the related elements.

The model has been used as part of the computer code 'R-MAN' [6] to calcu-
late the committed equivalent dose in target organs and tissues of the adult Reference
Man. The mathematical phantom of the adult Reference Man developed by the Oak
Ridge National Laboratory [7], the modern model of aerosol particle deposition in
the human respiratory tract [8] and recent ICRP recommendations [9] were used in
the dose calculations.

The available data suggest that the inhalation intake of fuel particle aerosols
by adult Chernobyl victims living in the Gomel region in April–May 1986 shows a
logarithmic-normal distribution, with a median value of 11 Bq $^{239}$Pu + $^{240}$Pu and
a geometric standard deviation of 5.0. In that case the Pu isotopes are used as
radionuclide tracers of nuclear fuel particles. This amount of inhaled fuel particle
aerosols leads to a committed effective dose of 1.3 mSv. The contribution of
$^{239}$Pu + $^{240}$Pu to that figure is only 6%.

4. CONCLUSIONS

Our studies indicate unique properties of the Chernobyl aerosol of nuclear fuel
particles. In these particles, all radionuclides acquire similar biokinetic character-
istics and are cleared from the lungs in the same way as UO$_2$. It was observed that
nuclear fuel particles contain fission products as well as transuranium radionuclides
that are strongly bound in the matrix of U oxides. Under the conditions of the
Chernobyl accident, Pu isotopes were released only as a part of nuclear fuel
particles. The contribution of $^{239}$Pu + $^{240}$Pu to the committed effective dose of
residents of the Gomel region affected by the Chernobyl accident is negligible, but
these isotopes are very important as tracers of fuel particles. The findings should be
taken into consideration in the planning of protective measures and in the assessment
of health effects following a major accident at a nuclear power installation.
REFERENCES


AERIAL GAMMA MAPPING SYSTEM FOR CHARACTERIZING THE IMPACT OF RADIOACTIVE DEPOSITION ON THE ENVIRONMENT

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Abstract
AERIAL GAMMA MAPPING SYSTEM FOR CHARACTERIZING THE IMPACT OF RADIOACTIVE DEPOSITION ON THE ENVIRONMENT.

The Commissariat à l'énergie atomique has developed an aerial system of gamma cartography called HELINUC. This system makes it possible to establish, within a few hours, a map of radioactivity for areas (several dozens to several hundreds of hectares) by identifying the radioelements present, in a range from the level of natural radioactivity to that of man-made radioactivity resulting from a large scale accident. HELINUC has been operational for about ten years and is part of the French intervention system in the event of a civil or military nuclear accident. The HELINUC system is mounted on a light helicopter (equipped with a NaI detector) and is linked to ground based equipment. HELINUC has been used at the request of the Marine Environment Laboratory (IAEA-MEL), Monaco, to estimate the radioactivity of nine industrial or nuclear sites between Budapest and the Black Sea, as well as that of a stretch of 2000 km of the river banks of the Danube. The paper presents a few main results of this measurement campaign (carried out in early 1992), particularly those concerning the Paks nuclear power station in Hungary. The cartography of the Turnu Severin site (Romania) was a good demonstration of the method; the maps obtained showed the presence of spots of $^{137}\text{Cs}$ from the accident at the Chernobyl nuclear power plant. Use of three-dimensional digitized maps in support of isoactivity maps improves the readability of the maps and increases the quantity of available information. Furthermore, this mapping method is applicable to environmental monitoring for the purpose of surveillance of industrial and agricultural pollution. The results obtained with the HELINUC system during the different measurement campaigns have shown that the aerial gamma cartography method is a rapid, powerful and efficient technique for measuring and controlling the radioactivity in the environment of industrial or nuclear sites.

1. PRINCIPLE AND DESCRIPTION

The Commissariat à l'énergie atomique has developed an aerial system of gamma cartography called HELINUC. This system makes it possible to establish, within a few hours, a map of radioactivity for areas (several dozens to several
hundreds of hectares) by identifying the radioelements present, in a range from the level of natural radioactivity to that of man-made radioactivity resulting from a large scale accident.

1.1. **Experimental device**

The helicopter borne device is composed of a NaI detector containing 16 L (soon it will contain 32 L), which is connected to a spectrometer for signal analysis, with 512 channels, ranging from 30 to 3000 keV.

Positioning of the helicopter is ensured by either a Trident transponder (Thomson C.S.F.) or a global positioning system (GPS) (Trimble).

The Trident transponder queries two or three ground-level beacons set up at known geodesic points and computes by triangulation the distances between the helicopter and these beacons, inferring the helicopter position with an accuracy of within a few metres.

The GPS is a satellite positioning system that has been used in many applications (described in several publications). The present GPS will be replaced soon by a differential GPS having an accuracy equivalent to that of the Trident transponder.

The flight altitude is accurately measured with a radio-altimeter. Spectrum and position data management is ensured by a computer which stores the data on a Bernoulli magnetic disk.

1.2. **Data collection**

For a detailed site analysis, the following parameters are required: pattern size, altitude, flight velocity and integration time.

Every two or three seconds, the system records a full spectrum within the 30–3000 keV energy range together with the helicopter co-ordinates.

1.3. **Data processing**

The data collected and stored on a disk are processed by a dedicated ground-based computer. On the basis of the flight data records, colour maps are processed and overprinted on topographic maps.

Data processing starts with a check of the trajectories. Once the possible aberrant points are corrected, reading of the spectra corresponding to every point measured and identification of the involved radioelements begin. Selecting a detection window for each radioelement allows further computation and mapping at one reference altitude to be performed.

Such maps provide, in a very short time (1–2 h), the positions of contaminated areas, the contamination levels and the nature of the radioelements. In the event of a nuclear incident, this information could be handed over to competent authorities for use.
2. RESULTS AND APPLICATIONS

2.1. Equivalent surface activity

Any emission of a radioelement which is detected by the data acquisition unit comprises both the emission of the radioelement at the ground surface and the emission of the radioelement when it has migrated into the soil. The absorption of the detected signal is all the more significant as the radioelement is deep in the soil and its radiation energy is low. The equivalent surface activity refers to the activity arising from both the remaining surface contamination of the radioelement and the attenuated portion of the radioelement in the soil. For example, in the case of $^{137}$Cs, only the outer five centimetres contribute to the measurement [1].

2.2. System detection limit

Under normal operating conditions the integration time is 3 s, the flight altitude is 40 m and the flight velocity is 70 km/h.

At present, detection limits by aerial means are:

- for $^{241}$Am: 15 kBq/m$^2$ (surface source of 2000 m$^2$),
- for $^{137}$Cs: 1-2 kBq/m$^2$ (surface source of 2000 m$^2$),
- for $^{60}$Co: 1 kBq/m$^2$ (surface source of 2000 m$^2$),
- for $^{137}$Cs: 20 MBq (point source).

2.3. Application to incidental or accidental situations

Four persons are necessary to operate the equipment. In France, it can be operational within 12 h and moved to any point within the country in an air transport vehicle. Fitting out the helicopter, surveying an area of 5–10 km$^2$ and plotting the first contamination maps requires only about four hours [2, 3].

2.4. Application to environmental surveys

The airborne gamma mapping system offers multifarious applications. In France, the functions assigned to this system cover:

- Systematic radiological surveillance of military sites;
- Radiological surveillance and monitoring of nuclear sites, e.g. environmental surveillance of nuclear power plants;
- Radiological surveillance of the environment of industrial sites.

The results of the surveillance of the Danube river banks performed in 1992 [4, 5] on behalf of the IAEA are presented below.
The distribution of $^{40}$K in an agricultural area to the south of the Paks nuclear power plant has been measured. It might be expected that the level of $^{40}$K is higher in areas where fertilizers have been applied to the soil, thus raising the natural background level, but, on the contrary, the level of $^{40}$K is lower on the river banks of the Danube. An isoactivity map of $^{137}$Cs in the same area has also been prepared. The distribution of $^{137}$Cs is spatially homogeneous and uniformly low — $^{137}$Cs has been buried in the soil by agricultural activities — except for a small region on the eastern bank close to the river.

The campaign was performed when the water level of the Danube was low. Along the river we detected contamination points at all locations where the river flow velocity was lower, i.e. inside the bends. The deposited sediments acted like a natural filter, retaining the small particles of $^{137}$Cs from the Chernobyl accident.

It is in the Hungarian Danube plain that the caesium activity is lowest. The level of man-made contamination by radionuclides in the basin of the Danube is comparable with, although slightly higher than, that measured in France. The levels of $^{137}$Cs were highest on the western side of the southern Carpathians, for example in Turnu Severin (Romania), where the activities in certain spots were the highest ones that were mapped during the Danube campaign. On a small lawn in this city, the level of $^{137}$Cs, measured by gamma spectrometry with a high purity germanium detector, reached 18 kBq/m$^2$. It appears that the western and southern hillsides are more contaminated than the plain.

Our isoactivity maps have been treated with a new method of mathematical spectrum processing which has been developed in the Valduc/HELINUC laboratory [6]. It consists of searching for the absorption peaks over the whole energy range, 30–3000 keV, without making any assumptions on the nature of the radionuclides involved. The relevant algorithm includes the following principal steps:

— Use of filter algorithms adapted to the spectrum characteristics; this makes it possible to avoid high statistical fluctuations without changing the spectrum shape.

— Searching for the absorption peaks by means of one or two derivations with prior complex filtering. Hence, a low intensity peak, such as that of $^{241}$Am at 60 keV, may be located.

— Modelling and subtraction of the Compton scattering background enables the full absorption peaks of various radionuclides to be computed.

— Subjecting the absorption peaks derived from processing to validity checks regarding full width at half-maximum and regarding statistics.

The advantage of such a process is systematic detection of all radionuclides with absorption peak intensities higher than the detection threshold. This allows an improvement in the sensitivity of the system by factors of two to five in terms of signal-to-noise ratio to be achieved.
Implementation of this mathematical spectrum processing enables detection of the major natural or man-made radionuclides with sensitivities approximating the natural background activity levels.

2.5. Three-dimensional mapping

The conventional method for preparation of maps of the radiological status of different areas consists of superimposing the isoactivity map and the topographic map of a given site, through scanning of a printed map at a scale of 1:25 000 or 1:50 000. This technique is fast and cheap. However, the plotting quality of the topographic maps is rather poor. Therefore, we have investigated a high-tech method based on the digital Earth model (DEM). This model, developed by the ISTAR company (Sophia-Antipolis, France), is derived from pair stereoscopic pictures taken by the LANSAT or SPOT observation satellites. The DEM can be used to compute relief perspective views and to obtain three-dimensional views by matching the various points of the ortho-image.

These relief maps will be further used in supporting the airborne gamma spectrometry data. The results will provide very realistic information on the locations of contamination, including rivers, roads, cities, etc. These maps could be easily used in decision making in the case of incidental or accidental pollution.

3. CONCLUSION

The results obtained with the HELINUC system during the different measurement campaigns have shown that the aerial gamma mapping system is a powerful tool for plotting, very quickly, accurate maps of the radiological status of any area that is as large as several square kilometres, whether contaminated or not. This method is used in geophysical detection, as well as in environmental surveillance of industrial or nuclear plants or in emergency situations.

Recent advances, such as the use of digitized geographic maps and of higher resolution detectors (promising tests have already been done in a fixed wing plane using a high purity germanium detector), will make it possible to enlarge and improve the capacity and the field of application of such a system.

REFERENCES


RADIONUCLIDE RELEASES
TO THE AQUATIC ENVIRONMENT

(Session 3)

Chairman

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Sweden
MARINE RADIOACTIVITY STUDIES IN THE VICINITY OF SITES WITH POTENTIAL RADIONUCLIDE RELEASES

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Abstract

MARINE RADIOACTIVITY STUDIES IN THE VICINITY OF SITES WITH POTENTIAL RADIONUCLIDE RELEASES.

The paper gives a review of recent and current studies by the Marine Environment Laboratory (MEL) of the IAEA at four locations where radioactive materials have been intentionally or accidentally introduced into the marine environment. In each case the possibility of radionuclide leakage has led Member States to implement monitoring and assessment projects which have included an invited IAEA-MEL participation. The study sites are: (a) the former Soviet dumping sites in the Kara Sea, (b) the former European dumping sites in the north-east Atlantic, (c) the location in the Norwegian Sea where the Soviet nuclear powered submarine Komsomolets accidentally sank in April 1989, and (d) the former Soviet and Russian dumping sites in the Far Eastern seas. Generally, anthropogenic radionuclide levels observed in the investigated areas are low, the main contribution being from global weapons test fallout. In some dumping sites (Stepovovo and Tsivolki Bays, north-east Atlantic), higher concentrations of either $^{137}$Cs, $^{60}$Co or $^{239+240}$Pu have been measured in bottom waters and sediment, indicating a possible contribution from the disposed wastes. However, the IAEA-MEL assessments based on both radiometric survey and dispersion modelling suggest that the global radiological impact of these disposals is comparable to or less than the impacts resulting from other sources of anthropogenic radioactivity.

1. INTRODUCTION

The Marine Environment Laboratory (MEL) of the IAEA assists Member States in the measurement and understanding of marine radioactivity. Training, analytical quality assurance, the use of isotopes as tracers and site specific radiological assessments are central to the programme of the IAEA-MEL. The laboratory regularly provides assistance in the form of (a) independent observation for the United

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Nations, (b) radioanalytical and radiometric measurements, (c) analytical quality assurance and distribution of intercalibration materials, (d) maintenance of analytical databases, (e) measurement of biological concentration factors and associated biokinetics, (f) computer modelling of possible dispersions and transfers of radionuclides from dumped waste, and (g) radiological assessments of measured and modelled marine radioactivity data.

This paper gives a review of recent and current studies by the IAEA-MEL at four locations where radioactive materials have been introduced into the marine environment and where the possibility of nuclide leakage has led Member States to implement monitoring and assessment projects which have included invited IAEA-MEL participation. The study sites are: (a) the former Soviet high level waste dumping sites in the Kara Sea, (b) the former European dumping sites for low level wastes in the north-east Atlantic, (c) the location in the Norwegian Sea where the Soviet nuclear powered submarine Komsomolets, armed with nuclear weapons, accidentally sank in April 1989, and (d) the former Soviet Union and Russian dumping sites for high level nuclear wastes in the Far Eastern seas.

2. DUMPING SITES IN THE KARA SEA

In the Kara Sea, about 4.5 PBq of activity is at present associated with nuclear reactors dumped in shallow waters [1, 2]. The IAEA-MEL is contributing to the IASAP (International Arctic Seas Assessment Project) programme of the IAEA [3]. Staff of the IAEA-MEL have participated in four expeditions to the Kara Sea during the past three years, bringing to Monaco extensive sets of marine samples from each dumping site, from the open Kara Sea and from the mouths of the rivers Ob and Yenisey, which might carry major inventories of anthropogenic radionuclides from previously contaminated land in the Urals and Siberia. Samples have been analysed for both man made and natural radionuclides, the latter being important for the understanding of the rates and mechanisms of sedimentation and diagenesis [4–6]. The IAEA-MEL results on radiometric analyses of sediment samples taken from the open south-west Kara Sea during the 1992 Norwegian-Russian expedition have been published [7]. They show that the open Kara Sea is relatively uncontaminated, the main contributions being from nuclear weapons test fallout and from land based sources.

2.1. Stepovovo and Tsivolki Bays

Progress has been made in the assessment of sediment contamination on the basis of results from the 1993 Norwegian-Russian expedition (1993 W Kara Sea Cruise) to the Stepovovo and Tsivolki Bays. The presence of relatively high concentrations of $^{60}$Co and $^{137}$Cs in core CSB 9304/6 [8], sampled from the inner part of
the Stepovovo Bay, compared with the levels in the outer part of the Bay and in the open Kara Sea, where $^{60}$Co usually is not present, indicates contamination from local sources (Fig. 1). The plutonium concentrations in the core are similar to those in the open Kara Sea [7]. Higher $^3$H levels (by a factor of two to three) have been observed in bottom waters, suggesting $^3$H leakage from the wastes. As could be expected, because of the comparatively poor quality of the packaging barriers, leakage probably has occurred from dumped containers in the inner part of Stepovovo Bay. However, the leakage has not led to a measurable increase of radioactivity in the outer part of the Bay. The highly localized character of this sediment contamination is an interesting feature of the area investigated, perhaps suggesting leakage in particulate form. The $^{210}$Pb chronology indicates a maximum $^{239+240}$Pu input into the sediment around the year 1980.

The second sediment profile (CTB 9302/1) shown in Fig. 1 was taken from a shallow near-shore site of Tsivolki Bay in the proximity of a sunken ship, in an area where dumping operations were carried out. The concentrations of both $^{137}$Cs and $^{239+240}$Pu increase with depth. Dating of the core with $^{210}$Pb indicates rapid deposition of sediment. It is noteworthy that the $^{238}$Pu/$^{239+240}$Pu ratios vary from $0.036 \pm 0.005$ to $0.08 \pm 0.01$, a range which slightly differs from the values expected for weapons test fallout, whether global or local.

The main conclusion from these observations is that no radiologically significant leakage from the major objects containing nuclear fuel has been observed. However, minor local leakages from radioactive wastes dumped in metal containers have been observed and there is some evidence also of a plutonium contribution of an as yet undefined origin. Subsurface maxima in sedimentary plutonium ($^{210}$Pb — dated around the year 1980) have been observed elsewhere in the southern and western Kara Sea [7] and this non-fallout component may therefore be relatively widespread.

### 2.2. Sea-bed gamma spectrometry

The IAEA–MEL has recently introduced for survey purposes a dual detector (NaI(Tl) and HPGe) system for underwater and sea-bed gamma spectrometry [9]. High resolution data collected near one dumping site in Stepovovo Bay show the absence of gamma emitters from the dumped materials and the dominance of natural radionuclides (Fig. 2). The HPGe spectrum clearly shows the presence of global fallout $^{137}$Cs, which at such low levels is indistinguishable in the NaI(Tl) spectrum recorded simultaneously. The spectra obtained with the HPGe spectrometer represent the first set of high resolution sea-bed gamma spectra ever recorded in situ. The main implication of these results is that no large scale leakage has occurred in the past. The future thrust of IAEA–MEL's Kara Sea studies is therefore centred on further development of in situ gamma spectrometry (e.g. satellite linked telemetric
FIG. 1. Downcore profiles of $^{137}$Cs, $^{60}$Co, $^{239+240}$Pu and $^{238}$Pu in sediment collected in 1993 at depths of 57 and 28 m in the bays of Novaya Zemlya (sediment cores CSB 9304/6 and CTB 9302/1, respectively).
FIG. 2. Sea-bed gamma spectra taken by HPGe and NaI(Tl) detectors in Stepovovo Bay (1993 W Kara Sea cruise).
FIG. 3. Depth averaged concentrations of $^{137}$Cs during the second year after release of 1 PBq of $^{137}$Cs at the Kara Sea dumping sites.
monitoring systems) and on theoretical predictions of the dispersion, transfer and dosimetry of radionuclides which could possibly be released in future leakage from these Arctic wastes.

2.3. Modelling and radiological assessment

In the first instance, a box model approach has been used to assess the order of magnitude global dosimetry which could result from source leakage [6, 7]. Preliminary estimates of the maximum committed collective dose and the individual dose rate from fish ingestion are low (~10 man·Sv and <1 μSv/a, respectively) and, thus, only potential exposures to regional populations may be significant. The IAEA-MEL is giving priority to local three dimensional circulation and dispersion modelling in which potential source terms are superimposed on dynamic computer models driven by regional data on tides, temperature, salinity, wind stress, barometric pressure, river input and sea ice.

The numerical computations were carried out using the Hamburg Shelf Ocean Model (HamSOM), which is a three dimensional, baroclinic, finite difference model, based on non-linear shallow water equations [10]. It is forced with monthly mean wind stresses and river run-off, tides and seasonal mean temperature and salinity. The model is implemented on a stereographic grid with an average horizontal grid size of 20 km. The results of the regional scale model suggest that, even for the worst case scenario (instantaneous release of the total inventory of 1 PBq of $^{137}$Cs from all dumping sites), $^{137}$Cs concentrations in water of the east Kara Sea should not exceed 15 Bq/m$^3$ (Fig. 3) (removal of caesium from water to sediment has not been taken into account). A maximum individual dose of 5 μSv/a would be delivered to a fish eater in the Yamal and Taymyr coastal areas. For a gradual release rate scenario of 1 TBq/a of $^{137}$Cs from each of the Abrosimov, Stepovovo and Tsivolki Bays and from the Novaya Zemlya Trough ($^{137}$Cs inventories of 360, 130, 480 and 69 TBq, respectively), the maximum individual doses delivered through consumption of fish from the mainland coastal area would be below 0.2 μSv/a.

On a local scale, the HamSOM code was applied to the real topography of Abrosimov and Stepovovo Bays. The spatial resolution for the bay models was chosen to be 1/10 nautical mile (185.2 m). Wind was considered to be the most important driving force for bay water circulation. The stationary state $^{137}$Cs concentrations in water exiting the bays should be of the order of 1 kBq/m$^3$ per 1 TBq/a release, resulting in a maximum individual dose of below 1 mSv/a to a hypothetical individual consuming fish from the bays. The average flushing times of the investigated bays are in the range of 3–4 months if no ice cover is considered.

2.4. Ob and Yenisey estuaries

On the invitation of the Russian Academy of Sciences (via the Murmansk Marine Biological Institute), the IAEA-MEL took part in 1993 in a cruise to the
south-east Kara Sea (1993 E Kara Sea Cruise) to investigate the contributions of the Ob and Yenisey rivers to the radionuclide levels and inventories in the Kara Sea. A comparison of data from two sediment cores taken in the Ob (Station N6) and Yenisey (Station N13) estuaries shows very different $^{137}$Cs profiles (see Fig. 4). While the $^{137}$Cs inventories observed at the mouth of the Ob river are similar to those found in the open Kara Sea [7], Station N13 in the Yenisey estuary does indeed show much higher $^{137}$Cs inventories, suggestive of a local input. Extensive mixing to 9 cm depth is evident. Analyses of $^{60}$Co, $^{90}$Sr and $^{239+240}$Pu, and dating with $^{210}$Pb are in progress.
3. NORTH-EAST ATLANTIC DUMPING SITE

Prior to the above mentioned revelations about former Soviet disposals of radioactive wastes in the marine environment, it was believed that more than 98% of the packaged low level radioactive material disposed of in the oceans had been dumped at deep sites in the north Atlantic Ocean. In terms of activity, 92% of the total radioactive material disposed of was dumped in the eastern basin. While, in general, 98% of the total activity dumped comprised beta/gamma emitters, small quantities of alpha emitting nuclides were also included. At the two main sites in the north-east Atlantic (46°00' N, 16°45' W and 46°15' N, 17°25' W), a total activity of more than 30 PBq was disposed of. The alpha emitting inventory at these sites would be expected to be of the order of 0.5 PBq [11]. These dumping sites were used until 1982; previously and subsequently they were subject to radiological survey, normally on an annual basis.

In the past, the IAEA-MEL has regularly co-operated with the OECD/NEA and its Member States on analytical quality assurance aspects of this surveillance exercise. However, in March 1992, at the request of the Member States, the IAEA-MEL for the first time played a significant part in site specific measurements at the north-east Atlantic dumping site, since it was believed that maximum information on all scientific parameters associated with sea disposal of radioactive wastes should be collected. The IAEA-MEL contributed by analysing water samples, collected above the sea-bed of the main sites, for anthropogenic radionuclides such as $^{14}$C, $^{137}$Cs, $^{238}$Pu, $^{239+240}$Pu and $^{241}$Am. Samples were collected from the FRV Walther Herwig at one control site and at four locations in the area of the two main subsites, and were shared between the cruise organizers, the Bundesforschungsanstalt für Fischerei (BFA) in Hamburg (Germany), and the Fisheries Laboratory of the Ministry of Agriculture, Fisheries and Food at Lowestoft (United Kingdom) and the IAEA-MEL. The IAEA-MEL results show enhancements of 5-7 times in $^{238}$Pu concentrations in sea water collected at the dumping sites relative to those at the control site. The concentrations of $^{239+240}$Pu, $^{241}$Am, $^{137}$Cs and $^{14}$C are also higher in some samples from the dumping sites. The $^{238}$Pu/$^{239+240}$Pu activity ratio at the control site (0.029 ± 0.008) is similar to that expected for global fallout in the northern hemisphere and is considerably less than the ratios observed in the water at the dumping sites (from 0.08 ± 0.01 to 0.13 ± 0.01) (Fig. 5). These results suggest that measurable leakage is occurring at the dumping sites. It should be borne in mind, however, that the highest observed activities (~0.6 mBq $^{137}$Cs/L and ~20 μBq $^{239+240}$Pu/L) are extremely small compared with the naturally occurring radioactivity of open ocean water (12.5 Bq of beta/gamma emitters per litre and 115 mBq of alpha emitters per litre). Thus the localized enhancements represent increases of up to $10^{-2}$% and $10^{-2}$% in beta/gamma and alpha activities, respectively, and are therefore radiologically negligible.
FIG. 5. Concentrations of $^{239+240}\text{Pu}$ and $^{238}\text{Pu}/^{239+240}\text{Pu}$ ratios in bottom sea waters at and around the north-east Atlantic dumping site; all samples measured were collected ~10 m above the sea-bed. Station co-ordinates: CS-13: 45°59' N, 12°59' W (background station); DS-18: 46°03' N, 16°41' W; DS-22: 46°00' N, 17°00' W; DS-28: 46°07' N, 16°42' E; DS-33: 46°08' N, 16°41' E.
4. KOMSOMOLETS SITE

On 7 April 1989, a fire broke out in the stern section of the Komsomolets nuclear submarine, which sank to a depth of 1685 m at 73°43'16'' N, 13°15'52'' E, to the south-west of Bear Island. The site is about 300 nautical miles from the Norwegian coast. The wreck contains one nuclear reactor and two nuclear warheads, one of which was fractured. The radionuclide inventory includes 1.6 PBq $^{90}$Sr, 2 PBq $^{137}$Cs, about 16 TBq $^{239}$Pu in the two warheads and 5 TBq of actinides in the reactor core [1].

During June/July 1994, the IAEA-MEL provided assistance to an international expedition to the Komsomolets site at the request of the Russian Federation. The objectives of the scientific cruise on board the R/V Mstislav Keldysh were to close nine door holes, including torpedo tubes, by covering them with titanium metal caps, and to sample and monitor for ambient radioactivity. A series of 280-600 L seawater samples collected from a vertical profile, a suite of surface sediments and cores, and various biota samples obtained by trawling were brought to the IAEA-MEL for analysis.

Some of the results of radionuclide analyses are presented in Table I. The presence of $^{134}$Cs in sediment suggests a leakage of caesium originating from the submarine. The concentrations of $^3$H in deep waters of the north-east Atlantic (~1600 m depth) are expected to be below 3 TU [12]. The values given in Table I suggest that a leakage of $^3$H has probably occurred in the area close to the nuclear warheads. The analyses of $^{239+240}$Pu are currently being completed.

5. DUMPING SITES IN THE SEA OF JAPAN

The former Soviet Union also disposed of high, intermediate and low activity wastes in the Far Eastern seas, although, unlike in the Arctic, no reactors containing fuel were dumped there. Therefore, in the Sea of Japan, the inventory of dumped radioactive waste is relatively low, comprising ~440 TBq of liquids and ~140 TBq of solids [1]. Within the framework of a joint agreement between the Governments of Japan, the Republic of Korea and the Russian Federation, the IAEA-MEL was invited to participate in the first Japanese-Korean-Russian joint expedition to the radioactive waste dumping areas in their common seas. The cruise was carried out on board the R/V Okean during March-April 1994. Sea water from several depths, sea-bed sediments and biota were sampled at seven sites within the dumping zone and at an additional two sites outside this zone — the latter to reflect background levels.

The results show that the concentrations of $^{89}$Sr, $^{137}$Cs, $^{238}$Pu and $^{239+240}$Pu in the Sea of Japan are low and are predominantly due to global fallout [13]. Figure 6 shows the vertical profiles of the $^{90}$Sr, $^{137}$Cs and $^{239+240}$Pu concentrations in sea
TABLE I. CONCENTRATIONS OF RADIONUCLIDES IN THE VICINITY OF THE KOMSOMOLETS SUBMARINE IN JUNE–JULY 1994

<table>
<thead>
<tr>
<th>Sample</th>
<th>Radionuclide concentrations</th>
</tr>
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<tbody>
<tr>
<td></td>
<td>(Bq/kg dry weight)</td>
</tr>
<tr>
<td>Sediment</td>
<td>^137Cs</td>
</tr>
<tr>
<td>M 2-7-5 reactor</td>
<td>0-1 cm</td>
</tr>
<tr>
<td></td>
<td>1-2 cm</td>
</tr>
<tr>
<td></td>
<td>2-3 cm</td>
</tr>
<tr>
<td>M 2-3-5 nuclear warheads</td>
<td>0-1 cm</td>
</tr>
<tr>
<td></td>
<td>1-2 cm</td>
</tr>
<tr>
<td></td>
<td>2-3 cm</td>
</tr>
<tr>
<td>Background station</td>
<td>0-5 cm</td>
</tr>
<tr>
<td>Sea water</td>
<td>Close to reactor</td>
</tr>
<tr>
<td></td>
<td>Close to nuclear warheads</td>
</tr>
<tr>
<td></td>
<td>First compartment</td>
</tr>
<tr>
<td>Biota</td>
<td><em>Foraminifera</em></td>
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<tr>
<td></td>
<td><em>Macruridal</em></td>
</tr>
<tr>
<td></td>
<td><em>Lycodes</em></td>
</tr>
</tbody>
</table>

^a Errors represent 1σ counting errors.

water for one of the sites within the dumping zone. The ^90Sr and ^137Cs data represent typical distributions for conservative fallout radionuclides, while ^239+240Pu exhibits a distinct maximum at about 800 m depth and a slow gradient towards the bottom. This is also typical [14], since plutonium is scavenged in the euphotic zone and is subsequently associated with sinking particles and re-solubilized at depth. The water profiles together with the radionuclide inventories in the top 10 cm layer of underlying sediment were used to estimate the total radionuclide inventories per unit surface area. The results for ^90Sr (3.3 kBq/m²) and ^137Cs (4.8 kBq/m²) compare well with the deposition densities of global weapons fallout
FIG. 6. Vertical profiles of $^{90}$Sr, $^{137}$Cs and $^{239+240}$Pu concentrations in water from the Sea of Japan.

for the latitude band 40–50°N, namely 3.2 kBq/m² and 5.1 kBq/m² for $^{90}$Sr and $^{137}$Cs, respectively [15]. For $^{239+240}$Pu, the present measurements give twice the inventory predicted from global fallout, 0.12–0.13 kBq/m², compared with 0.06 kBq/m². However, this observation is consistent with previous data from the Atlantic and Pacific Oceans [16] and is explained by an underestimation of plutonium deposition over ocean areas compared with deposition on land. The predominance of weapons test global fallout in the study area is further supported by the radionuclide activity ratios of $^{137}$Cs/$^{90}$Sr, $^{239+240}$Pu/$^{90}$Sr and $^{238}$Pu/$^{239+240}$Pu, which indicate no statistically significant deviation from the expected global fallout derived values.
FIG. 7. Comparison of contributions from individual FAO fishing areas to the collective effective dose commitment by ingestion of $^{137}$Cs and $^{210}$Po in fish and shellfish caught in 1990 (based on biota data).
6. COMPARISON OF DOSES FROM $^{210}$Po AND $^{137}$Cs IN SEAFOOD

Perspectives on the results of all four of the above studies are maintained by their input into and comparison with the data recently produced by the IAEA–MEL in its MARDOS Co-ordinated Research Programme, in which an international team of experts has quantified the concentrations and the dosimetry of anthropogenic $^{137}$Cs and naturally occurring $^{210}$Po in each of the FAO ocean regions, with particular emphasis on public exposures via seafood consumption. Radioactivity levels of $^{210}$Po and $^{137}$Cs in sea water and biota (fish and shellfish) have been estimated for the FAO fishing areas on the basis of measurements carried out in recent years. Collective doses have been calculated for each FAO area, using radioactivity data for water and biota. Good agreement has been found between the results obtained with these two methods, except for the doses from $^{210}$Po by consumption of shellfish. The collective effective dose commitment for $^{137}$Cs in seafood in 1990 has been found to be 160 man·Sv, with an estimated uncertainty of 50%. The corresponding dose from $^{210}$Po is 30 000 man·Sv, with an estimated uncertainty of a factor of five.

The results confirm that the dominant contribution to doses comes from natural $^{210}$Po in fish and shellfish, and that the contribution of anthropogenic $^{137}$Cs (mostly from nuclear weapons tests) is negligible (100–1000 times lower) (Fig. 7). The results obtained provide the most comprehensive data set available on radioactivity levels in the marine environment and on doses to the world population through ingestion of seafood.

7. CONCLUSIONS

Radiometric and modelling contributions by the IAEA–MEL to international marine radioactivity studies in the vicinity of sites with potential radionuclide releases show that, in all cases, the radiological consequences on a global scale should not be of importance. Generally, the radionuclide levels observed in the investigated areas are low, the main contribution being from global radioactive fallout from atmospheric nuclear weapons tests. In some areas (e.g. Kara Sea dumping sites, north-east Atlantic dumping sites and the Komsomolets area), higher concentrations of $^{137}$Cs, $^{60}$Co, $^3$H and $^{239+240}$Pu have indeed been measured, indicating a localized contribution from radioactive sources situated on the sea-bed. However, these releases are shown to result in very restricted and minor contamination, no increases of radionuclide levels being measurable beyond the dumping areas.

Box and numerical dispersion modelling suggests that, in the Arctic, radiological effects on a local scale may be of importance. More precise assessment requires further studies on sedimentation rates, sediment resuspension and transport, ice formation, and values of distribution coefficients and concentration factors for the low temperature Arctic marine environment.
All of these current results are being input into the new IAEA-MEL GLOMARD database which, linked to a geographical information system (GIS), will provide information on radionuclide distributions in sea water, sediment and biota, on temporal and geographical trends, and on correlations with hydrography, bathymetry and geochemistry.

ACKNOWLEDGEMENTS

The authors are greatly indebted to the German, Japanese, Norwegian and Russian Governments for their various invitations to the IAEA-MEL to participate in the international investigatory cruises. The IAEA-MEL operates under an agreement between the IAEA and the Government of the Principality of Monaco.

REFERENCES


DUMPING OF RADIOACTIVE WASTE IN THE BARENTS SEA AND THE KARA SEA

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Abstract

DUMPING OF RADIOACTIVE WASTE IN THE BARENTS SEA AND THE KARA SEA.

In order to evaluate the level of radioactive contamination in the Kara Sea and to assess the short and long term consequences of dumped radioactive waste, joint Russian–Norwegian expeditions have been performed annually since 1992. The results from the 1992 joint expedition to the Kara Sea demonstrated very low concentrations of radionuclides in waters and sediments. Contributions from different sources could be identified: global fallout, river transport, marine transport of discharges from European reprocessing plants and fallout from the accident at the Chernobyl nuclear power plant. The expeditions in 1993 to the dumping sites confirmed local contamination in the Stepovogo Fjord and the Tsivolki Fjord due to leakage from dumped radioactive waste. The levels of radioactivity in the Kara Sea are, however, very low and have at present a very small impact on man and on the marine ecosystem.

1. INTRODUCTION

In autumn 1990, information from different unofficial sources claimed that the former Soviet Union had dumped radioactive waste in the Barents Sea and the Kara Sea. In October 1992, a Russian Governmental commission was appointed and
was given the task of collecting and presenting information on the extent of dumping. The result was published in April 1993 in the form of a ‘White Book’ (White Book No. 2, 1993) on dumping of radioactive waste. This official Russian information largely confirmed the unofficial information. In addition, the report gave information about dumping of nuclear waste in the Pacific off the shores of Japan and Russia.

Information from the White Book shows that a total of 17 reactors were dumped in the Kara Sea. Seven of these reactors contained spent nuclear fuel. With one exception, all of the reactors were dumped in the fjords on the east coast of Novaya Zemlya at a depth of 20–50 m (one reactor from a nuclear submarine was dumped in the Novaya Zemlya trough at a depth of approximately 300 m). The reactors were dumped in the period from 1965 to 1988. Other solid radioactive waste was also dumped in the Kara Sea. Liquid radioactive waste was dumped in five defined areas in the Barents Sea. There were also discharges of liquid waste outside of these areas, mainly in the Kara Sea.

A joint Norwegian-Russian expert group was formed in 1992 and has carried out annual expeditions to the Kara Sea since 1992. The results from the 1992 and 1993 expeditions are reported in this paper. The aims of the expert group were: to obtain information on the handling, storage and, specifically, dumping of radioactive waste in the Arctic seas, to establish the actual concentrations of radionuclides in the Arctic marine ecosystem, especially the Kara Sea, to identify different sources contributing to radioactive contamination, and to assess the present and future consequences to man and the environment.

2. VISUAL INVESTIGATIONS OF DUMPING SITES AND SAMPLING

Water samples and sediments were collected at two stations in the Barents Sea and 11 stations in the Kara Sea in the 1992 expedition. During the 1993 expedition, three dumping sites for nuclear waste were investigated: the Tsivolki Fjord, the Stepovogo Fjord and an area in the open Kara Sea (the Novaya Zemlya trough). Dumped waste was localized and inspected in the Tsivolki Fjord and the Stepovogo Fjord, using side-scanning sonar and underwater cameras. In the Stepovogo Fjord, the dumped nuclear submarine No. 601, containing unloaded nuclear fuel, was localized. In the Tsivolki Fjord the most noticeable object was the hull of a large cargo ship. Bottom sediment samples were collected very close to the hull, using a bottom sediment sampler mounted on the remotely operated vehicle (ROV). During the searches in the investigated areas the sonar indicated the presence of a number of small objects on the sea bottom, but, owing to lack of time, it was impossible to localize the objects on the bottom with the ROV and to inspect them visually. Samples of water, sediment and biota were collected at nine stations and later analysed for several radionuclides (gamma emitters, $^{90}$Sr, $^{238}$Pu, $^{239}$Pu/$^{240}$Pu and $^{241}$Am).
2.1. Analytical processes

All samples were analysed by gamma spectrometry, using various high resolution germanium detectors (resolution in the range of 1.7–2.0 keV, efficiencies in the range of 10–30%). Low level liquid scintillation counting, low level anticoincidence counting and low level scintillation spectrometry and beta spectrometry were used for $^{90}$Sr analysis.

Determination of $^{238}$Pu and $^{239,240}$Pu was based on alpha spectrometry [1]. Determination of $^{99}$Tc was based on anion exchange chromatography followed by liquid–liquid extraction, electrodeposition and beta spectrometry [2].

3. RESULTS AND DISCUSSION

3.1. Radioactive contamination in the open Kara Sea

The 1993 results demonstrated that the level of radionuclide contamination in the Barents Sea and the Kara Sea can be explained by global fallout, releases from the Sellafield reprocessing plant, contributions from the rivers Ob and Yenisey, and fallout from the accident at the Chernobyl nuclear power plant. The observed levels of radioactive contamination are often lower than or similar to those of other areas; for example, the level of radioactive contamination in the Kara Sea is lower than the current levels in the Baltic Sea, the Black Sea and the Irish Sea.

3.2. Radioactive contamination at the dumping sites

The concentrations of $^{137}$Cs, $^{134}$Cs, $^{40}$Sr, $^{239,240}$Pu and $^{241}$Am in seawater samples from the 1993 expedition are given in Table I. The concentration of $^{137}$Cs varies from 4.5 to 31.9 Bq/m$^3$ at the dumping sites. The highest levels have been observed in the near bottom water samples from the inner part of the Stepovogo Fjord, Station 6 (26–32 Bq/m$^3$). Excluding these samples, the concentration of $^{137}$Cs varies from 4 to 14 Bq/m$^3$. In general, the concentration of $^{137}$Cs increases with depth and is highest in the near bottom water. The activity ratio of $^{134}$Cs/$^{137}$Cs varies from 0.007 to 0.027.

The concentration of $^{90}$Sr in sea water is in the range of 3–25 Bq/m$^3$. The highest levels of $^{90}$Sr have also been observed in the near bottom water samples from the inner part of the Stepovogo Fjord (24.6 Bq/m$^3$). Excluding these samples, the maximum concentration of $^{90}$Sr is 5.7 Bq/m$^3$. In the Tsivolki Fjord, the $^{90}$Sr concentration in sea water is highest in the surface water.

The mean activity ratio for $^{90}$Sr/$^{137}$Cs is 0.9 ± 0.3 for surface water and 0.5 ± 0.2 for near bottom water. In general, the activity ratio decreases with depth at all sites investigated.
TABLE I. CONCENTRATIONS OF $^{137}$Cs, $^{134}$Cs, $^{90}$Sr, $^{239,240}$Pu AND $^{241}$Am IN SEAWATER SAMPLES FROM DIFFERENT STATIONS AND DIFFERENT DEPTHS

<table>
<thead>
<tr>
<th>Station No.</th>
<th>Depth (m)</th>
<th>$^{137}$Cs (Bq/m$^3$)</th>
<th>$^{134}$Cs (Bq/m$^3$)</th>
<th>$^{90}$Sr (Bq/m$^3$)</th>
<th>$^{239,240}$Pu (mBq/m$^3$)</th>
<th>$^{241}$Am (mBq/m$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0</td>
<td>5.0 ± 0.2</td>
<td>0.12 ± 0.02</td>
<td>5.0 ± 1.3</td>
<td>4.0 ± 2</td>
<td>5.0</td>
</tr>
<tr>
<td></td>
<td>43</td>
<td>11.6 ± 0.3</td>
<td>0.27 ± 0.05</td>
<td>3.4 ± 0.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>55</td>
<td>6.9 ± 0.1</td>
<td></td>
<td>4.1 ± 1.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>0</td>
<td>5.1 ± 0.2</td>
<td>0.14 ± 0.03</td>
<td>5.4 ± 1.4</td>
<td>4.5 ± 0.7</td>
<td></td>
</tr>
<tr>
<td></td>
<td>28</td>
<td>8.2 ± 0.3</td>
<td></td>
<td>4.0 ± 1.1</td>
<td>7.0 ± 2</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>0</td>
<td>4.5 ± 0.1</td>
<td>0.14 ± 0.04</td>
<td>5.7 ± 1.1</td>
<td>9.8 ± 1</td>
<td></td>
</tr>
<tr>
<td></td>
<td>40</td>
<td>6.1 ± 0.1</td>
<td>0.21 ± 0.06</td>
<td>3.5 ± 0.7</td>
<td>26 ± 2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>70</td>
<td>10.0 ± 0.2</td>
<td>0.23 ± 0.07</td>
<td>3.5 ± 0.7</td>
<td>8.1 ± 0.9</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>0</td>
<td>4.9 ± 0.1</td>
<td>0.16 ± 0.04</td>
<td>4.8 ± 0.9</td>
<td>5.4 ± 0.8</td>
<td></td>
</tr>
<tr>
<td></td>
<td>40</td>
<td>6.1 ± 0.1</td>
<td>0.16 ± 0.02</td>
<td>3.4 ± 0.7</td>
<td>8.9 ± 2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>72</td>
<td>9.5 ± 0.3</td>
<td></td>
<td>3.6 ± 0.7</td>
<td>5.8 ± 0.4</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0</td>
<td>5.2 ± 0.1</td>
<td>0.14 ± 0.02</td>
<td>4.9 ± 1.0</td>
<td>2.3 ± 0.4</td>
<td>1.5 ± 1</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>8.2 ± 0.4</td>
<td>0.21 ± 0.02</td>
<td>4.0 ± 0.2</td>
<td>2.3 ± 1</td>
<td>7.7 ± 0.8</td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>6.3 ± 0.1</td>
<td></td>
<td>4.7 ± 0.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>0</td>
<td>5.8 ± 0.3</td>
<td>0.13 ± 0.00</td>
<td>5.2 ± 1.0</td>
<td>1.9 ± 0.4</td>
<td>2.0 ± 0.4</td>
</tr>
<tr>
<td></td>
<td>40</td>
<td>31.9 ± 1.3</td>
<td>0.21 ± 0.01</td>
<td>24.6 ± 1.4</td>
<td>6.0 ± 0.8</td>
<td></td>
</tr>
<tr>
<td></td>
<td>45</td>
<td>26.5 ± 0.5</td>
<td></td>
<td>24.3 ± 4.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>0</td>
<td>5.1 ± 0.3</td>
<td>0.12 ± 0.12</td>
<td>5.5 ± 0.8</td>
<td>2.9 ± 0.4</td>
<td></td>
</tr>
<tr>
<td></td>
<td>39</td>
<td>6.9 ± 0.3</td>
<td>0.16 ± 0.00</td>
<td>4.2 ± 0.7</td>
<td>18 ± 2</td>
<td>5.3 ± 0.8</td>
</tr>
<tr>
<td>8</td>
<td>0</td>
<td>5.7 ± 1.0</td>
<td>0.12 ± 0.01</td>
<td>3.1 ± 0.1</td>
<td>3.8 ± 0.6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>140</td>
<td>7.1 ± 0.3</td>
<td></td>
<td>2.9 ± 0.6</td>
<td>5.3 ± 0.6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>360</td>
<td>7.4 ± 0.4</td>
<td></td>
<td>2.8 ± 0.6</td>
<td>7.0 ± 1</td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>0</td>
<td>5.2 ± 0.2</td>
<td>0.08 ± 0.02</td>
<td>2.6 ± 0.6</td>
<td>2.6 ± 0.5</td>
<td></td>
</tr>
<tr>
<td></td>
<td>207</td>
<td>8.6 ± 0.4</td>
<td>0.23 ± 0.05</td>
<td>3.3 ± 0.7</td>
<td>6.1 ± 0.6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>340</td>
<td>13.5 ± 0.6</td>
<td>0.32 ± 0.19</td>
<td>2.9 ± 0.6</td>
<td>12 ± 1</td>
<td></td>
</tr>
</tbody>
</table>
The concentration of $^{239,240}$Pu varies from 1.9 to 26.0 mBq/m$^3$. In general, the activity level increases with depth, while no site specific differences have been observed. Cross-flow membrane filtration in near bottom water indicates that the colloidal fraction is small. The concentration of $^{238}$Pu is above the detection limit only at Station 4 (0.9 mBq/m$^3$).

Twelve seawater samples were subjected to $^{241}$Am analysis. Concentrations from 1.5 to 7.7 mBq/m$^3$ were obtained in four samples (Stations 5, 6, 7), while the detection limit was reached in the remaining eight samples.

The concentrations of $^{137}$Cs, $^{134}$Cs, $^{90}$Sr, $^{238}$Pu, $^{239,240}$Pu, $^{241}$Am and $^{60}$Co in the 0-1 cm layer of sediment profiles are given in Table II.

The $^{137}$Cs concentrations in sediments were highest at Station 6 in the Stepovogo Fjord (100–200 Bq/kg), while concentrations in the range of 5–25 Bq/kg were obtained at other sites. In general, $^{137}$Cs is enriched in the upper 5 cm of the sediment layer and decreases with depth. Caesium-134 was below the detection limit in the sediment profiles. However, traces of $^{134}$Cs in surface sediments were observed at Stations 1, 3, 4, 5 and 6.

The $^{90}$Sr concentrations in sediments were in the range of 0.2–4.2 Bq/kg. The highest concentrations were observed at Station 6 in the Stepovogo Fjord. The concentration of $^{90}$Sr in sediments decreases relatively smoothly with the depth of the profile.

Cobalt-60 was observed only at Stations 3, 4, 5 and 6, with the highest concentrations being in the upper sediment layer at Station 6. However, $^{60}$Co was also observed in large surface samples at Stations 1 and 9.

The $^{239,240}$Pu concentrations were in the range of 0.03–0.94 Bq/kg, with the highest values observed at Station 6 (Stepovogo Fjord) and Station 8 (Novaya Zemlya trough). The concentration level was similar to that observed in sediment samples from the open Kara Sea collected during the 1992 expedition (higher levels were observed at Stations 1–92 and 5–92) [3]. In general, the upper 4–5 cm of the sediment layer are enriched in $^{239,240}$Pu, and the concentration decreases with depth.

The levels of $^{238}$Pu and $^{239,240}$Pu were very low. In samples collected with the ROV sampler close to the hull of the dumped nuclear submarine in the Stepovogo Fjord (Station 5), $^{152}$Eu and $^{154}$Eu were also observed.

The Tsivolki Fjord

As seen from Table I (Stations 1–4), the radionuclide concentration levels in sea water from the inner and the outer Tsivolki Fjord are similar to those observed in the open Kara Sea.

The vertical distribution of $^{137}$Cs in sediments from the Tsivolki Fjord is similar to that observed in sediments from the open Kara Sea (1992); in the upper sediment layer, $^{137}$Cs is enriched and its concentration decreases with depth.
<table>
<thead>
<tr>
<th>Station No.</th>
<th>$^{137}\text{Cs}$</th>
<th>$^{134}\text{Cs}$</th>
<th>$^{90}\text{Sr}$</th>
<th>$^{60}\text{Co}$</th>
<th>$^{238}\text{Pu}$</th>
<th>$^{239,240}\text{Pu}$</th>
<th>$^{241}\text{Am}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>14.8 ± 0.6</td>
<td></td>
<td>0.8 ± 0.2</td>
<td>0.9 ± 0.2</td>
<td>n.d.</td>
<td>0.47 ± 0.09</td>
<td>0.13 ± 0.02</td>
</tr>
<tr>
<td>2</td>
<td>4.0 ± 0.4</td>
<td></td>
<td>0.4 ± 0.2</td>
<td>n.d.</td>
<td>0.03 ± 0.01</td>
<td>0.06 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>13.8 ± 1.4</td>
<td>&lt;1.1</td>
<td>1.0 ± 0.1</td>
<td>&lt;1.7</td>
<td>n.d.</td>
<td>0.26 ± 0.03</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>12.8 ± 0.8</td>
<td>&lt;2.4</td>
<td>0.7 ± 0.1</td>
<td>&lt;2.4</td>
<td>0.03 ± 0.01</td>
<td>0.29 ± 0.03</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>12.8 ± 0.5</td>
<td></td>
<td>0.4 ± 0.2</td>
<td>n.d.</td>
<td>0.37 ± 0.06</td>
<td>0.07 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>110 ± 1</td>
<td>&lt;2.8</td>
<td>2.0 ± 0.3</td>
<td>5.1 ± 0.9</td>
<td>0.03 ± 0.01</td>
<td>0.73 ± 0.05</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>20.0 ± 1.0</td>
<td></td>
<td>0.8 ± 0.2</td>
<td>&lt;3.2</td>
<td>0.05 ± 0.02</td>
<td>0.94 ± 0.08</td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>16.0 ± 1.4</td>
<td>&lt;4.2</td>
<td>0.8 ± 0.2</td>
<td>n.d.</td>
<td>0.32 ± 0.04</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

TABLE II. CONCENTRATIONS OF $^{137}\text{Cs}$, $^{134}\text{Cs}$, $^{90}\text{Sr}$, $^{60}\text{Co}$, $^{238}\text{Pu}$, $^{239,240}\text{Pu}$ AND $^{241}\text{Am}$ IN THE UPPER 1 cm LAYER OF SEDIMENT PROFILES (Bq/kg)
The most striking feature in the Tsivolki Fjord is, however, the presence of $^{60}$Co in the upper 4–5 cm layer (up to 19 Bq/kg dry weight). Because of the relatively short half-life of the nuclide, it is possible that $^{60}$Co originates from the dumped nuclear waste. However, a contribution from radionuclide fallout in the Novaya Zemlya region cannot be excluded.

**The Stepovogo Fjord**

The concentrations of $^{137}$Cs in surface water and near bottom water of the Stepovogo Fjord are given in Table I (Stations 5, 6 and 7).

In the near bottom water of the fjord, the concentrations of $^{137}$Cs (30 Bq/m$^3$) are significantly higher than those in the surface water, especially at Station 6, which is situated in the inner part of the fjord. In the outer part of the fjord, near the dumped nuclear submarine, and at the fjord inlet (Station 7) the $^{137}$Cs contamination levels in the near bottom water are only slightly higher than those in the surface water. The lower concentration of $^{137}$Cs in the surface water can be explained by dilution with melted ice or run-off from Novaya Zemlya. The increased levels in the near bottom water can be explained by leakage from nuclear material (especially at Station 6), and by resuspension of $^{137}$Cs from contaminated sediments or run-off from Novaya Zemlya.

In one sediment sample collected with the ROV sampler close to the hull of the dumped nuclear submarine, the concentration of $^{137}$Cs was more than one order of magnitude higher (206 Bq/kg) than that in all other sediment samples collected at Station 5. In this sample, $^{152}$Eu and $^{154}$Eu were also identified (34 ± 1 Bq/kg and 2 ± 1 Bq/kg). According to the estimated inventory of fission products in the submarine reactors [4], these nuclides were expected to be the dominating gamma emitters by the end of 1993. Although the ratio of $^{152}$Eu/$^{154}$Eu was lower in the sediment sample than had been expected from the estimates, the overall agreement in the radionuclide composition strongly suggests that the enhanced activity originates from the reactor of the dumped nuclear submarine.

In the inner part of the fjord, where exchange of the deep water with that in the outer part of the fjord is slow, owing to a shallow threshold, a much higher deposition of $^{137}$Cs was observed (about 300 Bq/m$^2$). In addition to the increased concentration of $^{137}$Cs, $^{60}$Co was also identified in the bottom sediments.

The increased levels of $^{137}$Cs and the presence of $^{60}$Co in sediments from the inner part of the fjord may be due to radionuclide run-off from Novaya Zemlya or to the release of radionuclides from dumped solid radioactive wastes. Unfortunately, along the coast, collection of soil samples was not permitted. However, one soil sample from the area was collected during the Russian Navy expedition and was analysed by scientists from the Navy and the Scientific Production Association "Typhoon". The concentration of $^{60}$Co in the upper 10 cm layer of soil appeared to be lower than the detection limit (<0.9 Bq/kg for the 0–5 cm soil layer,
<0.5 Bq/kg for the 5–10 cm layer). Since $^{60}$Co could not be identified in the soil, the source of $^{60}$Co can hardly be attributed to run-off. Even though the result is based on one soil sample from the shore of the Stepovogo Fjord, it is assumed that the increased contamination levels of $^{137}$Cs and $^{60}$Co in the inner part of the Stepovogo Fjord are most likely due to leakage from the dumped solid radioactive wastes.

The concentration level of $^{90}$Sr in the surface water (4.9–5.5 Bq/m$^3$) is similar to that in water from the western and southern parts of the Kara Sea (3–4 Bq/m$^3$) observed in 1992. The concentration is relatively uniform throughout the water column at Stations 5 and 7. At Station 6 in the inner part of the fjord, however, the concentration of $^{90}$Sr in the near bottom water increased by a factor of about five (25 Bq/m$^3$). Strontium-90 was also identified in sediments, especially at Station 6 (Table II). The source of $^{90}$Sr can hardly be explained by global fallout or run-off from land, since $^{90}$Sr is rather mobile in water and usually shows low retention in soils and sediments. The level of $^{90}$Sr in the near bottom water at Station 6 most likely originates, therefore, from leakage from dumped nuclear waste.

The concentration of $^{239,240}$Pu in surface water and near bottom water is within the range of 2–6 mBq/m$^3$. This level is somewhat lower than that observed in 1992 in the open Kara Sea (2–16 mBq/m$^3$). The vertical distribution is relatively uniform. On the basis of these results, the presence of $^{239,240}$Pu cannot be attributed to leakage from dumped nuclear waste.

The Novaya Zemlya trough

No dumped objects could be identified in the area, since the geographical co-ordinates of the dumped material given in the White Book were far too general and the available time did not permit an extensive search with the towed high frequency side-scanning sonar. The concentration levels are similar to those observed in the open Kara Sea in 1992. In the surface sediment layer (0–3 cm) the concentration of $^{137}$Cs is within 7–30 Bq/kg d.w., while a range of 2–53 Bq/kg d.w. was obtained in 1992. Owing to the limited number of samples from the Novaya Zemlya trough, it was not possible to find an indication of leakage from dumped nuclear waste.

3.3. Analyses of biota

The concentrations of $^{137}$Cs in fish ($Boreogadus saida$ (polar cod), $Liparis fabricii$) were $5 \pm 1$ and 1, respectively. Seaweed, mainly $Fucus evanescens$, $Laminaria digitata$ and $Laminaria saccharina$, was collected at 4–8 m depth at Stations 1 and 2 in the Tsivolki Fjord (salinity 14–15%$o$) and at Stations 5 and 6 in the Stepovogo Fjord (salinity 17–20%$o$). The uptake of radionuclides by seaweed
<table>
<thead>
<tr>
<th>Station</th>
<th>Species</th>
<th>( ^{137}\text{Cs} ) (Bq/kg)</th>
<th>( ^{60}\text{Co} ) (Bq/kg)</th>
<th>( ^{90}\text{Sr} ) (Bq/kg)</th>
<th>( ^{239,240}\text{Pu} ) (mBq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>St.1(3)</td>
<td>( \text{F. evanescens} )</td>
<td>1.7 ± 0.4</td>
<td>1.5 ± 0.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.1(2)</td>
<td>( \text{F. evanescens} )</td>
<td>&lt;2.2</td>
<td>&lt;2.6</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.1(2)</td>
<td>( \text{F. evanescens} )</td>
<td>1.6 ± 0.4</td>
<td>&lt;1.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.1(4)</td>
<td>( \text{F. evanescens} )</td>
<td>2.7 ± 0.4</td>
<td>&lt;0.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.1(2)</td>
<td>( \text{L. digitata} )</td>
<td>5.1 ± 0.5</td>
<td>&lt;1.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.1(3)</td>
<td>( \text{L. digitata} )</td>
<td>2.8 ± 0.3</td>
<td>&lt;1.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.2(1)a</td>
<td>( \text{L. digitata} )</td>
<td>&lt;2.4</td>
<td>&lt;3.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.2(2)a</td>
<td>( \text{L. digitata} )</td>
<td>4.1 ± 0.6</td>
<td>&lt;2.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.2(2)b</td>
<td>( \text{L. digitata} )</td>
<td>&lt;1.7</td>
<td>&lt;1.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.2, mix</td>
<td>( \text{L. digitata} )</td>
<td>2.4 ± 0.3</td>
<td>&lt;1.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.2, mix</td>
<td>( \text{L. digitata} )</td>
<td>2.7 ± 0.2</td>
<td>&lt;0.6</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.2(1)c</td>
<td>( \text{L. digitata, L. saccharina} )</td>
<td>4.0 ± 1.1</td>
<td>4.6 ± 1.2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.1(2)</td>
<td>( \text{L. digitata leaves, some L. saccharina} )</td>
<td>2.7 ± 0.5</td>
<td>&lt;0.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.1(2)a</td>
<td>( \text{L. digitata leaves, some L. saccharina} )</td>
<td>&lt;1.6</td>
<td>&lt;2.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.1(3)</td>
<td>( \text{L. digitata leaves, some L. saccharina} )</td>
<td>6.8 ± 0.5</td>
<td>&lt;1.7</td>
<td>2.4 ± 0.2</td>
<td>63 ± 7</td>
</tr>
<tr>
<td>St.1(3)</td>
<td>( \text{L. digitata lower stem, some L. saccharina} )</td>
<td>8.1 ± 0.7</td>
<td>&lt;2.0</td>
<td>2.1 ± 0.2</td>
<td>40 ± 20</td>
</tr>
<tr>
<td>St.1(4)</td>
<td>( \text{L. digitata stems} )</td>
<td>6.2 ± 0.6</td>
<td>&lt;1.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.1(3)</td>
<td>( \text{L. digitata stems, some L. saccharina} )</td>
<td>4.3 ± 0.3</td>
<td>&lt;1.2</td>
<td>3.2 ± 0.2</td>
<td>64 ± 9</td>
</tr>
<tr>
<td>St.1(4)</td>
<td>( \text{L. digitata, L. saccharina} )</td>
<td>2.8 ± 0.3</td>
<td>&lt;0.8</td>
<td>5.4 ± 0.3</td>
<td>58 ± 9</td>
</tr>
<tr>
<td>St.2(1)b</td>
<td>( \text{L. saccharina} )</td>
<td>&lt;2.2</td>
<td>&lt;2.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.2(2)a</td>
<td>( \text{L. saccharina} )</td>
<td>1.1 ± 0.3</td>
<td>&lt;0.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>St.5(1)</td>
<td>( \text{F. evanescens} )</td>
<td>3.4 ± 0.2</td>
<td>1.1 ± 0.3</td>
<td>3.9 ± 0.3</td>
<td>400 ± 20</td>
</tr>
<tr>
<td>St.6(1)</td>
<td>( \text{F. evanescens} )</td>
<td>4.1 ± 0.3</td>
<td>&lt;0.9</td>
<td>3.5 ± 0.2</td>
<td>440 ± 50</td>
</tr>
<tr>
<td>St.5(1)</td>
<td>( \text{L. digitata} )</td>
<td>2.5 ± 0.4</td>
<td>&lt;1.3</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
depends on several factors, such as time of sampling, salinity and temperature conditions, and dominantly the growth pattern. The climatic conditions and the salinity are almost the same in the two fjords and, therefore, the results can be compared.

The concentration of $^{137}\text{Cs}$ varies from 1.1 to 8.1 Bq/kg d.w. (Table III). The $^{137}\text{Cs}$ concentration in Fucus samples from the Stepovogo Fjord is higher than that from the Tsivolki Fjord. However, one sample of Laminaria from Stepovogo shows a lower $^{137}\text{Cs}$ concentration than most of the about 20 Laminaria samples from Tsivolki. Cobalt-60 is observed in three samples collected at Station 1 (Fucus, 1.5 Bq/kg d.w.), Station 5 (Fucus, 1.1 Bq/kg d.w.) and Station 2 (Laminaria, 4.6 Bq/kg d.w.). Slightly higher $^{239,240}\text{Pu}$ concentrations are found in the samples from the Stepovogo Fjord than in the samples from the Tsivolki Fjord (0.4 Bq/kg and 0.06 Bq/kg, respectively).

The seaweed may reflect the actual seawater concentrations of anthropogenic radionuclides if they are present in a retainable form. The concentration factor for seaweed is species dependent, and for Fucus vesiculosus/sea water (Bq/kg d.w. per Bq/L) the factor is 100-200 times higher for $^{60}\text{Co}$ than for $^{137}\text{Cs}$ [5]. The concentration factors for Fucus and Laminaria for arctic climatic conditions are, however, not known.

4. CONCLUSIONS

Elevated levels of $^{90}\text{Sr}$ and $^{137}\text{Cs}$ and the presence of $^{60}\text{Co}$ have been observed in the inner part of the Stepovogo Fjord and in one sample collected close to the hull of a dumped nuclear submarine in the Stepovogo Fjord; $^{60}\text{Co}$ has also been observed in the Tsivolki Fjord. This radioactive contamination most likely originates from the dumped radioactive material and is probably due to leaching from the waste.

The enhanced contamination levels caused by dumped nuclear waste are, however, low and are restricted to small areas. Thus, radiation doses from the existing contamination would be negligible. The radioactive contamination levels outside these areas are similar to the activity levels in the open Kara Sea.

REFERENCES


THE INTERNATIONAL ARCTIC SEAS ASSESSMENT PROJECT (IASAP): INTERIM PROGRESS REPORT

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Abstract

THE INTERNATIONAL ARCTIC SEAS ASSESSMENT PROJECT (IASAP): INTERIM PROGRESS REPORT.

The International Arctic Seas Assessment Project (IASAP) was established in 1993 to address concern over the potential health and environmental impacts of the dumped radioactive wastes in the shallow waters of the Arctic seas. The work is being carried out as part of the IAEA's responsibilities with regard to the London Convention 1972. The project will last for four years and is run by the IAEA in co-operation with the Norwegian and Russian Governments, and with the involvement, through the IAEA, of experts from about 15 IAEA Member States. The objectives of the project are: (1) to assess the risks to human health and to the environment associated with the radioactive wastes dumped in the Kara Sea and the Barents Sea, and (2) to examine possible remedial actions related to the dumped wastes and to advise on whether these actions are necessary and justified. The project has been organized into five principal working areas: (i) source terms, (ii) existing environmental concentrations, (iii) transfer mechanisms and models, (iv) impact assessment and (v) remedial measures. Progress of IASAP is reviewed each year by a group of senior scientists. This group is also responsible for preparing the final report of IASAP and the recommendations for presentation to Contracting Parties to the London Convention in 1996.

1. INTRODUCTION

In 1992, the news that the former Soviet Union had, for over three decades, dumped large amounts of high level radioactive wastes in the shallow waters of the Arctic seas caused widespread concern, especially in countries with Arctic coastlines.

The IAEA responded by making a proposal for an international study to assess the health and environmental implications of the dumping. The proposal received support from the Contracting Parties to the Convention on the Prevention of Marine Pollution by Dumping of Wastes and other Matter (London Convention 1972); in addition, the Parties requested that the study should include consideration of possible remedial actions. Therefore, the objectives of the study, which became known as the International Arctic Seas Assessment Project (IASAP), are:
To assess the risks to human health and to the environment associated with the radioactive wastes dumped in the Kara Sea and the Barents Sea; and

To examine possible remedial actions related to the dumped wastes and to advise on whether these actions are necessary and justified.

The study was launched at a meeting jointly organized by the IAEA and the Norwegian and Russian Governments in Oslo in February 1993. The results and conclusions of the project will be reported to Contracting Parties to the London Convention in 1996.

2. BACKGROUND ON THE DUMPED WASTES

In May 1993, the Russian Federation provided information to the IAEA about the high and low level radioactive wastes dumped in the Arctic seas and in the north-east Pacific during the years 1959-1992 [1]. According to this report the total amount of radioactivity of wastes dumped in the Arctic seas was more than 90 PBq. The items dumped included six nuclear submarine reactors and a shielding assembly from an ice-breaker reactor containing spent fuel comprising a total of 85 PBq; ten reactors (without fuel) containing 3.7 PBq; liquid low level waste containing 0.9 PBq; and solid intermediate and low level waste containing 0.6 PBq. The packaged and unpackaged solid waste and the nuclear reactors were dumped in the Kara Sea, in the shallow bays of Novaya Zemlya, where the depths of the dumping sites range from 12 to 135 m, and in the Novaya Zemlya trough, at a depth of 300 m. The liquid low level waste was discharged in the open Barents Sea and Kara Sea. Figure 1 shows the locations of the Arctic dumping sites.

3. EVALUATION OF THE CURRENT ENVIRONMENTAL SITUATION

The Joint Norwegian-Russian expert group was established in 1992 in order to investigate radioactive contamination due to dumped nuclear wastes in the Barents Sea and the Kara Sea. It arranged exploratory cruises to the dumping areas, with the participation of a scientist from the IAEA Marine Environment Laboratory (IAEA-MEL), in 1992, 1993 and 1994. All four sites where spent nuclear fuel was dumped were visited by the cruises of the expert group, but only some of the objects were successfully localized. The group took samples, made measurements and used a side-scanning sonar and video camera in an attempt to identify and examine the wastes. The results obtained during the cruises did not indicate any significant radioactive contamination at the dumping sites, although the levels in the vicinity of some dumped objects were slightly elevated compared with those observed at other locations [2-4].
FIG. 1. Sea disposal of radioactive waste in the Arctic seas by the former Soviet Union and the Russian Federation.
While it appears that at present there are no significant regional and global effects from the dumped wastes, gradual deterioration of the waste containments could lead to impacts in the future. These could occur through contamination of the marine food-chain, possibly resulting in radiation exposure of humans through consumption of fish and other marine foodstuffs. Since the wastes are lying in shallow waters, the possibility of radiation exposure by other routes, such as movement and transport of waste packages by natural events (ice or storm action) or human actions, cannot be ruled out. The time-scales for consideration are very long (up to tens of thousands of years) and, therefore, the possible impact of climatic change has also to be taken into account. In order to provide solutions to these problems it is necessary to have a thorough understanding of the present and future physical, chemical and biological characteristics of the environment surrounding the wastes and of the wastes themselves.

Preliminary assessments have indicated that, even under the most pessimistic release conditions, the wastes would not have a significant global radiological impact. However, for evaluating the possible risks at local and regional scales, considerably more information will be required than was generally available when the project was launched [4, 5]. The IASAP project was established to find solutions to these and other related problems.

4. PROGRESS IN IASAP

The project is organized in five working areas:

— Source terms
— Existing environmental concentrations
— Transfer mechanisms and models
— Impact assessment
— Remedial measures.

Progress made in all working areas of IASAP is reviewed each year by a group of senior scientists (IASAP Advisory Group Meeting).

4.1. Source terms

The aim of the Source Term Working Group is to determine the information on the wastes required for impact assessment calculations. This involves a knowledge of the waste form and of its likely behaviour with time in the marine environment. The efforts of the Group have been focused on the dumped objects containing spent fuel which pose the highest potential risk.

The official information on dumped wastes provided by the Russian Federation in May 1993 did not include information on the radionuclide composition of the
wastes or on the characteristics of the fuel in the different types of dumped reactors. In order to obtain more detailed information, it has been necessary to investigate the archives of the former Soviet Union and to reconstruct the history of the reactor fuel prior to dumping [6–8].

In January 1994, a detailed inventory of the radionuclide composition and information on the structure of the dumped reactor compartment of the commercial nuclear ice-breaker ‘Lenin’ was obtained [9] (OK-150 in Table I). In July 1994, the Russian authorities declassified essential details of the structure, the operational history and characteristics of the fuel of the dumped submarine reactors. As a consequence, the corresponding radionuclide inventories of the lead/bismuth cooled submarine reactor (No. 601 in Table I) [7] and of the water cooled submarine reactors (Nos 254, 260, 285, 421, 538 and 901 in Table I) [8] were made available to IASAP.

The total activity of the dumped reactors (with and without nuclear fuel) at the time of dumping is now estimated to be 37 PBq; the first estimate, provided in May 1993 by the Russian Federation, was 89 PBq. Because of radioactive decay, the total activity of the dumped reactors at the present time is only 4.7 PBq (Table I). Figure 2 illustrates the development with time of the total amount of radioactivity of the dumped nuclear fuel, taking into account radioactive decay. Actually, the maximum peak of radioactivity of the dumped material, 25 PBq, was reached in 1967, when the fuel assembly of one of the reactors of the ice-breaker ‘Lenin’, containing part of the spent fuel, was dumped.

For both impact assessment purposes and evaluation of the feasibility of possible remedial measures it is necessary to have information on protective barriers provided for the dumped reactors by their initial construction or through preparations prior to dumping. This information has been obtained through contracts with Russian institutes [6–8].

Fuel was removed from ten of the reactors prior to dumping. Those reactors that contained spent fuel when they were dumped (six reactors) had usually met with an accident prior to dumping in which the fuel was damaged. The dumping of the reactors took place by four principal means: (i) Most of the dumped submarine reactors were contained in their compartments; the reactors were filled with a special polymer — furfural. (ii) In some cases the reactors were taken out of the compartment and placed in a metal box shielded with lead. (iii) In the case of the lead-bismuth cooled reactors, the submarine compartment was filled with furfural and bitumen, and the whole submarine was dumped (No. 601). In this case, the solidified liquid metal coolant formed an additional protective barrier. (iv) The dumped components of the nuclear ice-breaker ‘Lenin’ included a reactor compartment with three reactor vessels from which the fuel was removed and which were filled with furfural. About half of the fuel from one of the reactors was dumped in a separate metal lined concrete box which was also filled with the sealant furfural.
TABLE I. DATA ON THE NUCLEAR REACTORS DUMPED NEAR NOVAYA ZEMLYA

<table>
<thead>
<tr>
<th>Site</th>
<th>Year of dumping</th>
<th>Depth of dumping</th>
<th>Factory number</th>
<th>Dumped unit</th>
<th>Number of reactors</th>
<th>Total activity (PBq)</th>
<th>Initial data (Ref. [1])</th>
<th>Further studies (Refs [6, 7, 8])</th>
<th>At time of dumping</th>
<th>At time of dumping</th>
<th>1993/1994</th>
</tr>
</thead>
<tbody>
<tr>
<td>Abrosimov Fjord</td>
<td>1965</td>
<td>20 (10-15)</td>
<td>No. 285</td>
<td>Reactor compartment</td>
<td>1</td>
<td>29.6</td>
<td>11.6</td>
<td>0.655</td>
<td>1965</td>
<td>20 (10-15)</td>
<td>29.6</td>
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<tr>
<td></td>
<td></td>
<td>20 (10-15)</td>
<td>No. 901</td>
<td>Reactor compartment</td>
<td>-</td>
<td>14.8</td>
<td>2.95</td>
<td>0.727</td>
<td>1965</td>
<td>20 (10-15)</td>
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<td></td>
<td>20</td>
<td>No. 254</td>
<td>Reactor compartment</td>
<td>2</td>
<td>-</td>
<td>b</td>
<td>0.093</td>
<td>1965</td>
<td>20</td>
<td>-</td>
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<tr>
<td></td>
<td>1966</td>
<td>20</td>
<td>No. 260</td>
<td>Reactor compartment</td>
<td>2</td>
<td>-</td>
<td>b</td>
<td>0.044</td>
<td>1965</td>
<td>20</td>
<td>-</td>
</tr>
<tr>
<td>Tsivolki Fjord</td>
<td>1967</td>
<td>50</td>
<td>OK-150</td>
<td>Reactor compartment and box</td>
<td>3</td>
<td>-</td>
<td>b</td>
<td>19.5</td>
<td>1993/1994</td>
<td></td>
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</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>containing fuel</td>
<td>Box containing</td>
<td>0.6</td>
<td>3.7</td>
<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>East Novaya Zemlya</td>
<td>1972</td>
<td>300</td>
<td>No. 421</td>
<td>Reactor</td>
<td>-</td>
<td>1</td>
<td>29.6</td>
<td>1.05</td>
<td>0.293</td>
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<tr>
<td>Trough</td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>Stepovogo Fjord</td>
<td>1981</td>
<td>50 (30)</td>
<td>No. 601</td>
<td>Submarine</td>
<td>-</td>
<td>2</td>
<td>7.4</td>
<td>1.72</td>
<td>0.838</td>
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<tr>
<td>Techeniye Fjord</td>
<td>1988</td>
<td>35-40</td>
<td>No. 538</td>
<td>Reactors</td>
<td>2</td>
<td>-</td>
<td>b</td>
<td>0.006</td>
<td>0.005</td>
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<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>10</td>
<td>6.6</td>
<td>89</td>
<td>37</td>
<td>4.7</td>
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<td></td>
</tr>
</tbody>
</table>

* Values from Ref. [1]; values in brackets are from Refs [2, 3].

b Reactors without spent fuel, not more than 3.7 PBq total.
FIG. 2. Development with time of the total amount of radioactivity of the nuclear fuel dumped into the Kara Sea.

On the basis of an analysis of the weak points of the protective barriers the Source Term Working Group prepared sets of possible time patterns for radionuclide release and release rates.

Further studies are needed before more accurate predictions of releases of the different radionuclides from each of the dumped reactors can be made. In this context, information on the physical and chemical characteristics of furfural and of its stability against radiation, heat, saline water, etc. would be valuable. In co-operation with the IASAP project, a study on the resistant properties of furfural was started early in 1995 as a Russian–US bilateral project.

The findings of the Norwegian–Russian exploratory cruises, for example the results of visual investigation of reactors by means of underwater cameras, in situ radiometric measurements, and water and sediment sampling and analyses are very important in the evaluation of the potential release rates. It is planned that the video tapes of the sunken objects taken during the exploratory cruises will be carefully analysed, as part of the Norwegian–Russian co-operation, by experts on naval reactor and submarine design and on problems of corrosion.

4.2. Existing environmental concentrations

Information on the levels of radioactive contamination in the target area and in other areas of the Arctic seas is being collected as input to the global marine radio-
activity database (GLOMARD) of the IAEA-MEL. The database is designed to provide up-to-date information on radionuclide levels in sea water, sediment and biota. It will enable evaluation of nuclide ratios, identification of the different contributions to radioactivity in the region, investigation of time trends and calculation of inventories. All available radionuclide data from the Arctic seas have been entered into the database. The first report with a preliminary evaluation of the existing radionuclide data will be available at the end of 1995.

4.3. Transfer mechanisms and models

Laboratories in Denmark, Japan, Netherlands, Russia, Switzerland, the United Kingdom, and the IAEA-MEL are participating in the co-ordinated research programme (CRP) entitled "Modelling of the radiological impact of waste dumping in the Arctic seas". The objective of the CRP is to develop realistic and reliable assessment models for the Arctic sea areas and to co-ordinate the efforts of different laboratories in the field. The final results of the modelling exercise will form the basis of the assessment to be presented to Contracting Parties to the London Convention in 1996.

A staged approach to the final modelling assessment is being taken. The participating modelling groups first analysed scenarios based on assumed unit releases into a simplified environment. Subsequently, the initial benchmark scenarios were supplemented with improved information on the oceanography, sedimentology and biology of the Kara Sea and the Barents Sea; this information was made available to the IASAP by Russian institutes [9–11].

At the same time, each modelling group participating in the CRP is in the process of developing its models, using the improved information that is gradually becoming available from the target area. The available models differ, inter alia, in their spatial and temporal resolutions. Thus, the results from the different models will be taken for different particular end-points. For example, the results from models with greater spatial resolution will be used for calculations of impacts on critical groups. As the next step in scenario development, release rates based on the results of the Source Term Working Group will be included in the scenarios.

4.4. Impact assessment

Impact assessment calculations will be carried out on the basis of the concentration fields prepared by the Modelling Working Group, using appropriate environmental transfer factors and demographic data.

Radiation doses at various times in the future will be predicted for local, regional and global populations, taking into account both average consumers and those individuals whose diet includes considerable amounts of seafood. The assessments will also include estimates of radiation doses to local fauna such as marine mammals.
Information on radionuclide concentration factors for biota and on distribution factors between sediment and water relevant to Arctic conditions is being acquired through the projects of the IAEA-MEL and other laboratories. The radiological, physical and chemical measurements made on samples taken from the Arctic area are being analysed and compiled, and a literature review is being conducted. For radionuclides and biological species for which appropriate local data are not available, the applicability of concentration and distribution factors derived at moderate latitudes will have to be considered.

4.5. Remedial measures

The Contracting Parties to the London Convention requested the IAEA to consider possible remedial actions in relation to the dumped wastes and to consider the feasibility of such actions. A group of technical experts was convened early in 1995 to consider possible remedial measures, mainly from the standpoint of technical feasibility. While the findings of the group must be considered preliminary at this stage, certain general conclusions can be drawn:

(a) Objects containing spent nuclear fuel should be considered as the prime potential subjects of remediation;
(b) Well developed techniques are available for in situ remedial measures such as capping and underwater burial;
(c) If a remedial measure involving transport of wastes were to be chosen, underwater transport is a noteworthy option.

This subject will be discussed again at meetings in 1995 and 1996, and special attention will be given to the radiological impact of the various possible remedial options. One of the waste management options which has to be considered is leaving the wastes as they are, without remediation. A decision to carry out remediation must be based, at least partly, on the judgement that potential future radiological risks from the dumped wastes are unacceptable. In any case, the IASAP project can address only technical aspects such as the engineering feasibility and the radiological implications of different options; and it can provide a technical basis for deciding whether remediation is necessary or not, but the decision itself must be taken by relevant national authorities.

REFERENCES


REVIEW OF THE DISCHARGE HISTORY AND POPULATION DOSES FROM THE SELLAFIELD REPROCESSING PLANT IN CUMBRIA, UNITED KINGDOM

The Sellafield Environmental Assessment Model (SEAM)

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United Kingdom

Abstract


A complete discharge history for liquid and aerial effluents has been established for the Sellafield nuclear fuel reprocessing plant. The discharge history was constructed from a detailed search of archived documentation and was validated by comparison with environmental concentration and environmental inventory measurements through use of the Sellafield Environmental Assessment Model (SEAM). In a few cases the cumulative discharge had to be estimated from environmental inventories through the model, rather than from documentary records. Whilst the previously published liquid effluent discharge chronology was essentially unchanged, the aerial effluent chronology was revised, with the cumulative discharge of many nuclides increased substantially. The revised chronologies together with SEAM account very well for the temporal and spatial distribution of radionuclides in the environment of Sellafield. Doses to the population in the nearby village of Seascale have decreased marginally since earlier assessments, despite the increase in assumed atmospheric emissions. This reflects the fact that environmental concentrations were a constraint on the earlier assessments, and also that the dosimetric models of the International Commission on Radiological Protection for some radionuclides have been revised.

1. INTRODUCTION

In the last decade, the existence of a higher than average rate of childhood leukaemia in young people from the village of Seascale in Cumbria has led to speculation that radioactive discharges from the nearby reprocessing plant at Sellafield may be a causative factor [1], even though the calculated doses are too small by
around two orders of magnitude to account for the apparent elevation of leukaemia risk observed in epidemiological studies [2, 3].

However, these estimates of historical doses from discharge from the plant have relied largely on calculations based on recorded discharges and conventional environmental models. This has left open the question of whether the recorded discharges, particularly in the earlier years of plant operation, might have been seriously underestimated.

In 1989 and 1990, writs were served on British Nuclear Fuels plc (BNFL) which commenced civil actions on behalf of a number of families from West Cumbria whose children had suffered from leukaemia and related conditions. The subsequent preparation of technical evidence involved the review of new experimental studies on discharges carried out since 1986, a further and very extensive review of historic documents disclosed as part of the legal process, and also a review of environmental monitoring data and their correlation with discharges from the site.

This paper comprises a summary account of the resulting reassessment of discharge chronologies and the associated reassessment of doses to the local population. A fuller account is published elsewhere [4].

2. APPROACH TO REVIEWING DISCHARGE CHRONOLOGIES

The objective of the work described in this paper was to produce a revised discharge chronology for the Sellafield site, and the associated assessment of radiation doses to the local population, in such a way as to minimize the uncertainties associated with the earlier estimates. This involved bringing together as much information as possible on the operation of the plant, in particular discharge records and associated documentation, information on the accuracy and efficiency of the effluent sampling arrangements, and environmental monitoring information.

Recorded discharges were reviewed against all the available documentary evidence, and amendments are made to the chronology, if necessary. The discharge chronologies so derived were then validated against measured concentrations in the environment using a predictive model. Only when the measured environmental concentrations could be adequately accounted for were the discharge chronologies considered to be reliable.

3. THE SELLAFIELD ENVIRONMENTAL ASSESSMENT MODEL

An essential step in the reassessment was the development of the Sellafield Environmental Assessment Model (SEAM), which was used both to calculate doses and to build confidence in the discharge chronology from recorded measurements of environmental concentrations and current assessments of environmental inventories.
This model is described more fully elsewhere [5]. It brings together established models of atmospheric dispersion and deposition [6, 7], terrestrial food-chains [8], marine dispersion and concentration in marine biota [9, 10], and the sea to land transfer of radionuclides [11]. Environmental measurements from a wide variety of sources have been compared with values calculated from the discharge chronology and from SEAM in order both to validate the model and to build confidence in the discharge chronology.

4. CHRONOLOGY FOR LIQUID DISCHARGES

Review of documentary evidence revealed no reasons for amending significantly the chronology of liquid effluent discharges published previously [3].

Records of discharges of radioactivity to the sea have been made since 1952. During the 1950s, radiochemical determinations were carried out for a range of radionuclides: \(^{89}\text{Sr}\), \(^{90}\text{Sr}\), \(^{95}\text{Zr}\), \(^{95}\text{Nb}\), \(^{103}\text{Ru}\), \(^{106}\text{Ru}\), \(^{137}\text{Cs}\), \(^{144}\text{Ce}\), uranium, plutonium, 'total beta' and 'yttrium and rare earths' [12, 13]. Polonium-210 was also reported during the period 1955-1963 and \(^{230}\text{Th}\) during 1955 and 1956.

Gamma spectrometric methods were introduced in 1962. Determinations of additional specific nuclides in liquid effluent were made: \(^{241}\text{Am}\) from 1968; \(^{241}\text{Pu}\) from 1972; and \(^{3}\text{H}\) from 1963.

In 1978, the analytical schedule for liquid effluents was expanded significantly to include a number of specific nuclides, including \(^{35}\text{S}\), \(^{54}\text{Mn}\), \(^{55}\text{Fe}\), \(^{60}\text{Co}\), \(^{63}\text{Ni}\), \(^{65}\text{Zn}\), \(^{99}\text{Tc}\), \(^{110}\text{Ag}\), \(^{125}\text{Sb}\), \(^{152}\text{Eu}\), \(^{154}\text{Eu}\), \(^{155}\text{Eu}\), \(^{237}\text{Np}\), \(^{242}\text{Cm}\) and \(^{243,244}\text{Cm}\). In that year, separate determination of the plutonium isotopes \(^{238}\text{Pu}\) and \(^{239,240}\text{Pu}\) began (previously, these nuclides had been included in the plutonium alpha determinations).

In the earlier discharge chronology prepared between 1984 and 1986 it was necessary to fill some gaps for the earlier years by extrapolation. However, such extrapolations had been made with reasonable confidence, based, for example, on available measurements of 'total alpha' and 'total beta' discharges, or based on available measurements of a radionuclide closely related to that for which the discharges were being estimated.

The only subsequent refinement in the data is the separation of plutonium alpha values for the period 1952-1977 into separate values for \(^{238}\text{Pu}\) and \(^{239,240}\text{Pu}\) [9].

The discharge chronology, taking account of the above refinement, is set out in Table I for the most radiologically significant nuclides [3].

5. CONFIRMATION OF THE LIQUID DISCHARGE CHRONOLOGY

In principle, environmental measurements should permit confirmation of the discharge chronology if they can be related to discharge levels by means of a suitable
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Notes: Additional authorized $^3$H discharges averaging 5.0E+15 Bq/a, due to transfer of consignments of tritiated waste from other locations, occurred in the late 1960s and early 1970s. Discharges of $^{230}$Th in 1955 and 1956 were 1.9E+12 Bq and 1.8E+12 Bq, respectively. Small amounts of $\alpha$ and $\beta\gamma$ emitters were discharged in 1951.
environmental model. We have therefore used SEAM [5] to correlate the discharge rates with available environmental data.

The modules of SEAM dealing with liquid effluents describe long term dispersion of marine discharges through the Irish Sea and calculate radionuclide concentrations in solution, and in suspended sediment and bed sediment phases. They incorporate an established model of the Irish Sea [9, 14].

There are two main sources of environmental monitoring data for the Sellafield area: the routine programmes operated by BNFL [15] and previously the United Kingdom Atomic Energy Authority (UKAEA); and the programme operated by the Ministry of Agriculture, Fisheries and Food (MAFF) [16]. In addition, an analysis of a sediment core obtained from a disused dock basin in Maryport harbour contains a chronology of radionuclide concentrations in suspended sediments extending back to the 1950s [17].

5.1. Activity in marine biota

SEAM is used to calculate radionuclide concentrations in marine biota by applying concentration factors (CFs) to the calculated radionuclide concentrations (dissolved phase) in sea water. Default values of CFs assumed by the model are those recommended by the International Atomic Energy Agency [10].

We found that the most complete and reliable time series measurements on marine biota were for the edible seaweed Porphyra umbilicalis (laverweed), which has been sampled and analysed for $^{106}$Ru and other radionuclides since 1951, and for fish, which have been sampled and analysed for $^{137}$Cs and other radionuclides since the 1960s. Agreement between calculated and measured concentrations is good (Figs 1 and 2), so confirming the discharge chronology for these radionuclides.

For the alpha emitting radionuclides $^{238}$Pu, $^{239,240}$Pu and $^{241}$Am, measurements in the marine environment only commenced in the mid-1970s with measurements on Porphyra and molluscan shellfish (Littorina littorea, Mytilus edulis). Whilst the time series from the 1970s can be well reproduced [4], the discharge chronologies from earlier years cannot be confirmed by these measurements.

5.2. Maryport sediment core

The chronology established for a sediment core obtained from a harbour basin in Maryport, about 30 km north of Sellafield [17], enables confirmation of the discharge chronologies of plutonium and americium back to the late 1950s. Concentrations of $^{239,240}$Pu and $^{241}$Am in suspended sediments were calculated using SEAM for the location nearest to Maryport, assuming distribution coefficient ($K_d$) values of $3 \times 10^3$ and $2 \times 10^6$, respectively [14]. Corrections were made to the $^{241}$Am concentrations to allow for ingrowth from $^{241}$Pu subsequent to deposition of the sediment. The measured and calculated values for the different core sections
**FIG. 1.** Calculated concentrations of $^{106}$Ru in Porphyra umbilicalis from West Cumbria compared with environmental surveillance measurements.

**FIG. 2.** Calculated concentrations of $^{137}$Cs in fish from the West Cumbrian coast compared with environmental surveillance measurements.
FIG. 3. Calculated concentrations of $^{238}\text{Pu} + ^{239,240}\text{Pu}$ and $^{241}\text{Am}$ in suspended marine sediments, compared with levels in the dated sediment core obtained from Maryport harbour by Kershaw et al. [17].

compare well (Fig. 3), thus confirming the discharge chronologies for these radionuclides.

5.3. Irish Sea inventory

Because plutonium and americium partition strongly into the sediment phase, the north-eastern Irish Sea contains in its sea-bed and intertidal sediments a large fraction of the cumulative discharge of these radionuclides. Model predictions for the sediment inventory and estimates from the many existing surveys of intertidal and sea-bed sediments which have been compared agree quite closely [18], giving additional confidence in the recorded discharge chronology.

Overall, the above comparisons give additional confidence in the chronology of the most radiologically significant radionuclides discharged throughout the site’s history. The reproduction of concentrations of $^{106}\text{Ru}$ in Porphyra and of $^{137}\text{Cs}$ in fish, including reproduction of temporal trends throughout most of the period of site operations, gives reassurance that no releases of major radiological significance have gone undetected.

Similarly, the Maryport core data give confidence in the $^{239,240}\text{Pu}$ and $^{241}\text{Am}$ discharge chronologies, again in terms of both absolute concentrations in sediments and their temporal pattern.
6. ATMOSPHERIC DISCHARGE CHRONOLOGY

On the basis of information provided by BNFL, Stather et al. [3] gave a chronology of discharges for high and low release heights, nominally at 100 m and at ground level, respectively, attributable to operations associated with reprocessing, operation of the Windscale Piles, and fuel storage in the ponds. Besides routine releases, the chronology included other releases where these were considered significant (including releases of uranium oxide particles from the piles), an exception being the discharge from the 1957 fire in Windscale Pile No. 1 which was given separately.

A number of uncertainties were recognized in the chronology given by Stather et al. [3]. For example, radiochemical analyses of stack samples were not carried out prior to 1964. Therefore, for plutonium and americium, the average annual discharge for the period 1964–1967 was projected backwards to 1952. Annual discharges of $^{90}$Sr, $^{95}$Zr, $^{95}$Nb, $^{106}$Ru, $^{137}$Cs and $^{144}$Ce during the period 1958–1963 relative to those in 1964–1967 were estimated on the basis of fuel irradiation and throughput rates. The calculated annual discharge for 1958 for these nuclides was projected backwards to 1952.

In addition, it was recognized that there were unmonitored sources of atmospheric discharge from the site in the early years, primarily from the early fuel storage ponds and plutonium plants. These discharges were estimated from the few measurements of cumulative plutonium deposition in soil then available and from data on the radionuclide concentrations in storage pond water.

In order to address these uncertainties, we have used measurements of total alpha and total beta, recovered from plant archives, to improve the estimates of pre-1964 stack discharges, and a much larger database of soil deposition measurements has been used to re-evaluate the early unmonitored emissions [19].

Subsequent to the publication by Stather et al. in 1986, further studies have been carried out in relation to aerial effluents from Sellafield which also require the discharge chronology to be reconsidered. These relate to investigations of the efficiency of stack sampling systems and the effective release heights of the stacks; the review of historic documentation also occasioned a few changes to the previous chronologies.

On the basis of the above considerations, we have made revisions to the atmospheric discharge database given by Stather et al. [3], as summarized below:

(a) Revision of routine plutonium releases from tall stacks in the period to 1964 based on results of air samples taken in the reprocessing plant stack.
(b) Backfitting of scaling factors known as sampling efficiency factors (SEFs) to past atmospheric discharges to allow for inadequacies in stack sampling.
(c) Revision of effective heights from 100 m to 80 m for discharges from tall stacks and to 10 m for atmospheric discharges of caesium and strontium, from 1973 to 1985.
(d) Addition of a specific release of plutonium in May 1968 to discharges from tall stacks.
(e) Postulation of a ground level plutonium source term of about 70 GBq, with high deposition velocity (0.1 m/s), released during the early years, to account for plutonium deposition and isotopic ratios found in soil cores taken close to the site.
(f) Review of \(^{35}\)S emissions from the Calder Hall reactors.
(g) Estimation of \(^{14}\)C discharges from reprocessing derived from available emission measurements and \(^{14}\)C measurements in local tree rings.

The revised discharge chronologies derived from these assumptions are given in Table II (80 m effective height) and Table III (10 m effective height). Additional data on significant abnormal releases which may require separate consideration, including the emission of uranium oxide from the Windscale Pile chimneys and the release from the Pile fire, are given in Table IV.

7. CONFIRMATION OF THE ATMOSPHERIC DISCHARGE CHRONOLOGY

As for liquid discharges, it is possible to confirm the major features of the atmospheric discharge chronology from environmental measurements by use of SEAM.

7.1. Air sampling

Some air sampling measurements have been made on and around the Sellafield site since the early 1950s. However, it is only since the early 1970s that individual radionuclides have been measured with sufficient sensitivity for the data to be useful in model validation. For this period, the calculated concentrations of \(^{137}\)Cs, \(^{239,240}\)Pu and \(^{241}\)Am agree well with measured values [4].

7.2. Fission products in milk

Milk is a good indicator of a number of fission products in the environment, including \(^{90}\)Sr, \(^{137}\)Cs and \(^{131}\)I. Milk from the vicinity of Sellafield has been routinely sampled and analysed for these radionuclides by the UKAEA and BNFL, since 1958, 1961 and 1958, respectively. Thus, milk samples have the potential to confirm the atmospheric discharge chronology of these important fission products. Generally, the results for \(^{131}\)I are below the limit of detection, except for times of known abnormal releases of \(^{131}\)I (and the period following the Chernobyl accident in 1986). Results for \(^{90}\)Sr and \(^{137}\)Cs are consistently positive, but, of course, sources such as weapons testing fallout need to be considered in interpreting the time series monitoring data.

Text continued on page 185.
### TABLE II. DISCHARGE OF RADIONUCLIDES TO THE ATMOSPHERE FROM SELLAFIELD DURING THE PERIOD 1951-1992 AT AN ASSUMED STACK HEIGHT OF 80 m (Bq/a)

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<th>Year</th>
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<th>$^{35}$S</th>
<th>$^{41}$Ar</th>
<th>$^{60}$Co</th>
<th>$^{90}$Sr</th>
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Notes: An estimated discharge of 1.8E+11 Bq of $^{210}$Po occurred in 1952. The $^3$H discharges from 1955 to 1962 inclusive are upper estimates based on materials accountancy data for $^3$H processing plants which operated over that period.
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<th>Nuclide</th>
<th>Release (Bq)</th>
<th>Basis of release estimate</th>
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FIG. 4. Model calculations of $^{137}$Cs concentrations in milk from a farm near Sellafield, compared with the results of environmental surveillance measurements.

Figure 4 shows the concentrations of $^{137}$Cs measured in milk at a farm 0.8 km north of the Sellafield site, which had been sampled consistently from 1958 to 1989, compared with concentrations predicted using the LANDFOOD module of SEAM. Clearly, the discharge chronology can account well for the measured concentrations, but during the period for which measurements are available the contributions from Sellafield are only dominant in the late 1970s.

Similar comparisons for $^{90}$Sr show that the discharge chronology and model generally overestimate $^{90}$Sr concentrations in milk.

7.3. Plutonium deposition and lake sediment cores

Because of the lack of environmental measurements prior to the 1970s, confirmation of the atmospheric discharge chronology for plutonium can only be obtained from cumulative deposition measurements. Soil core measurements have been used to assess the magnitude of unmonitored emissions from the site [19]. Two sediment cores taken from Ponsonby Tarn, a small lake located 1.8 km from Sellafield, have been shown to contain deposition chronologies [20, 21].

Although a number of uncertainties arise in interpreting the sediment core profile as a simple sequential record of atmospheric deposition, it is reassuring that
FIG. 5. Deposition of $^{238}\text{Pu} + ^{239,240}\text{Pu}$ in the sediments of Ponsonby Tarn, 1.8 km from Sellafield. Calculations of deposition rates from SEAM compare well with measured deposition in dated sections of a sediment core.

the deposition rates calculated from SEAM account well for the profile (Fig. 5). Thus, the discharge chronology accounts for both the total deposition of plutonium alpha near the site and its temporal distribution.

7.4. Autopsy data

Further verification of the atmospheric discharge chronology for plutonium may be obtained by reference to analysis of autopsy tissues of eight non-occupationally exposed persons, resident in Seascale or in the surrounding area from the early days of site operation until the time of their death between 1980 and 1984 [22]. In each case, death was due to cardiovascular disease.

A large fraction of the plutonium in the body which enters systemic circulation is taken up by bone and liver, with long retention half-times. The content of these organs at autopsy is an indication of the cumulative intake and the subsequent uptake into systemic circulation over several decades or more.

The intake of plutonium by inhalation has been calculated for an adult continuously resident in Seascale from 1950 to 1984 using SEAM. From these intakes the plutonium content of bone and liver has been calculated using metabolic models of the International Commission on Radiological Protection [23, 24]. It has been
assumed that plutonium was inhaled as an aerosol of 1 µm activity median aerodynamic diameter (AMAD), with 12% of the inhaled material passing to systemic circulation.

The calculated organ content for the skeleton is 1.6 Bq, compared with autopsy results in the range of 0.035–0.13 Bq; for the liver the calculated organ content is 1.0 Bq, compared with autopsy results in the range of 0.03–0.3 Bq.

The overestimation in predicted organ contents indicates that the estimation of discharges from high stacks in the pre-1964 period may have been overly pessimistic. On the other hand, the assumptions regarding the form and metabolic behaviour of plutonium (which are conventional choices for radiological assessment purposes) may be cautious. In either case, this exercise suggests that, in combination, the discharge chronology and the overall method used for assessment of plutonium intakes are likely to result in an overestimate of the systemic uptake of plutonium by individuals.

8. DISCUSSION AND CONCLUSIONS

Compiling the liquid discharge chronology has been relatively straightforward. However, the position for atmospheric discharges is more complex.

Substantial confirmation of the discharge chronology has been obtained by using SEAM in conjunction with available environmental measurements. A good match is obtained for discharges of important radionuclides and measured concentrations in environmental materials. It should be noted that many more comparisons have been made than are reported here.

A further assessment of doses to Seascale children is being undertaken by the National Radiological Protection Board on behalf of the Committee on Medical Aspects of Radiation in the Environment (COMARE). As a result of revisions to the discharge chronology and other revisions in the assessment methodology, the radiation dose to red bone marrow for an average child born in Seascale in 1950, summed to the age of 25, is now given as 5.8 mSv [25], compared with 6.2 mSv, given by Stather et al. [3] for a child born in 1950, summed to the age of 20.

According to these calculations, the conclusions of Stather et al. [3] would remain essentially unchanged: the assessed exposure to environmental radioactivity fails to explain the incidence of childhood leukaemia in Seascale. Further, the radiation dose to every child (and young adult) resident in Seascale would need to have been in excess of 50 mSv every year (i.e. ca. 1000 mSv, summed to the age of 20) if environmental radioactivity were to account for the leukaemia incidence (i.e. exposure comparable to occupational dose limits for workers at the plant) [22].

It may be concluded that the likelihood of such an underestimation of radiation exposure or risks to children in Seascale is highly improbable.
REFERENCES


RADIATION DOSES TO CRITICAL GROUPS SINCE THE EARLY 1950s DUE TO DISCHARGES OF LIQUID RADIOACTIVE WASTE FROM SELLAFIELD

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Abstract

RADIATION DOSES TO CRITICAL GROUPS SINCE THE EARLY 1950s DUE TO DISCHARGES OF LIQUID RADIOACTIVE WASTE FROM SELLAFIELD.

To set in context the precautions taken to protect man and the environment, the paper reviews some of the work on exposure pathways in connection with proposed and early liquid radioactive waste discharges from Sellafield. The main historical features of these discharges, affected by relevant plant operations, are briefly described. The important radiological exposure pathways resulting from the discharges and from people's consumption and occupancy habits are considered. To place the changing scenario onto a consistent basis using present-day methodology, a reconstruction of exposures has been carried out. The three major pathways examined are Porphyra/laverbread consumption in South Wales, fish and shellfish consumption near Sellafield, and external exposure over local and more distant sediments. A modelling approach based on recently measured radionuclide concentrations per unit rate of discharge was used to reconstruct exposures generally for the period up to 1977, after which time measured radionuclide concentrations were used directly; however, where reliable monitoring data were available prior to 1977, and important in relation to dose, these data were used in preference. Where practicable, quality assurance comparisons of modelling with direct measurement are presented. Possibly the greatest source of uncertainty is in the assumptions about people's consumption rates, sources of relevant food and occupancy habits. The reconstruction is based on what are believed to be reasonably robust data representative of long term behaviour, applying caution where there are known uncertainties. The results show that over the period 1952 to about 1970 the laverbread pathway was likely to have been critical, taking a cautious approach. Effective dose rates fluctuated at around 1 mSv/a from about 1956 to 1971. From about 1970 to 1985, the fish and shellfish pathway was likely to have been critical, with effective dose rates peaking at about 2 mSv/a in 1975-1976. These dose rates were within the contemporary ICRP dose limit for the public of 5 mSv/a. External exposure was likely to have been of lesser importance until about 1985, when with the retention of previously released radiocaesium on sediments it became dominant. This phenomenon applies particularly further afield where radiocaesium concentrations have been slower to
decline; in the Ribble estuary, houseboat dwellers have been the critical group from about 1985. Effective doses have been about 0.3 mSv and declining; they are due to the effects of radiocaesium discharges in earlier years.

1. INTRODUCTION

Low level liquid radioactive wastes have been discharged from the Sellafield site to the Irish Sea since 1952. The major source of these wastes was at first the processing of nuclear fuel for military purposes, but from the late 1950s operations were dominated by reprocessing of fuel from commercial nuclear power programmes in the United Kingdom, with similar fuel from abroad. Since discharges began, controls were applied to protect the public and the environment. These controls took account of the developing knowledge of hydrographic dispersion; of uptake of radionuclides by environmental materials and by man; and of international guidance on radiological protection principles. Additional safety factors were applied if there were significant uncertainties [1].

Protection of the public was, and continues to be, focused on those people likely to be the most exposed, who (perhaps unfortunately) became known as the ‘critical group’. This paper briefly reviews some of the early work on exposure pathways to set in context the precautions taken to protect man and the environment. The developing history of the Sellafield liquid radioactive waste discharges is then briefly described. The radiological exposure pathways resulting from the changing discharge pattern as well as from changes in people’s consumption and occupancy habits are reviewed. Over the years, the methodology for assessing radiological significance has developed; for example, prior to the recommendations of ICRP-26, controls were typically based on critical organ doses. To place the changing scenario onto a consistent basis, a reconstruction of exposures using present-day (ICRP-60) methodology has been carried out. A modelling approach based on discharges was used as well as available environmental monitoring data. The exposures from the different pathways over time are compared and contrasted, and conclusions drawn.

2. EARLY ASSESSMENTS

"Before the erection of an atomic energy production plant on the coast of the Irish Sea, it was essential to make an estimate of how much radioactivity could be disposed continually into the Sea without danger to the neighbourhood or to animal and plant life" [2]. In this section, these precautionary assessments are summarized.

Hydrographic dispersion off the Cumberland coast (Fig. 1) was investigated in a series of dye tracer experiments, ‘operation peanuts’ [3]. The results were supplemented by studies of tidal streams and of salinity data for the Irish Sea [1].
Assessments were made [4] of a wide range of potential pathways to man and the environment, including: external pathways from contact with sea water and the sea shore; internal irradiation from consumption of fish and seaweed and from breathing silt or sea spray; and irradiation of fish. In general, a concentration factor approach was used, involving for fish extensive studies of movements in the Cumberland
coastal area and experiments of uptake from labelled sea water in tanks [1]. The edible seaweed *Porphyra umbilicalis*, which was harvested commercially for consumption in South Wales (Fig.1), was also studied using labelled sea water. The assessments showed that the pathways likely to give rise to the greatest exposures were the food-chains through fish and seaweeds and direct irradiation from the shore [4]. Controls based on these pathways to man would also protect the fish population. Radiological guidance from the ICRP related to occupational exposures (0.3 R per week)\(^1\); this was modified by a safety factor of ten to apply to the public. The result is equivalent in today's units to \(\sim 12\) mSv/a. A further safety factor of ten was applied in order to account for uncertainties in the assessments. The most restrictive pathway for each radionuclide of importance was used to derive permissible discharge levels. Following consultation with Government Departments, discharges commenced in May 1952. Extensive environmental monitoring programmes were conducted [5] to investigate the effects of the discharges. The safety of the permissible levels was confirmed. The developing knowledge enabled modifications to be made to discharge levels over subsequent years. From August 1954, following the Atomic Energy Authority Act 1954, discharges became formally authorized by Government Departments. In 1971, operation of the site became the responsibility of British Nuclear Fuels Limited (BNFL).

3. **SELLAFIELD LIQUID RADIOACTIVE WASTE DISCHARGES**

Extensive work has been carried out by BNFL to document the Sellafield discharges, carrying out reconstruction where necessary [6, 7]. The results of this work have been used here. Discharges of some of the radiologically significant nuclides are shown in Figs 2 and 3.

3.1. **Ruthenium-106**

From the start of operations, \(^{106}\text{Ru}\) discharges increased, particularly in the late 1950s, reflecting the plant throughput. In 1964, the second reprocessing plant came into operation; this plant was associated with facilities for concentration of medium-active liquors by evaporation to allow increased decay storage prior to discharge. These facilities controlled \(^{106}\text{Ru}\) discharges in a scenario of increased fuel throughput and irradiation times; there was also increased decay of fuel in the ponds in the mid/late-1970s. Further decay storage was introduced in 1980, and the decision was taken in 1984 to construct the Enhanced Actinide Removal Plant (EARP) which would process the stored liquors. Meanwhile, some of the process streams

\(^1\) 1 R = \(2.58 \times 10^{-4}\) C/kg.
were re-routed to the high-active evaporator and, in 1985, an additional salt evaporator system was introduced to concentrate salt bearing medium-active streams. By all of these means the $^{106}$Ru discharges progressively decreased from the mid-1970s. Since the introduction of EARP in 1993, the ability to control $^{106}$Ru discharges is being investigated further.

3.2. Radiocaesium

Radiocaesium discharges arise not only from reprocessing operations but also from the fuel storage ponds. Discharges from 1952 onwards broadly reflected pond and reprocessing activity until the early to mid-1970s when increased storage and corrosion of Magnox fuel caused significantly greater discharges from the ponds. Interim abatement was provided by the use of containers of zeolite in the ponds through which the pond water was pumped: this method of control lasted until 1985 when the Site Ion Exchange Effluent Plant (SIXEP) came into operation, significantly reducing the pond water source of radiocaesium discharges. Reprocessing effluents too were reduced by the cessation of discharges of stored medium-active liquors and the use of the salt evaporator with storage. The EARP has been in use since 1993 to treat liquors containing radiocaesium.

3.3. Actinides

Discharges of actinides are summarized in Fig. 3. Increased reprocessing and recovery of residues led to greater discharges up to the mid-1970s. Discharges were reduced by a combination of: operation of a flocculation plant from the mid-1970s; cessation of discharging stored medium-active liquors from 1980; re-routing of some process streams to high-active evaporation and storage in the early 1980s; and operation of the salt evaporator since 1985. Since 1993, the EARP is being used to treat liquors containing actinides.

4. RADIOLOGICAL EXPOSURE PATHWAYS

Extensive marine monitoring, which continued after the first discharges, showed that uptake of $^{106}$Ru by Porphyra seaweed and consumption as laverbread was the limiting factor in disposal of radioactive effluent from Sellafield [8, 9]. Other pathways continued to be monitored, but they were shown to be of lesser importance, including that of fish consumption; at that time, discharges of nuclides with significant uptake into fish flesh were minor. Much effort was put into investigating the laverbread pathway and quantifying its importance on the basis of habits and market surveys [10–12]. A significant factor was not only the consumption rate but also the dilution of Cumberland Porphyra with seaweed from other locations. For control
purposes, doses were assessed assuming no dilution. Radiological guidance also developed. Assessments on the basis of ICRP Publication 2 [13], with a dose limit to the lower large intestine of 1.5 rad/a for members of the public, put the doses to the critical group at about 50% of this limit for 1959; during the early to mid-1960s [11], the doses were reduced to about 20% of this limit.

Regular publication of monitoring data and an assessment of radiological impact by the Ministry of Agriculture, Fisheries and Food (MAFF) commenced in 1967. The first of these reports included monitoring data from 1959 [14]. Following changes in discharge patterns with the reprocessing of commercial fuel, the exposures of the laverbread eaters increased in the late 1960s, but the doses were reported to be well within ICRP limits, even on the basis of no dilution of Cumberland Porphyra. From 1970, however, dilution began to be a more significant factor, and in 1972 [15] the harvesting of Cumberland Porphyra decreased abruptly owing to changes in transport facilities. The importance of other pathways had been kept under review; for 1972–1973 the critical pathway was reported as external exposure over mud in the Ravenglass estuary (Fig. 1) at about 7% of the ICRP whole-body dose limit of 0.5 rem/a [15].

It was also recognized that shellfish contribute to the diet of the local seafood consuming group and that shellfish, particularly molluscs, accumulate radionuclides with an affinity for adsorption to sediments, including $^{106}$Ru and actinides. Consumption rates were kept under review using habits surveys, including consideration of children. The dose rates to the critical group of fish and shellfish consumers were reported as 69% of the 5 mSv ICRP-26 limit in 1981; this result was based on a cautious gut transfer factor for plutonium [16]. Thereafter, reductions in discharges (Figs 2 and 3) caused progressive reductions in concentrations of radionuclides in fish and shellfish, and hence in the dose to the critical group. These reductions lagged behind the reductions in discharges due to the delayed effect in the environment particularly for actinides adsorbed on sediments. In 1990, the dose to fish and shellfish consumers had reduced to 0.16 mSv/a on the basis of ICRP-26 methodology [17]. In the same year, however, a further effect of adsorption of radionuclides on sediment had become apparent. At a distance from Sellafield, the time lag effect of reductions in concentrations of radionuclides was more pronounced. The external exposure of houseboat dwellers in the Ribble estuary (Fig. 1), some 90 km from Sellafield, which is mainly due to $^{137}$Cs adsorbed on sediments, emerged as slightly greater (0.18 mSv) than that of the fish and shellfish consuming group near Sellafield [17]. This relative importance has persisted until the present, and is being kept under surveillance.
5. RECONSTRUCTION OF DOSES TO CRITICAL GROUPS

5.1. Introduction

In the MAFF publications referred to in the previous section, doses to the critical groups have been reported on different bases over the 40 year history. One of the main differences has been the development of radiological protection guidance, with the particular change from organ doses to effective dose equivalent brought about by ICRP-26. Further, different doses per unit intake have been introduced because of changing models and parameters; one such change has been based on revised gut transfer factors for actinides. To bring these radiological factors onto a common basis, a reconstruction has been carried out on the basis of ICRP-60 methodology, using dose per unit intake data mainly from ICRP-67 [18]. However, some of the differences over time are due to the changing habits of the critical groups and other factors such as the source of particular foods and exposure locations. It is these last factors which are more difficult to allow for where uncertainty exists. Therefore, the reconstruction is based on what are believed to be reasonably robust yet cautious data for the habits of critical groups, representative of long term average behaviour. The three pathways of laverbread consumption, fish and shellfish consumption and external exposure are dealt with, other pathways having been kept under review over the years but shown to be of minor importance (Ref. [14] and subsequent MAFF reports). Also of relatively minor importance in terms of critical group dose have been pathways associated with routine airborne discharges.

5.2. Porphyra/laverbread pathway

The laverbread industry and consumption by high rate consumers in South Wales was thoroughly investigated and kept under review during the period of importance of this pathway [10–12, 19]. The two large surveys of 1967 and 1972 indicated representative critical group consumption rates of 160 g/d and 130 g/d, respectively. However, it would be unrealistic to apply these figures directly in respect of Cumberland Porphyra because of dilution during manufacture with seaweed from locations further afield, and the absorption by water during cooking. A guide to the significance of these factors may be gained from $^{106}\text{Ru}$ measurements of the laverbread product compared with Cumberland Porphyra. Data for the late 1960s, prior to the transport difficulties of the early 1970s, indicate dilution factors in the range of 4–16, depending upon manufacturer ([12]; [15] and earlier reports). Some manufacturers used more Cumberland Porphyra than others; assessments for 1959–1967 of the content of this seaweed in receipts by these manufacturers varied from 11% to 32% [12]. The dilution due to cooking, about a factor of two [12], would need to be taken into account to obtain the overall dilution. Clearly, the sources of variability make specification of a single consumption rate to cover the
FIG. 4. Comparison of calculated concentrations of $^{106}$Ru in Porphyra with results of environmental monitoring.
period 1952–1976 for comparative purposes difficult. However, for the purposes of this study, to take cautious account particularly of the variability in manufacturers’ sources of supply, a laverbread consumption rate of 150 g/d was combined with a dilution factor of six (including the effect of cooking) to give an equivalent consumption rate of 25 g/d (wet weight) of Cumberland Porphyra.

Radionuclide concentrations in Cumberland Porphyra from 1952 to 1969 were calculated using the method of normalized activity concentrations [20]. The results of environmental monitoring generally from 1971 to 1981 (Ref. [16] and previous reports) were used when more radionuclides than in the early period were analysed, but the data for some nuclides had not yet been influenced by the lag effect following decreased discharges. The monitoring results were normalized to discharge data for each year [7] prior to averaging. Data for the following radionuclides were included: \(^{90}\)Sr, \(^{95}\)Zr/Nb, \(^{106}\)Ru, \(^{134}\)Cs, \(^{137}\)Cs, \(^{144}\)Ce, \(^{238}\)Pu, \(^{239/240}\)Pu, \(^{241}\)Pu, \(^{241}\)Am. Contributions to the dose from other radionuclides are minor. An allowance was made for grow-in of \(^{241}\)Am from \(^{241}\)Pu. To illustrate the prediction of radioactivity concentrations, Fig. 4 shows a comparison of calculated concentrations of \(^{106}\)Ru in Porphyra from 1963 to 1985 with the results of environmental monitoring. This period includes that of averaging, but there is no apparent systematic effect for 1971–1981 and concentrations for 1982–1985 are in good agreement. The predicted concentrations appear to be systematically higher than the results of environmental monitoring for 1963–1970, but their use would be cautious. In these calculations, data from this laboratory have been used. Other data are available, particularly those of BNFL and its predecessors, and the data are generally in good agreement [21].

It has already been mentioned that, since 1970, transport difficulties led to a reduction of the content of Cumberland Porphyra in South Wales laverbread. Therefore, for 1970–1976, an additional reduction factor was applied to the concentrations of all nuclides, based on the relative ratios of \(^{106}\)Ru measured in laverbread as compared with Cumberland Porphyra. Calculations were only carried out for the years up to 1976 because, from 1971, this pathway declined in importance. Annual effective doses, based on a consumption rate of 25 g/d Porphyra and ICRP-60 dose coefficients [18], are shown in Fig. 5. The doses were mainly due to \(^{106}\)Ru, but from the mid-1960s the actinides began to contribute (35% of the dose in 1970). The reconstructed effect is that doses increased to about 1.2 mSv/a by 1956 and fluctuated about the 1 mSv/a level until 1971 when they decreased, becoming no longer of radiological importance.

5.3. Fish and shellfish consumption pathway

Whilst this pathway was kept under review from the commencement of discharges, more effort was put into quantifying it from the early 1970s [19], with the decline in the Porphyra/laverbread pathway. Variabilities in consumption rates by
Committed effective
dose rate (μSv/a)

Dose to seaweed consumers
Dose to fish/shellfish consumers
External exposures
Measured exposures to houseboat dwellers

the critical group of fish and shellfish consumers were reported particularly up to the mid-1980s (e.g. Ref. [16]). Specification of a single set of consumption rates for the purposes of comparison of doses with those of different exposed groups and reconstruction of doses for years in which consumption rate data were unavailable was based on the relatively stable consumption levels which were reported for the late 1980s. These levels have proved to have been fairly robust as long term averages outside this period. Accordingly, consumption rates of 36.5 kg fish per year, 6 kg crustaceans per year and 8.3 kg molluscs per year were used and, because mixtures of these foods are eaten by high rate consumers, the doses were summed. Following studies of foods consumed, fish were taken to consist of an equal mixture of plaice (Pleuronectes platessa) and cod (Gadus morhua); crustaceans were taken to consist of a 2:1 mixture of crab (Cancer pagurus) and lobster (Homarus gammarus); and molluscs were taken to consist entirely of winkles (Littorina littorea). Other molluscan species are eaten, but winkles represent a high proportion of the total and can be taken to be representative of the other species.

Radioactivity concentrations up to 1973, when not all foods were included in monitoring programmes, were based on calculations using normalized activity concentrations, generally derived from available data for 1977-1990. However, for radiocaesium, data for the period 1977–1983 were used to avoid the unrepresentative lag effect of reduced discharges. For actinides, normalized activity concentrations were derived using the method of availability times [22]. Radioactivity concentrations used for all species for 1978–1993 were as measured by environmental monitoring (e.g. Ref. [17]). For the period 1974–1977, monitoring data were available for the species of interest for radiocaesium, an important contributor to dose, so these data were used, supplemented by calculated concentrations for other radionuclides.

Annual effective doses were calculated, using the consumption rates and radionuclide concentrations derived as above. Dose coefficients given in ICRP Publication 67 [18] were used, except for plutonium and americium in winkles, for which a gut transfer factor of 0.0002 instead of 0.0005 is justified [23]. The results are shown in Fig. 5. In the period up to the late 1960s, doses were mainly due to $^{106}$Ru in shellfish; subsequently, radiocaesium in fish and actinides in shellfish also became of significance. From the mid-1970s to the mid-1980s radiocaesium in fish was the main contributor to dose, such that doses peaked at about 1.9 mSv in 1975. From the mid-1980s, radiocaesium discharges decreased following the operation of SIXEP, and actinides in shellfish provided the most important contributions to the dose.

An indication of the reliability of the predicted doses compared to those based directly on monitoring data is given in Fig. 6. This shows a comparison of the two methods for the period from 1978 to 1985, for which measurements were available; the lag effect due to reduced radiocaesium discharges became noticeable in 1984, and this is apparent in the figure. Prior to 1981, the agreement is within about ±10%.
FIG. 6. Comparison of dose rates from fish/shellfish consumption.
5.4. External irradiation over exposed sediments

This pathway was recognized as being of potential importance in the early studies (e.g. Ref. [4]). Dose rate monitoring over Cumberland beaches, estuarine sediments and harbours continued to be carried out as a significant part of aquatic monitoring programmes both by the operators and authorizing Ministries. However, a combination of the data with observations of public occupancy times showed that the doses were lower than those to the laverbread eaters. This situation persisted until about 1970-1971, when, with the growing importance of $^{95}$Zr/Nb and $^{144}$Ce discharges from the second reprocessing plant and the decline in Cumberland Porphyra transports, external exposure in the Ravenglass estuary (Fig. 1) was reported as critical [24]. However, by 1974 [25], fish and shellfish consumption had assumed critical importance because of the increased discharges of radiocaesium and its uptake into these foods. External exposures were kept under review by means of habits surveys and environmental monitoring; in 1980, because of the decline of the salmon trap at Ravenglass, the doses to boat dwellers in Whitehaven harbour (Fig. 1) were reported as the most important doses from external exposures [26]. In 1989, bait diggers were reported as the most exposed people in Whitehaven harbour. However, before this, in 1986, with the decreases in radiocaesium discharges being of greater effect near Sellafield, exposures further afield of houseboat dwellers in the Ribble estuary (Fig. 1) were identified as the most important because of the longer occupancy times; the dose rates over exposed sediment continued to be higher in west Cumbria than further afield.

The choice of a long term, representative location and occupancy time for the purpose of reconstruction has been complicated by the number of changes. Early discharges would have given more important external exposures in west Cumberland than at a distance and much of the data which can be extrapolated have been gathered in connection with Whitehaven harbour; therefore this location was chosen as a basis for deriving a representative dose. A suitable long term average occupancy time for boat dwellers (to include the effect of shielding by the hull) or bait diggers from habits survey observations was taken as 500 h/a over exposed sediment. The dose rates to houseboat dwellers in the Ribble estuary have been monitored directly from the time they became important, and were treated as a separate case.

Since 1981, gamma dose rate monitoring data from Whitehaven harbour have been used directly. For the period prior to this, doses were calculated using normalized activity concentrations for sediment in Whitehaven harbour, averaged from 1977 to 1990, except for radiocaesium, which was averaged from 1977 to 1982 in view of the rapid reductions in discharges after that period. Dose rates over exposed sediment were derived using a model [27] which takes account of the sedimentation rate, assumed to be 2 cm/a in Whitehaven harbour, although the results are not unduly sensitive to this parameter. Doses from beta particles, which could irradiate the gonads of bait diggers, were included, using a model derived for such a pur-
FIG. 7. Comparison of external dose rates, Whitehaven harbour.
pose [28]. The penetrating beta particles from $^{106}$Ru and $^{144}$Ce may be shown to increase the gamma component to the dose by factors of about two and five, respectively. Low adsorption of $^{90}$Sr by sediment leads to only a small contribution to external dose. Allowances for the beta component of dose were added to the gamma monitoring data used since 1981.

The dose rates derived from this process are plotted in Fig. 5. Also plotted are the data for the Ribble houseboat dwellers from 1981 to 1992, based on environmental monitoring. The calculations show that from 1952 to the mid-1960s, $^{106}$Ru was the most important contributor to the dose from the external pathway; with the operation of the second separation plant from 1964, the doses began to be enhanced by $^{95}$Zr/Nb and $^{144}$Ce in discharges. In the mid-1970s, radiocaesium became the most important contributor to external dose following the increased discharges. The peak dose rate was about 0.9 mSv/a in 1975. With the reduction in discharges, external doses declined, but the rate of decline was slower than that of doses from fish and shellfish, presumably because of the retention of radionuclides, including radiocaesium, on and within the sediment. Since 1985–1986, the external exposures of Ribble houseboat dwellers have been more important than the external doses in west Cumbria — at about the 0.3 mSv level and slowly declining.

The ability of the external dose rate model used to reconstruct doses prior to 1981 is illustrated in Fig. 7. Strictly comparable data are only available over the period from 1981, when monitoring was carried out under more consistent conditions, to 1983, when decreases in radiocaesium discharges began to render the model invalid because of retention of radiocaesium on sediments. This last effect can be seen clearly. Over this period there is good agreement, and prior to this the model appears to be predicting on the cautious side for the less consistent data.

6. DISCUSSION AND CONCLUSIONS

The history of doses to the Sellafield critical groups, assessed as described in Section 5, is summarized in Fig. 5. The ability of the models used to reconstruct doses has been shown to be reasonably good. Annual variations due to environmental conditions have not been allowed for in the case of reconstructed data, which apply mainly prior to the mid/late-1970s; thus these data should be seen in the context of longer term average behaviour. Probably the greatest systematic uncertainties are due to the use of long term average parameters for consumption and occupancy. These have been chosen to be reasonably, but not excessively, cautious; perhaps greater caution has been applied for the laverbread pathway than in other cases, in order to take account of the uncertain mix of Porphyra from the west Cumberland coast with seaweed from other locations.

Figure 5 verifies that, over the period from 1952 to about 1970, the laverbread pathway was likely to have been critical. The peak effective dose rates to the high
rate laverbread eaters fluctuated at about the 1 mSv/a level from 1956 to 1971. This is well within the then current whole-body dose limit of 5 mSv/a. From about 1970 to 1985, the fish and shellfish pathway was likely to have been the most important, with effective dose rates peaking at about 2 mSv/a in 1975-1976, still within contemporary limits. In 1985, the ICRP made known its view [29] that the principal dose limit for the committed effective dose equivalent to the public was 1 mSv/a. By 1985, the doses had been reduced to about 0.3 mSv/a, well within this lower limit. As regards the external radiation exposure pathway, this is likely to have been of lesser importance than the intake pathways up to about 1985; but, with the decreases in Sellafield discharges and the retention of previously released radiocaesium on sediments, it has become of dominant importance, especially in the Ribble estuary. It is noteworthy that this situation is a consequence of past discharges, with slower rates of decrease in radionuclide concentrations in sediments further afield. If a modelling approach as described in Section 5 is used to calculate only the effects of current discharges, the most important pathway ('critical' in terms of controls which can be applied now) is from fish and shellfish consumption near Sellafield. The enhancement due to past effects, present for all pathways, is not the result of a 'practice' in current, ICRP-60, terms; however, as Fig. 5 shows, the current overall dose rates are well within the 1 mSv/a public dose limit for practices.

REFERENCES


ENVIRONMENTAL IMPACT OF THE
LA HAGUE REPROCESSING PLANT

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Abstract

ENVIRONMENTAL IMPACT OF THE LA HAGUE REPROCESSING PLANT.

When authorizing a capacity increase for the La Hague site from 400 to 1600 Mt U per year, the French regulators did not agree to an increase in the release limits for radioactive liquids and gases, nor did they allow of subsequent licence amendments to raise those limits. The past four years of plant operation indicate that not only have these limits been respected but there has also been a noticeable decrease in radioactive liquid releases. Actual operating experience put the average exposure of plant personnel at less than 0.26 mSv in 1994, below the design criteria of 5 mSv/a and well below the maximum allowable dose of 50 mSv/a, and there is every reason to believe that this plant performance will continue. The favourable performance of the La Hague plant is the result of an approach in which safety features are incorporated early in the design phase, particularly those relating to containment, not only for normal operations but also for maintenance and repair. Furthermore, the operating experience, particularly in the area of waste minimization and environmental impact, is factored into design and operations. The La Hague monitoring programme, approved by the Ministry of Health, is a source of valuable data. All results of the radiological measurements, tracing the effect of radioactive releases on the environment, are forwarded to the Office of Protection against Ionizing Radiations. The results achieved over the last ten years indicate that the environmental impact of the La Hague site has been reduced by a factor of three, while the amount of reprocessed spent fuel has increased by a factor of four.

1. INTRODUCTION

Industrial reprocessing of spent fuel at La Hague started in 1966. Since that time, approximately 5000 Mt of metallic fuel and 7000 Mt of oxide fuel have been reprocessed in the plant, up to the beginning of 1995. The capacity was 400 Mt/a until 1989, when the new process line UP3 started operation with a capacity of 800 Mt/a. In 1994, the total plant capacity was raised to 1600 Mt/a by the commissioning of UP2-800, the twin process line of UP3. The total capacity is now devoted to the reprocessing of French and foreign spent fuel in equal amounts.

Spent fuel reprocessing has a double purpose:

— To separate the fissile materials, uranium and plutonium, from the fission products, which permits recycling of the fissile materials and extraction of their residual energy content;
— To properly sort and condition the ultimate residues, thus ensuring safe and clean long term disposal.

Therefore, reprocessing and recycling is a well proven industrial method for solving problems in connection with the back end of the nuclear fuel cycle: spent fuel is not a burden, it is a valuable energy concentrate; simultaneously, the ultimate waste volume and the toxicity are minimized.

However, beneficial as nuclear operation may be, we know that its radiological impact should be minimized according to the ALARA principle. In the La Hague plant, this principle has been followed in the management of the exposure of operating personnel: in 1994, the average annual exposure reached only 0.26 mSv, i.e. about one tenth of the exposure to natural radioactivity (the latest recommendations of the International Commission on Radiological Protection propose 20 mSv/a as the maximum allowable dose). From 1987 to 1993, the total integrated annual dose to operating personnel decreased from 4.99 to 1.91 man·Sv, while the annual amount of spent fuel reprocessed at the plant rose from 425 to 955 Mt.

The ALARA principle has also been followed in the management of releases to the environment. From the beginning, care has been taken to minimize the impact of liquid and gaseous releases: in the selection of the site, the design of the plant, the optimization of process parameters and the management of effluents. This will be detailed in this paper, focusing on the case of liquid releases.

2. MINIMIZATION OF LIQUID EFFLUENTS FROM THE PROCESS

In the original design of the UP3 plant, medium and low activity liquid process wastes were divided into three categories:

A — Active effluents from the process, which are routed to the effluent treatment facility for decontamination by chemical precipitation, followed by incorporation of the resulting sludges into bitumen;

B — Tritiated water from the process, to be discharged to the sea after monitoring and filtration;

C — Effluents from utilities, to be discharged to the sea after monitoring and filtration.

Experience gained from the first years of operation in the UP3 plant revealed much lower volumes and concentrations of radioactive effluents than had been anticipated from the design values; this was due to the high efficiency of the implemented processes and technologies. These observations led to an improved effluent management scheme. The general principle was to minimize mixing of low level and medium level effluent streams. Thus, the former can be directly released
as waste in category C, and the latter can be recycled in higher proportion after concentration, rather than forming category A effluents.

As a result, the annual volume of medium activity effluents of category A could be dramatically reduced, to the extent that chemical precipitation and bituminization would be completely discontinued in the near future. It should be mentioned that the chemical precipitation parameters (pH, solid in pulp concentration, etc.) strongly determine the residual concentrations of several radionuclides. Consequently, the final amount of released radionuclides will be approximately proportional to the treated effluent volume: the less the volume to be treated, the less is the amount of radionuclides released to the sea.

3. MANAGEMENT OF LIQUID RELEASES TO THE SEA

After filtration, all effluents are stored in buffer tanks, stirred and sampled for chemical and radiological analysis. Release to the sea can only be performed in batches, after complete batch analysis results have been collected and approved. The measurements include:

- beta activity per release
- alpha activity per release
- tritium content
- gamma spectrometry
- pH measurement
- suspended solid particles.

Complementary analyses are performed on a delayed time basis, including:

- strontium-90 activity per release
- plutonium activity per release
- uranium content.

The written approval for release is provided by the Radiological Protection Department (SPR), on behalf of the plant director, who is legally responsible for compliance with regulatory limits.

Concentration limits to alpha and beta activities are imposed on each batch before release. This rule ensures a smooth spread of releases over the year, thus maintaining the lowest possible concentration of radioactivity at the point of release in the sea.

The effluent pipe to the underwater outlet includes a 2500 m long line on land and a 5010 m long line under water from the coast to the end point. It is made of 1 cm thick steel and has a diameter of 22 cm. Annual regulatory control of the pipeline is performed.
4. PARTICULAR CHARACTERISTICS OF THE SEA NEAR LA HAGUE

The La Hague plant is located at the north-west end of the Cotentin peninsula. Tides of large amplitude are observed in this area. The sea strait between the La Hague cap and the nearest island, Aurigny, 16 km farther to the west, is called Raz Blanchard. Violent tidal streams run through it, with a normal speed of 6–8 knots, reaching top speeds of 10 knots, southward during the ebb and northward during the flow. The coast itself consists of a number of cliffs, separated by small sandy coves. The nearest deep sea bottom is covered with pebbles, gravel and coarse sand, since the existing hydrodynamics prevent sedimentation of fine particles, which more readily occurs in the quiet coves.

The point of effluent release is located in Raz Blanchard, 1700 m from the coast, at a depth of 28 m. The most favourable period for release is determined by the tide table, i.e. the period begins two-and-a-half hours before high tide and ends half an hour after it; during this period, 300 m$^3$ effluents can be released at a maximum rate of 180 m$^3$/h. Under such conditions, maximum dilution is obtained, reaching a dilution factor of one million within 1000 m from the release point. The site hydrodynamics have been confirmed to be most favourable for effluent dispersion, given the powerful local currents.

5. REGULATORY FRAMEWORK AND AUTHORIZED RELEASES

The regulatory constraints for liquid releases to the sea are determined by the decree of 28 March 1984. The annual limits are stipulated as follows:

- Tritium $37\,000$ TBq
- Other beta, gamma elements: total $1\,700$ TBq
- $^{90}$Sr + $^{137}$Cs $220$ TBq
- Alpha emitting nuclides $1.7$ TBq

The decree also specifies the required conditions for release, effluent monitoring procedures and environmental monitoring programmes, as well as statutory control by the governmental regulatory body for radiological protection (Office of Protection against Ionizing Radiations, OPRI).

The above mentioned limits obviously take into account differences in radiotoxicity between the radionuclides. They are based on the assessment of the regional radiological impact by the experts of the French Institute of Nuclear Safety (IPSN). The impact study was performed on the basis of:

- A detailed mapping of sea streams;
- Calculated transfer pathways to man of radionuclides through water, vegetation, animals and food;
- The habits of the local population.
TABLE I. ALPHA AND BETA ACTIVITY RELEASES

<table>
<thead>
<tr>
<th>Year</th>
<th>Beta, gamma releases (10^4 GBq)</th>
<th>Alpha releases (GBq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1987</td>
<td>111</td>
<td>456</td>
</tr>
<tr>
<td>1988</td>
<td>58</td>
<td>365</td>
</tr>
<tr>
<td>1989</td>
<td>59</td>
<td>367</td>
</tr>
<tr>
<td>1990</td>
<td>31</td>
<td>369</td>
</tr>
<tr>
<td>1991</td>
<td>12</td>
<td>153</td>
</tr>
<tr>
<td>1992</td>
<td>7.6</td>
<td>107</td>
</tr>
<tr>
<td>1993</td>
<td>7.3</td>
<td>101</td>
</tr>
<tr>
<td>1994</td>
<td>7.0</td>
<td>97</td>
</tr>
</tbody>
</table>

According to the study, the maximum annual exposure caused by authorized releases would not rise above 1% of the regulatory 5 mSv, for the most exposed people (local fishermen's families).

6. EVOLUTION OF REAL RELEASES

The total amounts of released beta, gamma and alpha activities from 1987 to 1994 are reported in Table I. The evolutions of releases and production rates are compared in Figs 1 and 2. The following comments should be made:

— In Figs 1 and 2 it can be seen that radiological releases to the sea have been reduced, while the plant productivity has been rising. The amount (tonnes) of material reprocessed is the first indicator; still more meaningful is the corresponding electricity yield (expressed in GW/a), which depends on the fuel burnup.

— The two steps contributing to this reduction are: the startup of the new chemical process for liquid effluents of medium activity in 1989; and, since 1991, the improved effluent management as a result of which smaller volumes had to be treated.

— Since 1991, the radiological releases are correlated with the treated volumes of effluents (see Table I).

Comparison of the actually released quantities (1994) with the site authorized releases clearly suggests a negligible environmental impact:
FIG. 1. Beta activity of reprocessed fuel released to the sea and production rates of the La Hague plant.

FIG. 2. Alpha activity of reprocessed fuel released to the sea and production rates of the La Hague plant.
7. ENVIRONMENTAL MONITORING PROGRAMME

The annual monitoring programme consists of extensive sampling campaigns of sea water, sand, seaweeds, sea shells and fish, for laboratory analysis and registration on a database. It is performed by the Department of Radiological Protection at the La Hague plant under regulatory control by OPRI. Complementary investigations and measurements are provided by the French navy and by a research team of IPSN which is specialized in radioecology.

7.1. Coastal survey

The survey of the French coast is performed over 200 km. It includes the following targets:

— **Sea water**: A sample is taken daily in the nearest cove. In other places, sampling is done every third month. Measurements include beta, gamma emitters, plutonium and natural potassium. For example, the natural radioactivity in sea water reaches 12 Bq/L. The plutonium limit of detection is $5 \times 10^{-5}$ Bq/L.

— **Sands and sediments**: These are sampled every third month for gamma emission measurements.

— **Seaweeds**: Five species have been selected for survey, according to their ability to fix and concentrate radionuclides such as $^{137}$Cs and $^{106}$Ru/$^{106}$Rh. They are sampled every third month for gamma measurements.

— **Coastal flora**: Studies have suggested possible radioactivity transfer by sea-spray onto the local flora along the coast. Monthly sampling is performed.

— **Sea shells and molluscs**: Oysters and scallops in particular pump and filtrate large volumes of water and fix the contained radionuclides. Gamma emitters are monitored.

7.2. Deep sea survey

The survey in the deep sea extends from the west coast of the Cotentin peninsula to the Seine estuary near Le Havre. It consists of monthly or three-monthly samplings of water, sand and sediments on 29 spots. Sampling is done by professional divers from the French navy. Gamma emitters are monitored. Several species of fish are also caught near the coast every month. The species surveyed are

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- Tritium 22% of the annual limit
- Other beta, gamma elements: total $^{90}$Sr + $^{137}$Cs 4% of the annual limit
- Alpha emitting nuclides 12% of the annual limit
- Alpha emitting nuclides 6% of the annual limit.
mackerels, basses, whittings and soles, which have different diets and different living places.

7.3. Analytical work

Approximately 1000 samples are taken every year from the marine environment for analysis; afterwards, they are thrown back into the sea. These samples provide the basis for 2200 measurements.

The analytical laboratory belongs to the La Hague Department of Radiological Protection. The methods include standard analytical chemistry, use of glove boxes for sample preparation, and measurements of medium level radioactivity, using gamma and alpha spectrometry.

7.4. Results

The results from the environmental monitoring programme are reported monthly, every three months and yearly. The latest results (1994) are shown in Table II. It can be seen that the artificial radioactivity is much lower than the natural activity ($^{40}$K) in all samples.

8. COMPLEMENTARY STUDIES: FAR FIELD SURVEY

The development of very sensitive measurement methods for very small concentrations of radionuclides has made it possible to initiate large scale investigations on the dispersion of radionuclides in the marine environment. The Laboratory of Marine Radioecology of IPSN has developed a strong expertise in this domain over many years. This R&D team is particularly engaged in the survey of radionuclide migrations across the integrated system of the Channel Sea and the North Sea. The programme includes the following objectives:

— To evaluate the impact of the diverse sources of radionuclides,
— To estimate the characteristic transition times of waters,
— To determine the influence of weather conditions,
— To follow and understand the time variations of radionuclides in a given place.

The expertise acquired involves hydrodynamics and mass transfer models as well as biochemistry of sea animals. For instance, the radionuclide $^{125}$Sb is a kind of radiological signature of the La Hague plant. The Channel Sea-North Sea system is fed by currents from the Atlantic Ocean and conversely pours its water into the Norway Channel. Thus, traces of $^{125}$Sb at very low concentrations can be followed quite far to the north. Releases of about 300 GBq per month from La Hague during summer 1993 were estimated in December 1994 to generate concentrations of
TABLE II. NATURAL AND ARTIFICIAL RADIOACTIVITY (GLOBAL ACTIVITIES) MEASURED IN THE ENVIRONMENT OF THE LA HAGUE PLANT (Bq/kg fresh weight)

<table>
<thead>
<tr>
<th>Type of sample</th>
<th>Natural</th>
<th>Artificial</th>
</tr>
</thead>
<tbody>
<tr>
<td>Seaweeds</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fucus</td>
<td>246</td>
<td>12</td>
</tr>
<tr>
<td>Lichen</td>
<td>199</td>
<td>14</td>
</tr>
<tr>
<td>Corallina</td>
<td>102</td>
<td>13</td>
</tr>
<tr>
<td>Laminaria</td>
<td>244</td>
<td>4</td>
</tr>
<tr>
<td>Sand and sediment</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sand</td>
<td>433</td>
<td>11</td>
</tr>
<tr>
<td>Sediment</td>
<td>338</td>
<td>48</td>
</tr>
<tr>
<td>Foodstuffs</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Crabs</td>
<td>70</td>
<td>7</td>
</tr>
<tr>
<td>Oysters</td>
<td>53</td>
<td>2</td>
</tr>
<tr>
<td>Mussels</td>
<td>36</td>
<td>3</td>
</tr>
<tr>
<td>Limpets</td>
<td>70</td>
<td>10</td>
</tr>
<tr>
<td>Fish</td>
<td>99</td>
<td>2</td>
</tr>
</tbody>
</table>
i.e. 200 times less than the dose from exposure to natural radioactivity. This favourable performance of the plant results from a long standing and sustained approach. Safety features were incorporated early in the design phase and the operating experience was also effectively factored into optimized process implementation.
ENVIRONMENTAL RISK ASSESSMENT FOR THE SILLAMÄE WASTE REPOSITORY

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Abstract

ENVIRONMENTAL RISK ASSESSMENT FOR THE SILLAMÄE WASTE REPOSITORY.

An environmental risk assessment has been performed for the repository of the Sillamäe metallurgy plant where, among other things, uranium from alum shale has been processed. The object of the study was to illustrate the consequences to man of a release of the hazardous substances contained in the repository into the sea. Two cases were studied: (1) the environmental impact under present conditions, with a relatively low continuous leakage of radionuclides from the waste repository, and (2) the impact of a sudden dam collapse. The paper presents the radiological impact assessment. The individual and collective doses to the population in the Baltic Sea area were calculated for an integration time of 50 years. The highest individual dose is less than 1 μSv for current leakage, while the dose from a dam collapse is of the order of 2 μSv. The dominant exposure pathway is the consumption of fish. The collective committed dose is about 1 man-Sv for both cases, taking into account the total catch of fish in all parts of the Baltic Sea.

1. BACKGROUND

The Sillamäe metallurgy plant in Estonia has, since 1948, processed uranium from alum shale. During the 1970s the plant also processed loparite (a mineral from the Kola peninsula containing thorium, niobium, tantalum and other rare earth metals) in parallel with uranium ores. Different types of processing and varying technologies during the time of operation have given rise to waste with different physical and chemical properties.

This waste has been deposited on the beach of the Narva Bay in the Gulf of Finland. The repository is an oval retention impoundment with a dam crest about 25 m above sea level. The area is about 330 000 m², and about 50% of it is occupied by a sedimentary pond containing about 150 000 to 200 000 m³ of waste water with a depth of 0-3 m. The bottom of the repository consists of a 1-8 m thick permeable sandy gravel-shingle. Beneath this layer there is a 2-10 m thick layer of clay [1].

Because of its high radioactivity content, its location and current construction, the Sillamäe repository was the object of a specific study. A joint project group was established, consisting of participants from Estonia, Finland, Norway and Sweden.
One of the recommendations of this group was to perform an analysis of the consequences of a radioactivity release due to continuous leakage from the waste as well as a postulated break of the dam. The study included radionuclides and some selected stable elements [2]. This paper presents the results of a study of the radionuclides $^{238}\text{U}$, $^{234}\text{U}$, $^{232}\text{Th}$, $^{226}\text{Ra}$ and $^{210}\text{Pb}$.

2. INVENTORY OF RADIONUCLIDES AND SOURCE TERMS

Samples were taken for analysis, both from within the repository and from the pond. On the basis of these results the inventories of radionuclides in the solid waste and the concentrations in the pond were estimated (Table I). The wide ranges of the estimated contents are due to the wide ranges in the data, which are probably due to the complex structure of the repository, with different layers containing different elements [3].

Continuous source terms were estimated from an analysis of the groundwater between the dam and the coastline, and from an estimation of the groundwater flow (Table I). Also in this case the variations in the data were considerable and, in order not to underestimate the exposure, the best-estimate values chosen were conservatively biased. The pH of the groundwater was also measured and was about 8, in contrast to the acid conditions in the pond water. The source terms were set to be constant during the period of study (50 years). For $^{210}\text{Pb}$, no value was reported; therefore, we used about the same value as for $^{226}\text{Ra}$.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Amount (t)</th>
<th>Concentration (μg/L)</th>
<th>Release (Bq/a)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>B.E.</td>
<td>Ranges</td>
<td>B.E.</td>
</tr>
<tr>
<td>U-238</td>
<td>1600$^a$</td>
<td>500–3200$^a$</td>
<td>2800$^a$</td>
</tr>
<tr>
<td>U-234</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Th-232</td>
<td>850</td>
<td>1000–9000</td>
<td>$&lt;5$</td>
</tr>
<tr>
<td>Ra-226</td>
<td>7.8$^b$</td>
<td>2–25$^b$</td>
<td>3$^b$</td>
</tr>
<tr>
<td>Pb-210</td>
<td>4950</td>
<td>1700–15 000</td>
<td>Not detected</td>
</tr>
</tbody>
</table>

$^a$ Total amount of U.
$^b$ Expressed as kg and pg/L, respectively.
Some assumptions were made for the case of an accidental dam collapse. The pond was expected to be emptied within a time period of 4–8 h. Furthermore, 1–20% of the solid waste in the repository was supposed to be transferred to the near-shore sediments. The continuous leakage from waste will remain at the current rate. After the initial phase, the elements deposited at the bottom of the pond will leak into the overlying water at a rate which depends upon their chemical solubilities (see below).

3. MODEL OF THE ENVIRONMENT

A local compartment model was designed for calculating the initial dispersion of the nuclides in the recipient (Fig. 1). A previously constructed compartment model of the Baltic Sea (compartments 5–11, Fig. 1) [4], with the Gulf of Finland as one compartment (compartment 4), was used together with the local model.

The division into compartments was based upon maps and upon the observed radioactivities in water and sediment. There were somewhat increased radioactivity levels in sediment and water close to the coast, and behind a tongue of land, the latter probably being due to prevailing currents.

The first compartment represents the water close to the shore along the repository, up to 70 m from the beach. The flow from this compartment is to a larger compartment which symbolizes the nearby bay. These two compartments are encircled by a compartment representing the Bay of Narva (400 km$^2$). The turnover of water in these compartments is based upon estimated stream velocities, on average 0.05 m/s along the coast.
Radionuclides and other substances released to the aquatic system are dispersed owing to hydrological conditions. Furthermore, they also interact with suspended matter and with sediments, depending on their physical and chemical properties. To each compartment of water, one compartment of sediments is attached. There are several processes, such as adsorption by particles, diffusion, bioturbation and resuspension, which affect the exchange of elements to, from and within the sediments. In this study, adsorption by suspended matter and resuspension were considered.

The two scenarios need different modelling approaches because of the actual amount of elements released and the chemical conditions. The continuous leakage from the repository is such that the nuclides can be treated as trace elements and do not affect the surrounding chemical environment.

A study of releases during a dam break has to take into account the solubilities of the elements, in contrast to studies of general trace elements. This implies that the solubility of inorganic compounds under various chemical conditions has to be considered. The maximum activity levels of an element in solution were estimated on the basis of chemical data on its solubility as an inorganic compound. This was calculated from solubility products and stability constants for species that are likely to occur, mainly oxides, hydroxides, carbonates and sulphates. It may be reasonable to assume a mixing with oxygen during the dam break event, resulting in oxidizing conditions for the precipitation/dissolution processes.

The pH and the sulphur contents in compartments 1 and 2 during the release of nuclides were approximated from the dilution of the water in the pond, giving pH values of 3 and 5, respectively, and sulphur concentrations of 25 g/L and 250 mg/L, respectively. The chemical conditions in compartment 3 and outwards were estimated from the mean values reported for the Baltic Sea.

The use of solubility products and stability constants for determination of the amount of an element in solution is, of course, a simplification, since processes such as formation of organic complexes are not considered. The kinetics of the processes may also be of importance, as many precipitation/dissolution processes have low reaction rates. The use of thermodynamic constants will, however, give an indication of the possible concentration of elements in solution for different conditions.

4. EXPOSURE PATHWAYS

The main exposure pathway to man from radionuclides in the brackish Baltic marine ecosystem is the consumption of fish [5]. However, milk and meat could also contribute to exposure, since it is possible that cattle drink the water when grazing at the shoreline.

As is common practice, the uptake of radionuclides by fish is described by bioaccumulation factors, taking into account the food-chain implicitly. In a similar
way the resulting levels in milk and meat are obtained from distribution factors. However, for a dam collapse, the concentration of radionuclides in fish in the vicinity of the repository is calculated dynamically.

No doses are given for the immediate vicinity of the plant, represented by compartments 1 and 2 in the model. The main fish for consumption migrates over large areas and it is assumed that there are no cattle in the vicinity of the plant. Consumption rates were set at 0.5 kg of fish and meat per week, and 2 L milk per week. The collective doses were based on the statistics of catches of fish in the different parts of the Baltic Sea.

5. RESULTS AND DISCUSSION

The dose calculations were performed probabilistically. All input parameters were obtained from given distributions according to observed ranges and data from the literature or values assumed by the authors.

The integrated individual dose was calculated at 0.9 µSv for exposure during 50 years of radionuclide release from the Sillamäe waste repository at the current release rate, and at 1.8 µSv for exposure in the event of a dam collapse (Tables II and III). The uranium isotopes dominate the doses at current releases, while $^{226}$Ra and $^{210}$Pb will contribute significantly to the dose for dam collapse. The same is true for the collective doses (Table IV).

Consumption of fish is the dominant exposure pathway (Table II). The average doses from the Gulf of Finland are about 10% of the doses from the Bay of Narva. However, using compartment 2 for calculating the exposures, the doses will increase by a factor of 100.

### TABLE II. INTEGRATED COMMITTED EFFECTIVE DOSES (Sv) DURING 50 YEARS OF RADIONUCLIDE RELEASE DUE TO CURRENT LEAKAGE FROM THE SILLAMÄE WASTE REPOSITORY

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Fish</th>
<th>Milk</th>
<th>Meat</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-238</td>
<td>$4 \times 10^7$</td>
<td>$9 \times 10^{10}$</td>
<td>$1 \times 10^8$</td>
<td>$4.5 \times 10^7$</td>
</tr>
<tr>
<td>U-234</td>
<td>$5 \times 10^7$</td>
<td>$1 \times 10^9$</td>
<td>$1 \times 10^8$</td>
<td>$5.0 \times 10^7$</td>
</tr>
<tr>
<td>Th-232</td>
<td>$3 \times 10^{10}$</td>
<td>$4 \times 10^{14}$</td>
<td>$5 \times 10^{13}$</td>
<td>$3.1 \times 10^{10}$</td>
</tr>
<tr>
<td>Ra-226</td>
<td>$2 \times 10^9$</td>
<td>$2 \times 10^{11}$</td>
<td>$4 \times 10^{12}$</td>
<td>$2.3 \times 10^9$</td>
</tr>
<tr>
<td>Pb-210</td>
<td>$3 \times 10^9$</td>
<td>$3 \times 10^{12}$</td>
<td>$8 \times 10^{13}$</td>
<td>$3.4 \times 10^9$</td>
</tr>
</tbody>
</table>
TABLE III. INTEGRATED COMMITTED EFFECTIVE DOSES (Sv) AS BEST ESTIMATE (B.E.) AND FOR A 95% CONFIDENCE INTERVAL AT DIFFERENT INTEGRATION TIMES AFTER DAM BREAK AT THE SILLAMÄE WASTE REPOSITORY

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Integration time</th>
<th>B.E.</th>
<th>95%</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-238</td>
<td>1 week</td>
<td>$7 \times 10^{12}$</td>
<td>$(0.6-30) \times 10^{12}$</td>
</tr>
<tr>
<td></td>
<td>1 month</td>
<td>$1 \times 10^{10}$</td>
<td>$(0.2-6) \times 10^{10}$</td>
</tr>
<tr>
<td></td>
<td>1 year</td>
<td>$6 \times 10^{9}$</td>
<td>$(1-20) \times 10^{9}$</td>
</tr>
<tr>
<td></td>
<td>50 years</td>
<td>$5 \times 10^{7}$</td>
<td>$(0.7-20) \times 10^{7}$</td>
</tr>
<tr>
<td>U-234</td>
<td>1 week</td>
<td>$7 \times 10^{12}$</td>
<td>$(1-20) \times 10^{12}$</td>
</tr>
<tr>
<td></td>
<td>1 month</td>
<td>$1 \times 10^{10}$</td>
<td>$(0.3-4) \times 10^{10}$</td>
</tr>
<tr>
<td></td>
<td>1 year</td>
<td>$6 \times 10^{9}$</td>
<td>$(1-20) \times 10^{9}$</td>
</tr>
<tr>
<td></td>
<td>50 years</td>
<td>$5 \times 10^{7}$</td>
<td>$(0.9-20) \times 10^{7}$</td>
</tr>
<tr>
<td>Th-232</td>
<td>1 week</td>
<td>$3 \times 10^{14}$</td>
<td>$(0.07-10) \times 10^{14}$</td>
</tr>
<tr>
<td></td>
<td>1 month</td>
<td>$8 \times 10^{13}$</td>
<td>$(0.2-50) \times 10^{13}$</td>
</tr>
<tr>
<td></td>
<td>1 year</td>
<td>$2 \times 10^{10}$</td>
<td>$(0.04-10) \times 10^{10}$</td>
</tr>
<tr>
<td></td>
<td>50 years</td>
<td>$1 \times 10^{8}$</td>
<td>$(0.02-6) \times 10^{8}$</td>
</tr>
<tr>
<td>Ra-226</td>
<td>1 week</td>
<td>$2 \times 10^{13}$</td>
<td>$(0.04-9) \times 10^{13}$</td>
</tr>
<tr>
<td></td>
<td>1 month</td>
<td>$8 \times 10^{12}$</td>
<td>$(0.1-50) \times 10^{12}$</td>
</tr>
<tr>
<td></td>
<td>1 year</td>
<td>$2 \times 10^{9}$</td>
<td>$(0.02-10) \times 10^{9}$</td>
</tr>
<tr>
<td></td>
<td>50 years</td>
<td>$2 \times 10^{7}$</td>
<td>$(0.02-10) \times 10^{7}$</td>
</tr>
<tr>
<td>Pb-210</td>
<td>1 week</td>
<td>$3 \times 10^{12}$</td>
<td>$(0.3-10) \times 10^{12}$</td>
</tr>
<tr>
<td></td>
<td>1 month</td>
<td>$2 \times 10^{10}$</td>
<td>$(0.1-7) \times 10^{10}$</td>
</tr>
<tr>
<td></td>
<td>1 year</td>
<td>$4 \times 10^{8}$</td>
<td>$(0.1-20) \times 10^{8}$</td>
</tr>
<tr>
<td></td>
<td>50 years</td>
<td>$6 \times 10^{7}$</td>
<td>$(0.2-30) \times 10^{7}$</td>
</tr>
</tbody>
</table>

For all nuclides included in these calculations the dose from the daughter nuclides can be neglected compared with the contribution from the parent nuclide.

At a dam collapse, the release of uranium with the pond water will be of the order of 10-50% of the annual leakage via groundwater. Consequently, this release will contribute relatively little to the integrated doses. The higher exposure from
TABLE IV. COLLECTIVE COMMITTED EFFECTIVE DOSES (man·Sv) FOR THE TWO RELEASE SCENARIOS\(^a\)

| Nuclide | Continuous release | Dam break | | | |
|---------|-------------------|-----------|---|---|
|         |                   | B.E. 95%  |   |   |
| U-238   | 0.5               | 0.6       | 0.1-2 | |
| U-234   | 0.6               | 0.6       | 0.1-2 | |
| Th-232  | 2 x 10\(^5\)     | 6 x 10\(^4\) | (0.09-50) x 10\(^3\) | |
| Ra-226  | 3 x 10\(^3\)     | 0.2       | 0.003-1 | |
| Pb-210  | 3 x 10\(^3\)     | 0.2       | 0.003-1 | |

\(^a\) Integration period 50 years; best estimate, and for the dam break case 95% confidence interval.

\(^{232}\)Th, \(^{226}\)Ra and \(^{210}\)Pb depends on the long term leakage from waste in the sediment of compartment 1.

In general, model calculations have an inherent uncertainty. This uncertainty arises from the different steps involved, such as description of the scenario, consideration of the relevant processes and choice of the parameter values. The contribution of the latter is partly considered in this case by the uncertainty analyses carried out. It is not always possible to estimate in detail the importance of the contribution of each source to the uncertainty. The calculations have shown that the major sources of error are as follows.

First, the waste is an inhomogeneous mixture and hence it is difficult to estimate the actual amounts of elements. Second, it is necessary in this study to estimate the dispersion volumes and turnover rates of water. Third, the mobility of the nuclides may be underestimated if organic complexes are not considered. On the other hand, coupled equilibria were not considered in the calculation of the solubility. This might reduce the assumed ability of the water to keep all elements in solution. Further sources of uncertainty are bioaccumulation factors and biological turnover times in fish.

One general conclusion from this study is that the doses are low — only several per mille of the general background values. The overall risk to man will be negligible.
ACKNOWLEDGEMENTS

This study was partly financially supported by the Swedish Radiation Protection Institute.

REFERENCES


MIGRATION OF RADIOACTIVE CONTAMINANTS DISCHARGED INTO THE RIVER YENISEI BY AN INDUSTRIAL PLANT BETWEEN 1960 AND 1993

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Abstract


The radioactive contamination of water, bottom sediments and fish of the Yenisei river ecosystem over a stretch of 2000 km from the site of discharges from the Krasnoyarsk Mining and Chemical Industrial Complex (KMCIC) is analysed on the basis of observational data from many years (1971-1993). Exposures of aquatic organisms in the distant zone and the near zone of KMCIC are estimated. Estimated doses and the radiation risk of the population consuming fish from the Yenisei river are presented.

1. INTRODUCTION

The Krasnoyarsk Mining and Chemical Industrial Complex (KMCIC) is located at a distance of 40 km from the city of Krasnoyarsk in Russia. For over thirty years, KMCIC has been a source of radioactive contamination of the environment, resulting from gas and aerosol releases, as well as liquid discharges of radionuclides. In 1992, two industrial straight-through reactors at KMCIC, which discharged their cooling waters near the right bank of the Yenisei river at a distance of 80 km from Krasnoyarsk, were stopped and removed from service. This resulted in considerable reduction of the radioactive contamination of the environment. The remaining reactor at KMCIC is currently in operation and is used for the heating of Krasnoyarsk-26. The results of investigations of the radiation situation in the Yenisei river basin, performed during a scientific expedition in August 1991, have been published in Ref. [1]. The present paper deals with an analysis of radioactive contamination levels and an assessment of the impact of KMCIC on the radioecological state of the Yenisei river, on the basis of observational data from many years (1971-1993).
2. RADIOACTIVE CONTAMINATION OF THE YENISEI RIVER

In the period from 1971 to 1993, scientific expeditions with the aim of surveying the radiation situation in the Yenisei river have been carried out on a stretch of about 2000 km of the river, from Krasnoyarsk to the small town Dudinka and further downstream (Fig. 1).

Among the most informative indications of radioactive contamination of the river ecosystem were the radionuclide concentrations in bottom sediments. Table I presents some results of a survey of bottom sediment contamination performed in August 1973. The major area of the river bottom is covered with coarse sand, pebble and gravel. Areas with a soft silty bottom are comparatively rare. These are, however, precisely the parts that are characterized by increased accumulation of radionuclides. Bottom sediment samples have been collected in 16 sections. The data for four of them are given in Table I. The highest levels of radioactive contamination of the river bottom were observed near the source of radionuclide discharge, where the total radionuclide density was as high as 5800 kBq/m². More than half of this density was accounted for by two radionuclides: ⁵¹Cr and ⁶⁵Zn. At a distance of 250 km from the source of discharge, practically the same radionuclides as in the near zone were observed; however, the total radionuclide density decreased by almost an order of magnitude. At a distance of 800 km from the source, the level of radioactive contamination of the river bottom decreased by an order of magnitude, with approximately 70% of this level being accounted for by ⁶⁵Zn and ¹³⁷Cs. At a distance of 1930 km from the source, the total activity of bottom sediments from technogenic radionuclides decreased by another order of magnitude and amounted to 5 kBq/m². Thus, the investigations of the radioactive contamination of the river bottom showed that the area impacted by KMCIC is about 2000 km. The levels of bottom sediment contamination at the boundary of the zone far from KMCIC are one thousand times lower than those in the near zone; however, they are higher than the level of contamination due to global fallout.

Components of the river ecosystem other than bottom sediments were also investigated during these scientific expeditions. Table II presents data on the radionuclide concentrations in fish. According to observational data, one of the major radionuclides accumulated in fish is ³²P. Noticeable amounts of this radionuclide were detected even at a distance of over 1400 km from the source of discharge. Table III gives data on the radionuclide concentrations in the river water in the near zone and the far zone of KMCIC. Outside the near zone, the water contamination level decreases markedly, although trace amounts of artificial radionuclides are detected at a considerable distance (over 1300 km) from the source of discharge. However, their content in the river water is lower by several orders of magnitude than the threshold levels stipulated by radiation safety norms. Following the removal from service of two straight-through reactors in 1992, the radioactive contamination of the river water was considerably reduced.
According to observational data, most of the radionuclides ($^{46}$Sc, $^{54}$Mn, $^{58}$Co, $^{59}$Fe, $^{60}$Co, $^{65}$Zn and others) were transported on suspended particles, except for $^{51}$Cr, which was mainly in dissolved form. The stage of $^{32}$P and $^{137}$Cs was intermediate, with their concentration in suspension amounting to about half of the total content in water.
TABLE I. RADIONUCLIDE CONCENTRATIONS (Bq/kg dry weight) IN BOTTOM SEDIMENTS OF THE YENISEI RIVER (0-5 cm layer) IN AUGUST 1973

<table>
<thead>
<tr>
<th>Radionuclides</th>
<th>Distance from the source of discharge (km)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>6</td>
</tr>
<tr>
<td>Sc-46</td>
<td>5 550</td>
</tr>
<tr>
<td>Cr-51</td>
<td>26 000</td>
</tr>
<tr>
<td>Mn-54</td>
<td>3 700</td>
</tr>
<tr>
<td>Co-58</td>
<td>3 300</td>
</tr>
<tr>
<td>Co-60</td>
<td>7 400</td>
</tr>
<tr>
<td>Fe-59</td>
<td>6 700</td>
</tr>
<tr>
<td>Zn-65</td>
<td>25 000</td>
</tr>
<tr>
<td>Cs-134</td>
<td>2 400</td>
</tr>
<tr>
<td>Cs-137</td>
<td>7 800</td>
</tr>
<tr>
<td>Ce-144</td>
<td>1 850</td>
</tr>
<tr>
<td>Eu-152</td>
<td>3 700</td>
</tr>
<tr>
<td>Eu-154</td>
<td>1 100</td>
</tr>
</tbody>
</table>

Radionuclide density (kBq/m²) | 5 800 | 620 | 60 | 5 |

3. RADIOECOLOGICAL ASSESSMENT

An assessment of the radioecological state of the Yenisei river was made on the basis of an assessment of the exposure of the population and the aquatic biota, i.e. on the basis of hygienic and ecological criteria [2].

Table IV presents estimated dose rates for aquatic organisms in the near zone of KMCIC, calculated from observational data on radioactive contamination of components of the Yenisei river ecosystem by the methods discussed in Refs [3, 4]. The actual values of the dose rates at the site of radionuclide discharge may exceed the assessed values given in the table. Practically for all groups of organisms the values of the absorbed dose from artificial radionuclides in the 15 km zone are substantially
TABLE II. RADIONUCLIDE CONCENTRATIONS (Bq/kg fresh weight) IN FISH OF THE YENISEI RIVER IN AUGUST 1973

<table>
<thead>
<tr>
<th>Species of fish</th>
<th>Distance from the source of discharge (km)</th>
<th>Radionuclides</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$^{32}$P</td>
</tr>
<tr>
<td>Dace</td>
<td>190</td>
<td>2800</td>
</tr>
<tr>
<td>Soroga</td>
<td>190</td>
<td>3600</td>
</tr>
<tr>
<td>Perch</td>
<td>190</td>
<td>2200</td>
</tr>
<tr>
<td>Perch</td>
<td>330</td>
<td>600</td>
</tr>
<tr>
<td>Dace</td>
<td>600</td>
<td>2800</td>
</tr>
<tr>
<td>Soroga</td>
<td>600</td>
<td>1040</td>
</tr>
<tr>
<td>Pike</td>
<td>1230</td>
<td>600</td>
</tr>
<tr>
<td>Soroga</td>
<td>1230</td>
<td>600</td>
</tr>
<tr>
<td>Dace</td>
<td>1230</td>
<td>740</td>
</tr>
<tr>
<td>Dace</td>
<td>1460</td>
<td>270</td>
</tr>
<tr>
<td>Soroga</td>
<td>1460</td>
<td>20</td>
</tr>
</tbody>
</table>

Higher than the natural radiation background. The highest values of absorbed dose are found in macrophytes, molluscs and fish. The short lived radionuclides $^{24}$Na, $^{32}$P and $^{56}$Mn contribute largely to the value of the dose from technogenic radionuclides. Outside the near zone, the doses for aquatic organisms decrease considerably. Table V presents the estimated annual dose rates for fish, which depend on the distance from the source of radionuclide discharge. Additional exposure of fish to artificial radionuclides in the Yenisei river takes place both downstream and upstream of KMCIC; this is associated with biotic transfer of radionuclides during migration of fish. Outside the near zone, the dose rates for fish are within the range of the so-called 'low doses'. However, even at a distance of 250 km from the source of discharge these values often exceed the natural radiation background level.
<table>
<thead>
<tr>
<th>Radionuclides</th>
<th>Distance from the source of discharge (km)</th>
<th>1985–1992 (average)</th>
<th>1992</th>
<th>1993</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sc-46</td>
<td>250</td>
<td>11 ± 6</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>850</td>
<td>5 ± 3</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>1360</td>
<td>5 ± 3</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Cr-51</td>
<td>250</td>
<td>1900 ± 1200</td>
<td>700 ± 80</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>850</td>
<td>1100 ± 700</td>
<td>150 ± 140</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>1360</td>
<td>700 ± 400</td>
<td>160 ± 140</td>
<td>—</td>
</tr>
<tr>
<td>Mn-54</td>
<td>250</td>
<td>8 ± 4</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>850</td>
<td>3 ± 2</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>1360</td>
<td>3 ± 2</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Co-58</td>
<td>250</td>
<td>26 ± 12</td>
<td>4 ± 2</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>850</td>
<td>12 ± 8</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>1360</td>
<td>7 ± 4</td>
<td>10 ± 8</td>
<td>—</td>
</tr>
<tr>
<td>Co-60</td>
<td>250</td>
<td>7 ± 5</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>850</td>
<td>4 ± 2</td>
<td>8 ± 4</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>1360</td>
<td>4 ± 2</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Fe-59</td>
<td>250</td>
<td>42 ± 20</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>850</td>
<td>7 ± 3</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>1360</td>
<td>8 ± 4</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Zn-65</td>
<td>250</td>
<td>23 ± 10</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>850</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>1360</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Cs-137</td>
<td>250</td>
<td>6 ± 3</td>
<td>2 ± 1</td>
<td>1.0 ± 0.5</td>
</tr>
<tr>
<td></td>
<td>850</td>
<td>5 ± 2</td>
<td>6 ± 4</td>
<td>2 ± 1</td>
</tr>
<tr>
<td></td>
<td>1360</td>
<td>5 ± 3</td>
<td>2 ± 1</td>
<td>2 ± 1</td>
</tr>
</tbody>
</table>
**TABLE IV. ESTIMATED DOSE RATES (mGy/d) FOR AQUATIC ORGANISMS IN THE NEAR ZONE (15 km) OF KMCIC (1991)**

<table>
<thead>
<tr>
<th>Organisms</th>
<th>Absorbed dose</th>
<th>Natural background</th>
</tr>
</thead>
<tbody>
<tr>
<td>Phytoplankton</td>
<td>0.01–0.1</td>
<td>0.0005–0.002</td>
</tr>
<tr>
<td>Zooplankton</td>
<td>0.03–0.4</td>
<td>0.001–0.002</td>
</tr>
<tr>
<td>Macrophytes</td>
<td>1.0–4.5</td>
<td>0.001–0.01</td>
</tr>
<tr>
<td>Molluscs</td>
<td>1.0–2.5</td>
<td>0.002–0.01</td>
</tr>
<tr>
<td>Fish</td>
<td>0.3–1.6</td>
<td>0.002–0.01</td>
</tr>
</tbody>
</table>

*1 Gy = 1 J/kg.*

**TABLE V. ESTIMATED ANNUAL DOSE RATES FOR FISH IN THE YENISEI RIVER**

<table>
<thead>
<tr>
<th>Distance from the source of discharge (km)</th>
<th>Dose rate from incorporated technogenic radionuclides (cGy/a)*</th>
</tr>
</thead>
<tbody>
<tr>
<td>80 (upstream of KMCIC)</td>
<td>0.02–0.2</td>
</tr>
<tr>
<td>16 (downstream of KMCIC)</td>
<td>0.40–1.3</td>
</tr>
<tr>
<td>60</td>
<td>0.30–8.0</td>
</tr>
<tr>
<td>250</td>
<td>0.13–1.7</td>
</tr>
<tr>
<td>850</td>
<td>0.003–0.01</td>
</tr>
<tr>
<td>1360</td>
<td>0.002–0.007</td>
</tr>
<tr>
<td>Natural radiation background</td>
<td>0.07–0.40</td>
</tr>
</tbody>
</table>

*1 Gy = 1 J/kg.*
4. CONCLUSIONS

According to results from observations and radioecological assessments made on the basis of these results, the discharge of radionuclides from KMCIC has a local impact and a regional impact on the Yenisei river ecosystem. An important task of further studies is a reconstruction of the doses from exposures and the radiation risks for the whole period of operation of KMCIC.

REFERENCES


RADIONUCLIDES IN THE TERRESTRIAL ENVIRONMENT

(Session 4)

Chairman

L. BULDAKOV
Russian Federation
RADIONUCLIDE DYNAMICS DURING LITTER DECOMPOSITION IN A HOLM-OAK FOREST

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Abstract

RADIONUCLIDE DYNAMICS DURING LITTER DECOMPOSITION IN A HOLM-OAK FOREST.

Migration of $^{134}$Cs and $^{110}$Ag$^m$ in a Mediterranean forest soil was studied by means of field incubation of artificially contaminated holm-oak leaves in plastic cylinders. The contaminated green leaves in the cylinders replaced the original litter layer, and the litter decomposition processes were studied. The following layers were sampled: L (remaining litter), F (fermented litter), H (humus layer) and mineral soil layers. The fraction of radionuclides that had migrated from the L-layer accumulated mainly in the F- and H-layers, whereas very little activity reached the mineral soil after an initial leaching period. During the second year of incubation, a significant transfer from the F-layer to the H-layer was observed for both radionuclides. At the first sampling, it was found that leaching of radionuclides from the L-layer was equivalent to the water extractable fraction of radionuclides obtained by sequential extraction. From this period onwards, radionuclide release was dependent on litter decomposition. A positive linear correlation was found between mass loss and radionuclide release after the first leaching period. The regression coefficient was 0.57 for $^{134}$Cs and 0.56 for $^{110}$Ag$^m$.

In agreement with these observations, radionuclide release from the litter layer followed a double negative exponential function. Radionuclide dynamics in the F-layer followed three different periods: In the first period, rapid accumulation of radionuclides occurred in the F-layer as a consequence of initial leaching from the L-layer. In the second period, there was both transfer from the L-layer and transfer to the H-layer of radionuclides, which was mainly due to degradation of easily decomposable compounds. The F-layer lost about 70% of its weight, but the radionuclide content remained constant. It is suggested that in this period, retention of radionuclides by recalcitrant compounds occurred. Micro-organisms could also play an important role in consuming carbon, with subsequent immobilization of radionuclides. Finally, a third period, with both carbon and radionuclide mineralization, was observed at the end of the incubation period. It can be concluded that decomposition processes and organic matter dynamics in the L- and F-layers are key factors controlling cycling of radionuclides in forest systems.
1. INTRODUCTION

Forest ecosystems were among the most contaminated environments after the Chernobyl accident. A general model for radiocaesium cycling in forests [1] showed that forest soil was the major compartment accumulating radionuclides.

Several studies reported a high capacity of the forest floor to retain radionuclides. This capacity has been attributed to biological retention [2] or specific retention by the lignin fraction [3]. Further mobility of radionuclides in the forest floor is initially regulated by litter decomposition and, secondly, by interaction of the radionuclides with the humic substances and clay minerals [4].

A review of radiocaesium cycling in forests [5] showed the necessity for a better understanding of the role of litter decomposition in radiocaesium availability, as well as the importance of better identification of the mechanisms involved in the biogeochemical process of caesium availability and uptake in forest soils.

The aim of this paper is to study the relationship between organic matter decomposition and radionuclide dynamics as a key process for modelling the cycle of radionuclides in forest soils.

2. MATERIALS AND METHODS

2.1. Experimental design

Migration of $^{134}$Cs and $^{110}$Ag$^m$ in a mediterranean forest soil was studied by means of field incubation of artificially contaminated green holm-oak leaves in plastic cylinders. The radioactive aerosol used in sample contamination was a thermo-generated aerosol containing $^{134}$Cs, $^{85}$Sr and $^{110}$Ag$^m$. The contaminated green leaves, placed in the cylinders, replaced the original litter layer, and the litter decomposition processes were studied. Over a period of two years, nine sample collections were performed. For each cylinder the following layers were sampled separately: L (remaining litter), F (fermented litter), H (humus layer), A1 (0–2.5 cm depth mineral soil) and A2 (2.5–5 cm depth mineral soil). More details of the experimental design and sampling methodology are given in Ref. [6].

2.2. Analytical techniques

The water content in the freshly contaminated green leaves and in L-layer samples was determined from a subsample dried at 60°C. Samples from the L- and F-layers were ground; samples from the H-layer and from mineral soil were passed through a sieve of 2 mm mesh size.

Activity measurements of $^{134}$Cs and $^{110}$Ag$^m$ were carried out by high resolution gamma spectrometry, using an intrinsic germanium detector (ORTEC
GMX15200-P), with 21.7% relative efficiency and 1.89 keV of resolution (FWHM) at 1.33 MeV. The radionuclide activities were decay corrected to the date of contamination. The radionuclide content in each layer sampled is expressed as the percentage of the initial contamination of the samples.

2.3. Statistical techniques

The significant differences in radionuclide content and standing mass of the F-layer were examined by analyses of variance and a test of the range of least significant differences (LSD). Pearson correlation was used to test the significance of mass loss and radionuclide release from the L-layer. All levels of significance are referred to at the 1% level. The SPSS program [7] was used to perform statistical analysis.

3. RESULTS AND DISCUSSION

3.1. Radionuclide migration

Figure 1 shows the distribution of $^{134}$Cs in the different layers during the incubation period. During the first year, radiocaesium, which had migrated from the
FIG. 2. Distribution of $^{110}$Ag$^{m}$ in the forest floor and in topsoil layers during the incubation period. Mean values ($n = 4$) with standard errors.

L-layer, accumulated mainly in the F-layer; from this period onwards, progressive transfer from these layers to the H-layer was observed. Migration to the deeper layers occurred rapidly and very little activity reached the mineral soil after this initial leaching period. These results are in agreement with those found after the Chernobyl accident in Mediterranean conditions [8]. At the end of the study, the H-layer contained the highest amount of radionuclides. In this layer, a large proportion of radiocaesium remains in available form, and therefore it can be an important source of radiocaesium uptake by plant roots.

Silver-$^{110}$m showed a similar behaviour, but with slower migration (Fig. 2), according to the observed higher incorporation into litter [9].

3.2. Radionuclide cycling

Litter decomposition, measured as mass loss, was about 60% for the whole period. The mass loss in litter bags used as a control for this method was about 35%. The difference is attributed to litter comminution by the soil fauna, which seems to enhance both litter decomposition and radionuclide release [6].

Figure 3 shows the relationship between the percentage of radionuclide remaining and litter mass loss. In the initial period, leaching of both radionuclides
FIG. 3. Relationship between the remaining litter mass and the radionuclide activity in the L-layer during the incubation period.

was equivalent to the water extractable fraction of radionuclides obtained by sequential extraction [9]. From this period onwards, radionuclide release was dependent on litter decomposition. A positive linear correlation was found between mass loss and radionuclide release after the first leaching period. The regression coefficient was 0.57 for $^{134}\text{Cs}$ and 0.56 for $^{110}\text{Ag}$m.

In agreement with these observations, radionuclides released from the litter layer (Figs 1 and 2) followed a double negative exponential function:

$$^{134}\text{Cs}: \quad \text{RA} = 66.13 e^{-0.039t} + 32.28 e^{-0.0015t}$$

$$^{110}\text{Ag}m: \quad \text{RA} = 29.88 e^{-0.015t} + 71.92 e^{-0.0014t}$$

where RA is the remaining activity and t is the incubation time (days).

The first part of the exponential, with a higher migration rate, is attributed to leaching processes; the second part, with a lower rate, is related to decomposition processes associated with litter mass loss. Leaching of $^{134}\text{Cs}$ is faster than that of $^{110}\text{Ag}$m, but the release due to decomposition is similar for both radionuclides. Other studies reported lower migration rates for radiocaesium [10] and different release rates, depending on the kind of plant litter under the same experimental con-
FIG. 4. Relationship between the standing mass and the $^{134}\text{Cs}$ activity in the F-layer during the study period.

ditions [11]. Rafferty and Kliashstorin [12] carried out a study in Chernobyl and Ireland and reported that the radiocaesium content in the L-layer remained constant during litter decomposition. These results are in contrast to those obtained in the present work. This could be due to the different nature of litter in the two experiments or, more probably, it could be related to the fact that, in the year when the experiment was carried out, the litter in Chernobyl was not contaminated by deposition of radionuclides (as was the litter in our experiment), but contamination was due to root absorption, which could affect the form of radiocaesium in the leaves. It is well known that during the decomposition process, the differential release of mineral elements attached to litter depends to a large extent on the kind of chemical bounds prevailing for these elements. Therefore, the radionuclide content in the different pools (labile or recalcitrant) of initial litter could determine further migration as a consequence of the differential decomposition rates for these pools.

The relationship between the standing mass of the F-layer and radionuclide accumulation in the F-layer can be seen in Figs 4 and 5. These results can be explained as follows: In the initial period, rapid accumulation of radionuclides occurred in the F-layer as a consequence of initial leaching from the L-layer (50 days). In the second period, there was both radionuclide transfer from the L-layer and transfer to the H-layer, related to easily decomposable compounds
FIG. 5. Relationship between the standing mass and the $^{110}\text{Ag}^m$ activity in the F-layer during the study period.

(600 days). The F-layer lost about 70% of its weight, but the radionuclide content remained constant. It is suggested that during this period, retention of radionuclides by recalcitrant compounds occurred. Micro-organisms also play an important role in consuming carbon, with subsequent immobilization of radionuclides [11, 13]. Finally, a third period, with both carbon and radionuclide mineralization, was observed towards the end of the incubation period.

It can be concluded that decomposition processes and organic matter dynamics in the L- and F-layers are key factors controlling radionuclide cycling in forest systems.

A mechanistic model for litter decomposition, including factors such as litter quality, forest floor and soil microclimate, and biological activity, will prove useful in modelling radionuclide behaviour in forest systems.

ACKNOWLEDGEMENTS

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REFERENCES


AGGREGATED TRANSFER COEFFICIENTS

A simple approach to modelling transfer of radionuclides to food products from semi-natural ecosystems

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Abstract

AGGREGATED TRANSFER COEFFICIENTS: A SIMPLE APPROACH TO MODELING TRANSFER OF RADIONUCLIDES TO FOOD PRODUCTS FROM SEMI-NATURAL ECOSYSTEMS.

A recent VAMP (Validation of Environmental Model Predictions) report collated values for aggregated transfer coefficients (T_{ag}) which can describe the integrated transfer of radiocaesium to food products from semi-natural ecosystems. Further T_{ag} data are presented for sheep and mushrooms which suggest that subdivision of T_{ag} values to take soil type and seasonal variation into account is potentially valuable. For sheep, T_{ag} values for organic soils are generally one to two orders of magnitude higher than those for sandy and clay soils. For most other semi-natural foodstuffs there are currently inadequate data to make such subdivisions.

1. INTRODUCTION

The Chernobyl accident focused attention on the transfer of radiocaesium through natural and semi-natural ecosystems to man. In semi-natural ecosystems high, and in some cases prolonged, transfer of radiocaesium into a wide range of foodstuffs has occurred. Furthermore, within the same species, high variation is often found in radiocaesium contamination levels.
Models developed before the Chernobyl accident were generally designed to consider agricultural food production, and they predicted radionuclide levels in such foodstuffs reasonably well. However, these models failed to predict adequately radionuclide levels in natural food products from semi-natural ecosystems. Semi-natural ecosystems were previously considered only to a limited extent (with the notable exception of reindeer) because the quantities of food consumed from these systems were assumed to be comparatively small and confined to a few individuals. They were therefore usually not considered in dose assessments.

After the Chernobyl accident it became evident that although semi-natural ecosystems provide comparatively small quantities of food in many countries, the long term dose commitment from these ecosystems to humans can be significant, particularly for radiocaesium. This is primarily due to the high levels of radiocaesium found in many semi-natural food products and the long effective half-lives of $^{137}\text{Cs}$ observed in some of these ecosystems compared with agricultural systems [1–4]. Whilst high radiocaesium levels in natural food products often persisted, those for agricultural areas generally declined quickly, so that with each year after the deposition of fallout the comparative importance of transfer of radiocaesium by natural food products from semi-natural ecosystems to man increased. Intake of food from semi-natural ecosystems is not evenly distributed in the population, and certain critical groups, such as hunters, sheep and reindeer breeders and mushroom pickers, can have particularly large intakes of these food products [5–7].

Modelling transfers of radiocaesium to food and finally to man is difficult for semi-natural ecosystems; this is due to (i) a lack of understanding of the processes involved in controlling the fate of radionuclides in these complex ecosystems and (ii) the difficulty of modelling the often complex transfer processes present. Many different factors can affect radiocaesium behaviour in semi-natural ecosystems. In particular, there is a pronounced heterogeneity in soil properties in these semi-natural ecosystems. The low capacity of many of these soils to immobilize radiocaesium, because of low clay and potassium contents, low pHs and high organic matter contents [8], is one of the main factors responsible for persistently high radiocaesium levels in plants and animals. Consequently, the proportion of radiocaesium that resides in vegetation of semi-natural ecosystems is often much greater than that which occurs in agricultural ecosystems. Herbivores feeding on this vegetation vary widely in their feeding habits, and this leads to substantial variation in the levels of radiocaesium contamination, both between individuals within species and with time. Marked seasonal differences in radiocaesium levels in game result from changes in the availability of forage plants. High concentrations of radiocaesium activity have been reported for animal products from many semi-natural ecosystems and are due both to the high radiocaesium levels in the vegetation species eaten by animals and to the comparatively high transfer of radiocaesium to small ruminant species such as sheep, goats and roe-deer, which predominate in these ecosystems [4].
The transfer of radionuclides to plants and animals is often expressed using concentration ratios (CR), or for animals transfer coefficients (defined as the equilibrium ratio between the activity concentration in milk or meat divided by the daily intake). However, for animals in semi-natural ecosystems such transfer parameters are of limited value because of the uncertainties in estimating herbage intake or the radiocaesium content of ingested vegetation.

Given such difficulties and uncertainties, alternative methods of estimating transfer need to be explored for semi-natural ecosystems. This has been attempted in the VAMP report [9] by collating easily derived, empirical transfer coefficients, which integrate the transfer of radionuclides through one or several physico-chemical or biological steps. Such coefficients, termed aggregated transfer coefficients ($T_{ag}$), can be used in predictive models instead of the commonly used transfer parameters. Aggregated transfer coefficients are calculated using the expression:

$$T_{ag} = \frac{\text{activity concentration in the food product (Bq/kg or Bq/L)}}{\text{activity of deposit per unit area (Bq/m}^2\text{)}}$$

with units of m$^2$/kg or m$^2$/L. Activity levels are commonly expressed as fresh weight (fw) for animal products and dry weight (dw) for plant products. The $T_{ag}$ values primarily refer to the situation where flora and fauna are contaminated via root uptake from the soil and the directly intercepted component is negligible. Furthermore, $T_{ag}$ values cannot be used for prediction in conditions of continuous deposition. Any transfer parameter is of limited usefulness when trying to predict radiological impact, unless it is combined with estimates of time dependent changes in transfer. Data available for effective half-lives for semi-natural food products which can be combined with $T_{ag}$ values are discussed in the VAMP report.

A summary of the $T_{ag}$ values presented in the VAMP report is given in Table I. The range of variation in most $T_{ag}$ values observed for species other than fungi was about one order of magnitude. For fungi the spread of values was much greater, often exceeding two orders of magnitude within the same species of fungi.

Comparatively high $T_{ag}$ values were noted in a variety of circumstances, including:

- honey derived from bees with access to heather,
- sheep grazing on heather moorland,
- roe-deer in autumn,
- reindeer in winter,
- sheep with access to large quantities of mushrooms.

These high values were excluded when deriving the best estimate. However, it was evident that $T_{ag}$ values would be affected by soil, vegetation and seasonal factors, and further data were needed to determine the comparative effect of these sources of variation.
<table>
<thead>
<tr>
<th>Food product</th>
<th>( T_{\text{ag}} ) (m(^2)/kg)</th>
<th>Range</th>
<th>Best estimate</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Fungi (dw)</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><em>Cantharellus cibarius</em></td>
<td>0.01–0.96</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td><em>Cantharellus tubaeformis</em></td>
<td>0.14–1.5</td>
<td>0.7</td>
<td></td>
</tr>
<tr>
<td><em>Lactarius trivialis</em></td>
<td>0.02–5.2</td>
<td>1.8</td>
<td></td>
</tr>
<tr>
<td><em>Leccinum versipelle</em></td>
<td>0.005–0.74</td>
<td>0.1</td>
<td></td>
</tr>
<tr>
<td><em>Leccinum scabrum</em></td>
<td></td>
<td>0.03</td>
<td></td>
</tr>
<tr>
<td><em>Boletus edulis</em></td>
<td>0.003–0.38</td>
<td>0.07</td>
<td></td>
</tr>
<tr>
<td><em>Rozites caperata</em></td>
<td>0.78–3.1</td>
<td>1.9</td>
<td></td>
</tr>
<tr>
<td><em>Xerocomus badius</em></td>
<td>0.19–7.1</td>
<td>1.5</td>
<td></td>
</tr>
<tr>
<td><em>Xerocomus chrysenteron</em></td>
<td>0.01–5.2</td>
<td>1.0</td>
<td></td>
</tr>
<tr>
<td><strong>Berries (dw)</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cloudberry</td>
<td>0.002–0.23</td>
<td>0.1</td>
<td></td>
</tr>
<tr>
<td>Cowberry</td>
<td>0.032–0.04</td>
<td>0.04</td>
<td></td>
</tr>
<tr>
<td>Bilberry</td>
<td>0.017–0.041</td>
<td>0.04</td>
<td></td>
</tr>
<tr>
<td><strong>Honey (fw)</strong></td>
<td>0.00004–0.13</td>
<td>0.005</td>
<td></td>
</tr>
<tr>
<td><strong>Game animals (fw)</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Moose meat</td>
<td>0.006–0.032</td>
<td>0.02</td>
<td></td>
</tr>
<tr>
<td>Roe-deer meat</td>
<td>0.001–0.2</td>
<td>0.05</td>
<td></td>
</tr>
<tr>
<td>Red deer meat</td>
<td>0.02–0.07</td>
<td>0.04</td>
<td></td>
</tr>
<tr>
<td>Arctic hare</td>
<td>0.0009–0.13</td>
<td>0.03</td>
<td></td>
</tr>
<tr>
<td>Brown hare</td>
<td>0.0002–0.053</td>
<td>0.004</td>
<td></td>
</tr>
<tr>
<td><strong>Domesticated animals (fw)</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sheep meat</td>
<td>0.01–0.074</td>
<td>0.04</td>
<td></td>
</tr>
<tr>
<td>Goat milk</td>
<td>0.002–0.015</td>
<td>0.004</td>
<td></td>
</tr>
<tr>
<td>Cow milk</td>
<td>0.001–0.018</td>
<td>0.002</td>
<td></td>
</tr>
<tr>
<td>Beef</td>
<td></td>
<td>0.006</td>
<td></td>
</tr>
<tr>
<td>Reindeer (August)</td>
<td>0.025–0.1</td>
<td>0.1</td>
<td></td>
</tr>
<tr>
<td>(winter)</td>
<td>0.6–1.1</td>
<td>0.8</td>
<td></td>
</tr>
</tbody>
</table>
At the time of writing of this text it was difficult to assess the general applicability of the $T_{ag}$ values presented in the VAMP report. The authors concluded that further consideration needed to be given to the validity and usefulness of aggregated transfer coefficients and that, in addition to more information on half-lives and consumption rates, more data were needed for relevant $T_{ag}$ values for the most important food products from a wider range of semi-natural ecosystems, since $T_{ag}$ values would be expected to vary with ecosystem. A number of additional studies have now been carried out which make it possible to further evaluate the potential usefulness of aggregated transfer coefficients. The first study provides data for sheep, which might be expected to integrate heterogeneous deposition, by grazing over a large area. The second study provides data for mushrooms, whose hyphae take up nutrients (and radiocaesium) from a comparatively smaller area, but still integrate from a large area compared with that of the fruiting body itself.

1.1. Sheep

After the Chernobyl accident, high radioactivity levels were observed in sheep from semi-natural ecosystems in many affected countries, with the observed values exceeding the intervention limits in many European countries. For sheep grazing in semi-natural ecosystems the major factors responsible for the persistently high radiocaesium activities in meat are the high rates of root uptake of radiocaesium by grazed vegetation and, in certain areas, the selective intake by sheep of highly contaminated fungi and ericaceous species. For instance, in some of the contaminated pastures in Norway, fungi occasionally produce a substantial crop of fruiting bodies. When this happened in 1988, consumption of highly contaminated mushrooms by sheep led to a twofold increase in $T_{ag}$ values, rising from 0.063 m$^2$/kg in 1987 to 0.136 m$^2$/kg in 1988 [1, 10].

Although radiocaesium levels have declined in many areas, sheep from semi-natural ecosystems in Norway, Sweden, Ireland and the United Kingdom still exceed the intervention limits 9 years after the Chernobyl accident. This parallels the long effective half-life values which were calculated in Norway for $^{137}$Cs from above-ground nuclear weapons tests [1].

Sheep meat $T_{ag}$ values were divided in the VAMP report into data for large areas of unfenced land and data for more discrete pasture areas. As might be expected, the range in reported values was greater for the large areas at 0.01–0.15 m$^2$/kg fw than for the pasture sites, which ranged from 0.01 to 0.074 m$^2$/kg. The best estimate (0.04 m$^2$/kg) and range was based on the pasture values.

Recent data for sheep come from a Nordic programme study in which $T_{ag}$ values were compared for sheep meat in six different Nordic countries: Denmark, Faroe Islands, Finland, Iceland, Norway and Sweden [11]. The majority of lamb
TABLE II. SITE CHARACTERISTICS OF THE NORDIC SHEEP T<sub>ag</sub> STUDY (after Ref.[11])

<table>
<thead>
<tr>
<th>Country (code)</th>
<th>Main radiocaesium source</th>
<th>Environment</th>
<th>Soil type(s)</th>
<th>Organic matter (%)</th>
<th>Vegetation type(s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Denmark (DEN)</td>
<td>Nuclear weapons tests</td>
<td>Coastal</td>
<td>Sandy</td>
<td>8.2 ± 0.6</td>
<td>Permanent grassland</td>
</tr>
<tr>
<td>Faroe Islands (FAI)</td>
<td>Nuclear weapons tests</td>
<td>Coastal</td>
<td>Peaty</td>
<td>50.8 ± 21.2</td>
<td>Permanent grassland</td>
</tr>
<tr>
<td>Finland (FIN)</td>
<td>Chernobyl</td>
<td>Inland</td>
<td>Clay</td>
<td>16.6 (forest)</td>
<td>Natural pasture</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>13.5 (field)</td>
<td>(50% grass)</td>
</tr>
<tr>
<td>Iceland (ICE)</td>
<td>Nuclear weapons tests</td>
<td>Coastal</td>
<td>Peat, gravelly</td>
<td>52.0</td>
<td>Lowland mire</td>
</tr>
<tr>
<td>Norway (NOR)</td>
<td>Chernobyl</td>
<td>Coastal</td>
<td>Peaty</td>
<td>60.1 ± 25.5</td>
<td>Permanent grassland</td>
</tr>
<tr>
<td>Sweden (SWE)</td>
<td>Chernobyl</td>
<td>Mountain forest</td>
<td>Peat, gravelly, sandy moraine</td>
<td>10.0 ± 4.0</td>
<td>Mountain moor, Betula forest, permanent grassland</td>
</tr>
</tbody>
</table>

production in these Nordic countries takes place on uncultivated pastures or in semi-natural ecosystems. The study aimed at determining possible variation in T<sub>ag</sub> values caused by regional differences in factors such as climate, soil type, agricultural practices and other environmental factors.

The Nordic study was carried out at sites where no countermeasures had been taken. At each site, soil and herbage was collected and analysed for 137Cs, and lambs were either live-monitored or slaughtered and muscle analysed. Sampling took place yearly from 1990 to 1993; site characteristics are given in Table II.

The soil to herbage transfer values were highly variable and were greatly influenced by the particular plant species present. Where the same species had been sampled in different countries, the soil type was an important determinant of the radiocaesium level in plants. Soil to herbage transfer factors could be divided into two groups: comparatively high mean values of 0.018–0.083 m<sup>2</sup>/kg were recorded for the four sites with peaty soils, whilst comparatively low mean values of
0.00004–0.00032 m²/kg were found for the two sites with clay or sandy soil. The values declined in magnitude in the following country order:

Norway > Sweden > Iceland > Faroe Islands > Finland > Denmark

The range in transfer ratios for soil to herbage and in $T_{ag}$ values for lamb meat are shown for each year and country in Fig. 1. As would be expected, the mean ratio between the $^{137}$Cs levels in meat and vegetation was similar (0.58–0.70, using fw
TABLE III. $T_{ag}$ VALUES FOR SHEEP MEAT FROM DIFFERENT STUDIES

<table>
<thead>
<tr>
<th>Data set</th>
<th>Soil type</th>
<th>Range of $T_{ag}$ (m²/kg)</th>
<th>Best estimate (m²/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>VAMP — large area</td>
<td>Mostly upland organic soils</td>
<td>0.011-0.15</td>
<td></td>
</tr>
<tr>
<td>VAMP — pasture</td>
<td>Mostly upland organic soils</td>
<td>0.01-0.074</td>
<td>0.04</td>
</tr>
<tr>
<td>Nordic study</td>
<td>Organic soils</td>
<td>0.011-0.0619</td>
<td>0.04</td>
</tr>
<tr>
<td>— high range</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nordic study</td>
<td>Clay/sandy soils</td>
<td>0.00015-0.0055</td>
<td>0.001</td>
</tr>
<tr>
<td>— low range</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

a Incorporating data for individual years.

meat and dw plant) at five sites, although the values for the Faroe Islands were surprisingly low (0.27).

The mean $T_{ag}$ values for lamb meat were clearly divided into two groups, with higher values of 0.0148–0.0467 m²/kg, contrasting with low values of 0.0003-0.003 m²/kg. The values declined in magnitude in the following country order:

Sweden > Norway > Iceland > Faroe Islands > Finland > Denmark

with the exception of a forested part of the Finnish site, which was separately studied in 1993 and which had a comparatively high $T_{ag}$ value of 0.011 m²/kg.

It follows that, since, in general, the CR values were similar, the difference in $T_{ag}$ values must be due to variation in soil to herbage uptake and diet selection. Possibly, variation in $^{137}$Cs intake by grazing selection contributed to the enhanced $T_{ag}$ values for the site in Iceland compared with its rather lower transfer values from soil to vegetation.

Values from the Nordic study, with our judgement of recommended estimates, are compared with those from the VAMP report in Table III.

The improved range of data available for sheep suggests that $T_{ag}$ values can usefully be subdivided into two robust categories: one category suitable for sites with organic soils and one suitable for sites with sandy or clay soils.

When the comparative consumption of lamb from semi-natural ecosystems was taken into account (high in Norway, Iceland and the Faroe Islands, low elsewhere) the soil–herbage–lamb pathway for radiocaesium intake by man was estimated to be of greatest importance in Norway and Iceland, intermediate in the Faroe Islands and of comparatively lesser importance in Denmark, Finland and Sweden.
1.2. Fungi

High radiocaesium activities have been reported in the fruiting bodies (mushrooms) of a number of fungal species both before and after the Chernobyl accident [12–14]. However, radiocaesium levels are highly variable among fungal species (e.g. Ref. [15]), and within species the variation can be considerable [16, 17].

Since there are large variations both between and within different species, dose estimates based on the mean values given in Table I are likely to be highly uncertain and reflect the lack of data for calculating $T_{ag}$ values for fungi. Because of the high transfers of radiocaesium to fungi and the potential importance for radiocaesium transfer to humans, the authors of the VAMP report stated that systematic collection of dry weight activity concentrations in fungi and deposition values for the soil in which they are growing were needed.

An alternative approach to quantifying transfer has been adopted by Wirth et al. [18] who have calculated transfer ratios from soil to fungi, based on radiocaesium concentrations in mushrooms and only in the organic horizon of the soil. This method was suggested because fungal mycelia are preferentially found in organic horizons and are unlikely to absorb much radiocaesium from lower mineral horizons, where it will be strongly fixed. This approach would potentially reduce variation and would be most appropriate for saprophytic fungi, but possibly less useful for symbiotic species.

Further data for mushrooms have been collected in September 1994 from a site in the Rovno region of Ukraine [19], and $T_{ag}$ values calculated using both the VAMP $T_{ag}$ approach and that of Wirth et al. [18]. For two of the three mushroom species, for which duplicate samples were taken at the same forest site, using the $T_{ag}$ value based on radiocaesium present in only the organic layer reduced the coefficient of variation (CV) (Table IV). However, the differences were not striking, and this might be due to the feeding strategy of the species sampled.

The $T_{ag}$ values calculated using total deposition for Boletus edulis are in the maximum previously reported range of collated values in the VAMP report. Such high values may be due to the soil type on the study site, which is a highly organic peaty soil.

It is evident that the variation in $T_{ag}$ values for mushrooms is considerable. Initial data presented here suggest that the variation may be reduced for certain species by calculating $T_{ag}$ values with reference to the organic layer only. However, further data are needed, showing how $T_{ag}$ values vary with species, soil type and ecosystem, before subdivisions into hopefully less variable subgroups could be recommended. The potential importance of radiocaesium intake via mushrooms for critical groups is considerable and, therefore, any potential method of predicting radiocaesium contamination in mushrooms merits further investigation.
TABLE IV. $T_{ag}$ AND TRANSFER RATIO VALUES FOR RADIOCAESIUM IN MUSHROOM SPECIES COLLECTED IN UKRAINE IN 1994

<table>
<thead>
<tr>
<th>Mushroom species</th>
<th>Number</th>
<th>$T_{ag}$ (m$^2$/kg) (total deposition)</th>
<th>Transfer ratio (organic layer)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Mean</td>
<td>SD</td>
</tr>
<tr>
<td><strong>Boletus edulis</strong></td>
<td>8</td>
<td>0.45</td>
<td>0.35</td>
</tr>
<tr>
<td><strong>Boletus luteus</strong></td>
<td>3</td>
<td>1.43</td>
<td>1.01</td>
</tr>
<tr>
<td><strong>Boletus luridus</strong></td>
<td>4</td>
<td>4.80</td>
<td>1.70</td>
</tr>
</tbody>
</table>

2. CONCLUSION

Additional information on $T_{ag}$ values indicates that subdivision of $T_{ag}$ values to take soil groups and seasonal variation into account may improve the prediction capacity of this approach. Combining $T_{ag}$ values with information on soil groups potentially gives a robust, rapid method of predicting areas where there might be high, long term radiocaesium contamination of food products from semi-natural ecosystems. However, more data are needed for the different food products and different soil groups before subdivisions could be recommended for most food products.

ACKNOWLEDGEMENTS

Much of this paper summarizes data and discussion points presented in the VAMP $T_{ag}$ report [9], the Nordic study on sheep $T_{ag}$ values [11] and the ECP9 report of December 1994 (EC contract No. COSU-CT93-0050) [19]. The expertise of the authors of each of these documents has therefore contributed to this appraisal.

REFERENCES


[19] STRAND, P., HOWARD, B.J., SKUTERUD, L., AVERIN, V. (Eds), Fluxes of Radionuclides in Rural Communities in Russia, Ukraine and Belarus. Transfer of Radionuclide to Animals, their Comparative Importance under Different Agricultural Ecosystems and Appropriate Countermeasures, ECP9 Annual Report to the EC, DGXII-F-6, Nov. 93 — Dec. 94 (1994).
Abstract

APPARENT NON-LINEAR RELATIONSHIP OF RADIONUCLIDE CONCENTRATIONS BETWEEN CROP AND SOIL.

In general radiological assessments, the radionuclide concentration in crops has been assumed to be linearly related to that in soil. For this assumption to be valid, the soil-to-plant transfer factor must be independent of the radionuclide concentration. The actual transfer from soil to plant, however, seems to be inversely related to the soil contamination level. The paper presents a numerical study which explains why the caesium concentration in crops appears to be non-linearly related to that in soil, and a new model describing soil-to-plant transfer is proposed. It is shown that the variation of the soil sorption characteristics among the sampled soils causes the apparent, inversely proportional relationship.

1. INTRODUCTION

Accurate prediction of radionuclide transfer from soil to plants is an important but problematic subject. The difficulties in evaluating the internal dose from radionuclides through the food-chain were brought into focus by the Chernobyl accident. Large differences in evaluated internal doses were found between the data of the Government of the former USSR and those of the International Advisory Committee (IAC), although the external doses were almost the same [1]. This may be attributed to differences in the methods applied. Assessments made in the former USSR of ingestion of $^{137}\text{Cs}$ used an environmental transfer model assuming no restrictions on local food consumption, whereas the IAC calculated the dose more directly, using whole body counters. Plots of the average whole body incorporation of $^{137}\text{Cs}$ in selected population groups versus the concentration of $^{137}\text{Cs}$ in the surface soil (Fig. 1) show that the intake of $^{137}\text{Cs}$ by residents is inversely proportional to its concentration in the surface soil. This inversely proportional relationship might be partly due to the restriction of local food consumption. However, it seems doubtful that strict food control was carried out in accordance with the contamination level; this had not been clarified before the measurement. It is possible that the
tendency shown in the figure is attributable to the spatial difference in the characteristics of $^{137}$Cs transfer from soil to agricultural products. Konshin [2] observed an inversely proportional relationship between the radiocaesium concentration in the soil and the soil-to-plant transfer factor. A similar tendency was also found in soil-to-milk transfer [3]. These results call into question the transfer factor concept, which assumes linearity of the radionuclide concentrations in environmental components. We have carried out a numerical simulation to investigate the reason for this non-linear relationship, and we propose a new model describing soil-to-plant transfer of radionuclides.
2. MODELS AND PARAMETERS

For instantaneously contaminated soil, the radionuclide concentration in the surface soil can be predicted approximately by the following exponential equation [4]:

\[ C_{\text{soil}}(t) = C_{\text{soil}}(0) \exp(-\lambda t) \]  

(1)

where \( C_{\text{soil}} \) is the radionuclide concentration in the surface soil (Bq/g); \( t \) is the time (years) and \( \lambda \) is the effective removal constant (a\(^{-1}\)). Assuming that radionuclides in surface soil are removed by radioactive decay, downward infiltration and transfer to plants [4], \( \lambda \) can be written as

\[ \lambda = \lambda_d + \lambda_i + \lambda_c \]  

(2)

where \( \lambda_d \) is the radioactive decay constant (a\(^{-1}\)), \( \lambda_i \) is the removal rate by downward infiltration (a\(^{-1}\)) and \( \lambda_c \) is the removal rate by crop root uptake (a\(^{-1}\)). \( \lambda_d \) and \( \lambda_i \) can be described in detail, using more physically specific parameters:

\[ \lambda_d = \frac{\ln 2}{\theta_h} \]  

(3)

\[ \lambda_i = \frac{v_w}{W_{\text{soil}}(\theta_i + K_d)} \]  

(4)

where \( \theta_h \) is the half-life of the radionuclide (a); \( v_w \) is the velocity of water infiltration per unit area (L·m\(^{-2}\)·a\(^{-1}\)); \( W_{\text{soil}} \) is the amount of surface soil per unit area (kg/m\(^2\)); \( \theta_i \) is the soil water content by weight (L/kg); and \( K_d \) is the distribution coefficient for the soil solution (L/kg). The radionuclides are assumed to be uniformly distributed in surface soil cultivated annually. Because the rate of radionuclide uptake by a crop is dependent on the growth stage and is often discontinuous in one year, it is inappropriate to determine a constant value for \( \lambda_c \). If only the concentration of radionuclides at harvest time is considered, however, \( \lambda_c \) can be approximated as follows:

\[ \lambda_c = \frac{T_f Y_c}{W_{\text{soil}}} \]  

(5)

where \( T_f \) is the soil-to-plant transfer factor (Bq/g crop per Bq/g soil); and \( Y_c \) is the annual crop yield per unit area (kg/m\(^2\)). The above-ground parts of the crops, excluding the edible parts, are assumed to return to the soil after harvest. Substituting Eqs (2–5) into Eq. (1),

\[ C_{\text{soil}}(t) = C_{\text{soil}}(0) \exp\left\{-\left(\frac{\ln 2}{\theta_h} + \frac{v_w}{W_{\text{soil}}(\theta_i + K_d)} + \frac{T_f Y_c}{W_{\text{soil}}}\right)t\right\} \]  

(6)
The radionuclide concentration in the crop ($C_{\text{crop}}$) is calculated by multiplying $T_f$ by $C_{\text{soil}}$:

$$C_{\text{crop}}(t) = T_f C_{\text{soil}}(0) \exp\left\{-\left(\frac{\ln 2}{T_h} + \frac{v_w}{W_{\text{soil}}(\theta_1 + K_d)} + \frac{T_f Y_c}{W_{\text{soil}}}ight)t\right\} \quad (7)$$

$T_f$ has been obtained empirically as the concentration ratio between crop and soil. However, it is known that radionuclide ions are taken up from the soil solution and transferred to the root xylem through the surrounding cells [5, 6]. The upward migration in the xylem to the above-ground parts is considered to be promoted by the water movement, i.e. transpiration [6]. On the basis of this information, it is proposed to describe $T_f$ by

$$T_f = \frac{S_c T_c}{\theta_1 + K_d} \quad (8)$$

where $S_c$ is the selective absorption coefficient representing the ion permeability from soil solution to root xylem; and $T_c$ is the transpiration coefficient (L/kg) representing the transpiration volume per unit weight of the edible parts of the crop. In this equation, the value of $C_{\text{soil}}$ during the growth period is approximated as that at harvest. $S_c$ may be affected by the function of the root cell membrane and the osmotic potential in the soil solution. $T_c$ is possibly related to nutrient and meteorological conditions. Use of $K_d$ to predict radionuclide transfer from the liquid phase of the soil to plants has been proposed previously [7]. The velocity of water infiltration per unit area in Eq. (4) is expressed as

$$v_w = v_b - T_c Y_c \quad (9)$$

### TABLE I. VALUES OF PARAMETERS USED FOR CALCULATION

<table>
<thead>
<tr>
<th>$T_h$ (a)</th>
<th>$v_b$ (L·m$^{-2}$·a$^{-1}$)</th>
<th>$Y_c$ (kg·m$^{-2}$·a$^{-1}$)</th>
<th>$W_{\text{soil}}$ (kg/m$^2$)</th>
<th>$T_c$ (L/kg)</th>
<th>$S_c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td>$1.4 \times 10^3$ $^a$</td>
<td>3.4 $^b$</td>
<td>240 $^c$</td>
<td>36 $^d$</td>
<td>0.53 $^e$</td>
</tr>
</tbody>
</table>

- $^a$ Average annual rainfall in Tokyo during the past 30 years [8].
- $^b$ Areal average of the total annual yield of leafy vegetables in Japan [9].
- $^c$ Taken from the USNRC Regulatory Guide [10].
- $^d$ Average value for young Chinese cabbage ($Brassica campestris$) [11].
- $^e$ Value for young $Brassica campestris$ when the electrical conductivity in feed solution was 1.0 mS/cm [11].
where $v_b$ is the water infiltration velocity per unit area without plants (L·m$^{-2}$·a$^{-1}$). Substituting Eqs (8) and (9) into Eq. (7), with the assumption that the $K_d$ value is much higher than $\theta_i$, gives

$$C_{\text{crop}}(t) = \frac{S_c T_c}{K_d} C_{\text{soil}}(0) \exp\left\{ -\left( \frac{\ln2}{T_h} + \frac{v_b - T_c Y_c}{W_{\text{soil}} K_d} + \frac{S_c T_c Y_c}{K_d W_{\text{soil}}} \right) t \right\}$$

(10)

In the following section, the concentration of caesium in the crop is calculated using Eq. (10). The values of parameters used for calculation are shown in Table I.
FIG. 3. Chronological changes of the $^{137}$Cs concentration in the crop calculated with Eq. (10) when the initial concentration in the soil is 1.0 Bq/g. The values in parentheses are the $K_d$ percentiles in the cumulative probability distribution shown in Fig. 2.

$K_d = 2.2 \times 10^2$ (L/kg) (1)

$4.2 \times 10^2$ (5)

$1.9 \times 10^3$ (50)

$8.2 \times 10^2$ (95)

Elapsed time (years)

FIG. 4. Calculated caesium concentrations in soils and crops versus the distribution coefficient after 1, 3, 5, 10 and 20 years.
as constant values [8-11]. The assumed variation characteristics of $K_d$ for $^{137}$Cs are shown in Fig. 2; they have been obtained for 36 agricultural soils collected throughout Japan [12]. The initial $C_{\text{soil}}$ is set as 1.0 Bq/g just after harvest.

3. CALCULATION RESULTS AND DISCUSSION

The chronological changes of the $^{137}$Cs concentration in the crop, calculated for four $K_d$s, are shown in Fig. 3; these $K_d$s are 1, 5, 50 and 95 percentile values of the caesium $K_d$s shown in Fig. 2. It is found that $C_{\text{crop}}$ at lower $K_d$ is higher initially and decreases more rapidly; conversely, $C_{\text{crop}}$ at higher $K_d$ remains at a low

![Diagram](attachment:image.png)

**FIG. 5.** Apparent relationships between the calculated caesium concentrations in crops and soils.
level over the whole period. Such tendencies are attributed to the differences in the removal rates and the soil-to-plant transfer factors, both of which become higher in soil with lower $K_d$ values, as predicted from Eqs (4, 5) and Eq. (8), respectively. The calculated caesium concentrations in soils and crops after 1, 3, 5, 10 and 20 years are plotted as a function of $K_d$ in Fig. 4. Although both $C_{soil}$ and $C_{crop}$ decrease more rapidly in soil with lower $K_d$ values, they have inversely shaped curves; $C_{soil}$ is smaller but $C_{crop}$ is larger in soil with lower $K_d$ values. As a result of these differences, a non-linear, inversely proportional relationship appears between $C_{crop}$ and $C_{soil}$, as shown in Fig. 5.

This apparent relationship is not due to the dependence of the soil-to-plant transfer factor on concentration, but it is due to the variation of the distribution coefficients among sampled soils. It should be noted that the results obtained in this study do not deny the effectiveness of the transfer factor concept, i.e. the assumed linearity of the radionuclide concentrations between crop and soil on a local scale. Although it has been pointed out that the soil-to-plant transfer factor shows a dependence on the radionuclide concentration in the feed solution [13], the radionuclide concentration in a real soil solution does not seem to be so high that such a dependence needs to be considered for an individual crop. The non-linear relationship found in observations may be partly due to the spatial variation of soil sorption characteristics, although data for validation are insufficient.

To reduce the errors of model prediction for the soil-crop pathway, we propose that the radionuclide concentration in crops should be predicted on the basis of the concentration in the liquid phase of the soil, which can be estimated by the sorption intensity of the soil, i.e. the distribution coefficient. It is also desirable to clarify the contribution of other factors, such as the permeability of root cell membranes for each crop species, which can be represented by the selective absorption coefficient $S_c$, and the migration of radionuclides from the root xylem to the above-ground parts, which can be evaluated for leafy crops by the transpiration coefficient $T_c$. Such information will validate the proposed model (Eq. (10)), which should lead to an improvement in the reliability of environmental impact assessments for a terrestrial ecosystem.

ACKNOWLEDGEMENT

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REFERENCES


PROTECTION OF THE NATURAL ENVIRONMENT IN RELATION TO THE OPERATION OF SWEDISH NUCLEAR FACILITIES

Needs and criteria from the Authority point of view

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Abstract

PROTECTION OF THE NATURAL ENVIRONMENT IN RELATION TO THE OPERATION OF SWEDISH NUCLEAR FACILITIES: NEEDS AND CRITERIA FROM THE AUTHORITY POINT OF VIEW.

For the protection of the natural environment against detrimental effects of ionizing radiation, targets for protective actions have to be identified. Furthermore, it will be necessary to discuss criteria, possibly in quantitative terms, that can be monitored or predicted with reasonable accuracy. It is proposed that emphasis should be placed on biodiversity (biological diversity), encompassing intra- and inter-species variation as well as ecosystem structure. By protection of biodiversity, protection of biological resources and their use in local and international communities is inferred. While populations are generally the organizational level that should be protected, they may in certain cases be small, which would necessitate protection at the individual level also. Biodiversity can, however, only be monitored or predicted in detail in very rare cases. Actions taken for the protection of the environment will, therefore, have to ensure adequate protection of conditions governing biodiversity. This can be done by assessing doses to biotic components of the ecosystem. It remains to be discussed whether quantitative criteria should be established, but these would help in assessing both the impact on and the area of concern in the environment. On the basis of the lowest observed effect levels of ionizing radiation for individuals and a safety factor of 100-1000 to account for a range of risk factors, the quantitative criterion for protection of non-human species should probably be set at less than 50 mGy per year.

1. INTRODUCTION

The life and well-being of man depends on the environment and, consequently, protection of the environment is of primary concern in relation to all human activities resulting in accumulation and/or discharges of harmful substances. The recognition of this fact has led to the establishment of criteria for the feasibility of human activities resulting in radiation from, e.g., point sources or radionuclides dispersed in the environment. These criteria are, to a large extent, based on the current knowledge of radiation effects on humans.
The issue of protection of non-human beings from detrimental effects of ionizing radiation has been addressed on a number of occasions, resulting in the commonly quoted, although indirect, statement that organisms in the environment are adequately protected when humans are protected [1], i.e. at radiation levels implied by the radiation protection standards of the International Commission on Radiological Protection (ICRP). It is not the aim of this paper to challenge this view.

It is the opinion of the Swedish Radiation Protection Institute (SSI), however, that the issue of protecting the natural environment from detrimental effects of ionizing radiation should be addressed in its own right, using direct approaches, where man is merely regarded as one of several indicator species. There are several reasons for this position: (a) Given the aim of protecting populations of non-human species, a few admittedly rare situations must be discussed, e.g. that a few species may be more radiosensitive than man and that, in a few cases, populations may be small enough that loss of even relatively few individuals could seriously affect the gene pool and thus, at least indirectly, the survival of the entire population. (b) Ethically, it is not self-evident that protection of populations is sufficient for non-human species. At least for some higher species, radiation induced disease among individuals may become unacceptable. (c) The concept of protecting other species via man is mainly geared to regulated releases from nuclear activities. Interventions aimed at reducing risks to man may, however, completely fail to affect other species.

The Swedish radiation protection act, revised in 1988, identifies protection of the environment as one of its major goals. Furthermore, the Swedish nuclear industry is rapidly advancing plans for the construction of a geological repository for spent nuclear fuel, which necessitates establishment of criteria for assessment of the feasibility of these plans in the near future. This paper gives a brief discussion of the background and presents a framework for reasoning that could apply in establishing criteria for environmental protection. The discussion is relevant for protection against radiation emitted by radionuclides dispersed in the environment, but, in terms of general goals, it also has relevance for point sources of radiation.

2. TARGETS FOR PROTECTION

2.1. The environment

The environment of humans has both abiotic and biotic components. Protective actions would, however, normally be targeted at biotic components, not abiotic components such as water and rocks. Nevertheless, changes in radionuclide levels in abiotic components, caused by anthropogenic activity, can be a useful criterion in screening for possible effects on biotic components. The effect on abiotic components can be assessed by using the environmental increment as a criterion. This defines the increment of a specified radionuclide in relation to the natural variation,
or inferred natural variation, of its abundance and is used as an initial step in the assessment model for waste repositories developed by Atomic Energy of Canada Ltd [2, 3].

Further assessment can be made of doses (observed or predicted) to biota. The biotic components directly relevant to man are normally considered via assessments of the human exposure pathway. Organisms in the natural environment may or may not be directly included in this pathway; however, criteria for protection should apply equally for all. The Swedish Environmental Protection Agency defines four general targets for protection: (a) human health, (b) biological diversity, (c) the production potential of natural resources, and (d) the (agri)cultural landscape [4]. It would seem that points (a) and (d) are normally covered by actions taken to protect humans and should thus be reasonably covered by the dose limits to the public (mean individual annual effective dose of 0.1 mSv to members of the critical group for Swedish nuclear facilities). Points (b) and (c) are given more attention below.

2.2. Biodiversity

We propose that emphasis should be placed on biodiversity when assessing the impact of anthropogenic radionuclides on the natural environment. The term biodiversity (biological diversity) has been defined at the United Nations Conference on Environment and Development in Rio de Janeiro as “the variability among living organisms from all sources including, inter alia, terrestrial, marine and other aquatic ecosystems and the ecological complexes of which they are part; this includes diversity within species, between species and of ecosystems” [5]. In principle, this equals the definition adopted by the Swedish Environmental Protection Agency, and can be adopted also in SSI’s work on defining criteria for protection against detrimental effects of radiation. The fact that the ecosystem level is mentioned is interesting, since it implies that not only particular organisms but also the transfer of material, energy, etc., that constitutes the backbone of the ecosystem are considered important. This highlights the importance of all organisms, not only certain conspicuous or in other respects publicly ‘interesting’ species. It includes also races, cultivars, etc., of, e.g., livestock and crops. However, these will be considered in the exposure pathways to man and are not of direct relevance to the theme of this paper.

Protection of the environment to uphold biodiversity also implies, although not fully, that the biological resources are protected. The term biological resources has been defined at the United Nations Conference as the “genetic resources, organisms or parts thereof, populations, or any other biotic component of ecosystems with actual or potential use for humanity” [5]. Biological resources are intimately coupled to biodiversity in a number of ways; biodiversity contributes to genetic resources of potential use, and the production potential of the natural environment as well as its capacity to withstand or recover from natural stress conditions is usually considered to be correlated with biodiversity. Also, in diverse use of arable land, the concept
of biodiversity is partially applicable. In conclusion, it appears that biological diversity has general relevance in environmental protection, also in relation to environmental radioactivity.

The SSI is well aware, however, that biodiversity is not in all situations considered an ultimate goal of efforts directed at environmental protection. Biodiversity may decline owing to natural causes. Furthermore, there is no general human interest in protection of a limited number of organisms that are in some way (e.g. for health reasons or on economic grounds) particularly harmful to man. Finally, monitoring — or prediction — of biodiversity are non-practicable options when assessing the impact of environmental radioactivity. Measures directed at protection of the natural environment will rather have to ensure that the conditions for maintained biodiversity remain favourable and/or that environmental contamination does not cause changes in biodiversity. The most practicable option is, probably, assessment of doses to different types of biota occupying different media.

2.3. Target organizational level

Biological life can be arranged in a hierarchy according to the level of organization, ranging from the molecular level to ecosystems. Although distinctions are not straightforward in all cases, it appears justified to define a target organizational level for actions taken to protect the natural environment, so that, by inference, higher organizational levels should be automatically protected.

The traditional view [6] is that for non-humans, populations should be protected. However, populations may be small and isolated, or have restricted gene exchange with other populations of the same species. Under such circumstances, protection should be directed towards individuals.

There is little doubt that reproduction is the most critical part to be affected by radiation; this is related either to radiation induced loss of fertility, or distorted embryonic development, or genetic disease of the offspring. This may result in the requirement that protection of populations should have more stringent limitations to exposures than protection of individuals.

3. CRITERIA FOR PROTECTION

3.1. Needs for quantitative criteria

The general desire for criteria for environmental protection was briefly touched upon in the Introduction. The question is whether the criteria should remain general or whether they should be given in quantitative terms. Quantitative criteria, in terms of doses to organisms in the environment, are helpful in assessments of environmental impacts of nuclear installations, and may also aid in defining the area
around a nuclear power plant, waste repository, etc., that is likely to be significantly affected. Ideally, the area influenced by any installation should not exceed the physical boundaries of the installation itself, i.e. the damage to natural life in the long run should not exceed that caused by the construction per se. Harmful effects outside this area could, depending on the magnitude of the area and of the effects, be interpreted as signs of warning or indicate that the installation is unacceptable.

3.2. Scientific basis

Quantitative criteria are accepted elements when discussing protection of man and are supported by a large body of scientific information that enables calculation of risk factors for, e.g., cancer and genetic disease. The question is whether we are in possession of enough information for setting quantitative criteria for protection of nature.

It is evident that the radiation sensitivity of the vast majority of the probably 30–40 million non-human species that exist on Earth is by no means as well documented as that of man. However, it can be argued that the scientific evidence is solid enough to indicate a range of biological sensitivities among organisms and developmental stages that would be of relevance for setting criteria. Rather than considering these ranges as being the result of imprecision or insecurity of data, it can be argued that there are scientific grounds for the contention that ranges are not likely to become smaller with increased scientific information (although increased research is interesting in its own right). One could consider, for example, the electrical gradients across cell membranes that range from a few tens of millivolts (inside negative) in, e.g., amphibian oocytes, to hundreds of millivolts in meristematic tissues of aquatic plants. For a radionuclide occurring in the environment as a divalent cation, the equilibrium distribution ratio between inside and outside would vary by several orders of magnitude over this range of electrical gradients, which subsequently bears on tissue distribution, internal doses, etc. Bearing in mind further variations related to species differences, developmental stage, synergistic stress effects, etc., there will always be a limitation in the precision of predictions of consequences to the natural environment. An updated compilation of radiosensitivity data is expected from UNSCEAR during 1995.

3.4. Ranges of acceptable doses

Ionizing radiation is likely to more severely affect organisms that are complex (large number of genes) and long lived. Generally, this applies to organisms such as vertebrates and plants. Although the variation between organisms and developmental stages is enormous, data suggest that doses lower than 1–5 Gy\(^1\) per year are

\(^1\) 1 Gy = 1 J/kg.
less likely to cause severe detrimental effects on individuals of these kinds of organisms [6]. A considerable safety factor should be applied. It is proposed that a safety factor of between 100 and 1000 should be used to account for the facts that experimental studies usually do not consider the possible effects of long term, low level radiation (consequently, risk estimates are scarce), and that genetic effects and effects on development and behaviour actually have been recorded at substantially lower radiation levels [6], and to account for the aforementioned lack of precise information (cf. the precautionary principle). This reasoning would lead to the conclusion that the quantitative criterion for protection of non-human species should be set at less than 50 mGy per year.

It is interesting to note that the maximum doses that terrestrial and aquatic biota are likely to receive at the current ICRP standard of a maximum dose of 1 mSv annually to members of the critical group are most likely less than 300–400 mGy [6]. The Swedish limit for exposure of the public is 0.1 mSv per year (mean individual annual effective dose to members of the critical group), which would reduce the doses to organisms in the environment by a factor of ten. The values thus arrived at are within the range defined according to the above reasoning. Although further analysis is required, this indicates that the standard set for man and discussed for organisms in the natural environment in this paper would at least be compatible. Still, the SSI considers it desirable that operators of nuclear installations provide direct arguments indicating that organisms in the natural environment are protected.

REFERENCES


INTERCEPTION AND POST-DEPOSITION RETENTION OF RADIONUCLIDES BY VEGETATION AND THEIR IMPORTANCE FOR DOSE ASSESSMENT

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Abstract

INTERCEPTION AND POST-DEPOSITION RETENTION OF RADIONUCLIDES BY VEGETATION AND THEIR IMPORTANCE FOR DOSE ASSESSMENT.

The interception of radionuclides represents the link between the atmospheric dispersion of radionuclides released to the atmosphere and their transport in food-chains. The paper gives an overview of these processes and their dependence on the characteristics of the deposit and the environmental conditions. For dry deposits, the important factors that control the interception are the particle size and the development of the vegetation. The interception fraction increases if the plant surfaces are wet or moist. The interception of wet deposited activity by vegetation depends on the chemical form of the radionuclide, the amount of rainfall during which the deposition occurs and the development of the vegetation. The loss of radionuclides from plants after deposition is due to removal by wind, rain, fog, abrasion of waxy particles from the leaves and loss of leaves. Regarding the decrease of the activity concentration of the plant, the diluting effect of the increase of biomass is very important. Further influencing factors are the particle size and the age of the plant. In some experiments the loss rate decreased with time after deposition. The processes of interception of dry and wet deposits, and the post-deposition retention are described and discussed. Some exemplary model calculations are given that underline the importance of the processes described for prospective and retrospective dose assessment.

1. INTRODUCTION

The interception of radionuclides represents the link between the atmospheric dispersion of radionuclides released to the atmosphere and their transport in food-chains. Especially the data available since the Chernobyl accident have indicated that radionuclide transfer from air and rain to food and feed crops is an important source of uncertainty in the estimation of the exposure due to ingestion of contaminated
In the first few months after radionuclide depositions during the growing season, the activity concentrations in foodstuffs will be dominated by initial and post-deposition retention of the deposited material. An extensive review of initial and post-deposition retention has been initiated in the framework of the VAMP programme [1]. The present paper summarizes the main findings of that review and gives a brief description of these processes and their dependences on environmental factors, the chemical form and seasonal factors.

2. INTERCEPTION OF DEPOSITED RADIONUCLIDES

When materials are deposited from the atmosphere, whether by dry or wet deposition processes, a certain fraction of the materials will be intercepted by vegetation, with the remainder reaching the ground. The interception fraction $f$, which is defined as the fraction of the deposited material that is initially retained, is often quantified as a function of the standing biomass $B$ (kg/m$^2$, dry mass) and the empirically derived absorption coefficient $\mu$ (m$^2$/kg) (see Ref. [2])

$$f = 1 - \exp(-\mu B)$$

(1)

In order to take into account the dependence of the interception fraction on the biomass, $f$ can be normalized to $B$ in order to obtain the mass interception factor $f/B$ (m$^2$/kg) according to:

$$
\frac{f}{B} = \frac{1 - \exp(-\mu B)}{B}
$$

(2)

which facilitates comparison of experiments performed with varying biomass. For small values of $B$, there is little difference between $f/B$ and $\mu$. In this paper, if not otherwise stated, $f/B$ is used to quantify the initial retention. However, the validity of Eqs (1) and (2) is limited to those parts of the growing periods during which the exposed leaf area increases in proportion to the standing biomass. This is only the case in the first part of the growing period; in the second part the biomass still increases, whereas the leaf area begins to decrease [3]. Furthermore, in all estimations of $f$ or $f/B$, it should be carefully checked whether the deposition is dry or wet, since the deposition mode has — as will be shown later — a large influence on the interception fraction. Correct application of $f$ and $f/B$ is therefore only warranted if the deposition scenario under which $f$ or $f/B$ has been derived is similar to the situation for which these parameters will be applied.

2.1. Interception of dry deposits

A number of experiments have been performed to determine the interception of dry deposits on vegetation. In these experiments, the absorption coefficient $\mu$ was
derived from the total deposition, the initially retained fraction and the biomass. The results of such experiments are summarized in Table I. It is interesting to note that the absorption coefficient is similar for small particles and elemental iodine. With increasing particle size, $\mu$ decreases considerably. This is probably due to the fact that larger particles roll off from the plant surface more easily than smaller ones. However, this is obviously not true for Lycodium spores which seem to adhere more strongly to the plant surface than sand particles. In the experiments where the vegetation was moist or wet, the observed absorption coefficients were considerably higher, which is probably due to the enhanced sticking effect. This is confirmed by the observations of Pinder et al. [9], who found lower interception fractions on plants with waxy leaf surfaces. However, with the exception of the near-field of the release point, for the purpose of dose assessment, particles with a diameter of up to a few micrometres are most relevant, since the depletion of large particles from the radioactive cloud is very effective.

### Table 1. Summary of Absorption Coefficients $\mu$ Obtained from Field Experiments for Dry Deposits

<table>
<thead>
<tr>
<th>Deposited material</th>
<th>Diameter ($\mu$m)</th>
<th>Crop</th>
<th>Absorption coefficient (m$^2$/kg)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Mean</td>
<td>Std. dev.</td>
</tr>
<tr>
<td>Lycodium spores</td>
<td>32</td>
<td>Grass</td>
<td>3.1</td>
<td>0.15</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Wheat, dry</td>
<td>3.2</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Wheat, moist</td>
<td>9.6</td>
<td>3.7</td>
</tr>
<tr>
<td>Quartz</td>
<td>44–88</td>
<td>Grass</td>
<td>2.7</td>
<td>0.3</td>
</tr>
<tr>
<td>Sand</td>
<td>40–63</td>
<td>Grass, dry</td>
<td>0.44</td>
<td>0.15</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Grass, wet</td>
<td>0.88</td>
<td>0.13</td>
</tr>
<tr>
<td></td>
<td>63–100</td>
<td>Grass, dry</td>
<td>0.23</td>
<td>0.07</td>
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<tr>
<td></td>
<td></td>
<td>Grass, wet</td>
<td>0.69</td>
<td>0.16</td>
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<td></td>
<td>100–200</td>
<td>Grass, dry</td>
<td>0.24</td>
<td>0.07</td>
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<tr>
<td></td>
<td></td>
<td>Grass, wet</td>
<td>0.46</td>
<td>0.11</td>
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<tr>
<td>Pu-238 particles</td>
<td>$\approx 1$</td>
<td>Corn</td>
<td>3.6</td>
<td>0.05</td>
</tr>
<tr>
<td>I-131</td>
<td>Vapour</td>
<td>Grass</td>
<td>2.8</td>
<td>0.14</td>
</tr>
<tr>
<td>Pb-212</td>
<td>Vapour</td>
<td>Artificial grass</td>
<td>13</td>
<td>—</td>
</tr>
</tbody>
</table>
TABLE II. MASS INTERCEPTION FACTORS f/B DETERMINED DURING THE CHERNOBYL FALLOUT AT NEUHERBERG, GERMANY [11]

<table>
<thead>
<tr>
<th>Date</th>
<th>Ru-106</th>
<th>I-131</th>
<th>Cs-137</th>
<th>Ba-140</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 May 1986</td>
<td>0.48</td>
<td>0.70</td>
<td>1.1</td>
<td>1.7</td>
</tr>
<tr>
<td>2 May 1986</td>
<td>0.95</td>
<td>1.0</td>
<td>1.3</td>
<td>1.8</td>
</tr>
<tr>
<td>3 May 1986</td>
<td>0.95</td>
<td>0.9</td>
<td>1.3</td>
<td>1.4</td>
</tr>
<tr>
<td>4 May 1986</td>
<td>1.0</td>
<td>0.9</td>
<td>1.0</td>
<td>1.4</td>
</tr>
<tr>
<td>5 May 1986</td>
<td>0.55</td>
<td>0.75</td>
<td>0.90</td>
<td>1.4</td>
</tr>
<tr>
<td>6 May 1986</td>
<td>0.55</td>
<td>0.75</td>
<td>0.85</td>
<td>1.2</td>
</tr>
</tbody>
</table>

2.2. Interception of radionuclides deposited during rainfalls

The interception of wet deposited radionuclides by plants is dependent on a variety of factors, the most important of which are:

- physico-chemical properties of the element,
- development of the plant at the time when deposition occurs,
- the amount of rainfall during which the activity is deposited.

The differences in the interception among different elements is due to their different valencies. The plant surfaces are negatively charged; they have properties of a cation exchanger [10]. Therefore, the initial retention of anions such as iodide is less than that of polyvalent cations, which seem to be very effectively retained on the plant surface. In Table II, the mass interception factor, as measured in Neuherberg (Germany), corrected for radioactive decay, is given for the period of 1–6 May 1986, for $^{106}$Ru, $^{131}$I, $^{137}$Cs and $^{140}$Ba [11]. Since most of the deposition occurred during a heavy shower on 30 April and 1 May, dry deposition of $^{106}$Ru, $^{137}$Cs and $^{140}$Ba did not contribute significantly to the total deposition [12]. The mass interception factors increase in the order of $^{106}$Ru, $^{131}$I, $^{137}$Cs and $^{140}$Ba, although these nuclides were deposited during the same rainfall event. The highest values were found for $^{140}$Ba, which behaves similarly to strontium. Barium is a bivalent cation, which seems to be retained stronger on the negatively charged plant surface than the monovalent caesium cation. The mass interception factor is smaller for iodine, which probably occurs as monovalent anion on the plant surface.
The mass interception factors for $^{106}$Ru are slightly lower than those for $^{131}$I. Accounting for the contribution of dry deposition due to the presence of elemental iodine, this may indicate that ruthenium behaves similarly to iodine concerning the interception of wet deposits. However, since the information about the chemical form of ruthenium is very poor and speculative, it cannot be decided whether this observation is typical for ruthenium or specific for the Chernobyl accident. In the days after 1 May, the differences in the mass interception factors were less than those on the first day owing to the increasing contribution of dry deposition in that period with only small amounts of precipitation.

These observations agree very well with those reported by Hoffman et al. [13], who measured the mass interception factors for a variety of anions and cations and for insoluble microspheres with a diameter of 9 $\mu$m on the leaves of several tree species (Fig. 1) during a simulated shower of 8.5 mm. The insoluble microspheres are also positively charged. The mass interception factors for cations are approximately a factor of three to five higher than those for anions such as iodide or...

**FIG. 1.** Mean values of mass interception factors for five different plant types, for radioactively labelled cations and anions, and for one size class of insoluble microsphere, under controlled laboratory conditions, during an artificial rainfall of 8.5 mm [13].
sulphate. Compared to the valency, the plant species seems to be of minor importance. It is obvious that the valency of the element considered is the controlling condition for the mass interception factor.

The effect of the amount of rainfall on the mass interception factors is obvious from Fig. 2, where the mass interception factors for $^7\text{Be}$, $^{131}\text{I}$ and insoluble microspheres, obtained in the experiments discussed in Ref. [14], are plotted against the amount of precipitation during which the activity was deposited. The difference due to the different charges of the tracers used is obvious; furthermore, Fig. 2 demonstrates that the mass interception factor is inversely proportional to the amount of rainfall. However, this effect is more pronounced for anions than for cations. The interception of iodine decreases in proportion to increasing precipitation; this relationship is also obvious for cations, but is much less pronounced. Especially for intensive precipitation the mass interception factor declines much faster for anions. According to Ref. [15], the interception fraction $f$ for iodine can be described very well by an approach which assumes that interception depends on the thickness of the water film that remains on the leaves subsequent to a precipitation event. It has been
observed that cations are retained more strongly than would be expected from the retention of the water film.

In Fig. 3, the interception fraction $f$ for $^{137}$Cs, determined for various species of cereals, is plotted against the leaf area index at the time of application [16]. The leaf area index gives the ratio of the leaf area of a canopy per unit soil area. The activity was applied at different stages of development during artificial rainfalls of 1 mm. The interception fraction increases in proportion to the leaf area. This correlation is very useful for estimating the initial retention on crops such as vegetables, grain and fruit because for these plants, in the second half of their growing period, the biomass is still increasing whereas the leaf area remains constant or is already declining.

3. POST-DEPOSITION RETENTION OF RADIONUCLIDES

After radionuclides are deposited on vegetation, their activity will decrease because of radioactive decay and environmental removal processes due to the influence of wind and water, surface abrasion and growth dilution. The exact
contribution of each of these processes is difficult to quantify, since they occur simultaneously and their effect can hardly be distinguished. The post-deposition retention is quantified by the weathering half-life $T_w$, which represents the period of time during which one-half of the activity deposited on plants is lost. There are two possibilities to express the weathering half-life: The first definition refers to the decrease of the activity concentration of the vegetation; this definition includes the contribution of growth dilution. The other definition excludes growth dilution, and in this case the weathering half-life refers to the activity which is lost from the vegetation per unit ground area. The total loss from vegetation including radioactive decay is estimated from the weathering half-life $T_w$ and the physical half-life $T_r$ according to:

$$T_{eff} = \frac{T_w T_r}{T_w + T_r}$$

(3)

Values for $T_w$ are compiled in Table III on the basis of unit mass and unit ground area. This compilation demonstrates that the pre-Chernobyl observations agree well with the post-Chernobyl observations. The weathering half-life seems to be slightly lower for iodine than for caesium. Some authors indicate that $T_w$ increases with time after deposition. For dormant and senescent vegetation, the weathering half-lives observed are much longer, since the metabolic activity of such plants is much lower than that of growing plants. The stage of development also has an influence on the post-deposition retention. The weathering half-lives increase during the maturing process, when the foliage becomes more and more similar to dormant or dead standing plant canopies.

In some experiments, the weathering half-life has been determined on the basis of unit mass and unit ground area. This shows that growth dilution may be a very effective process for reduction of the activity concentration in contaminated plants. However, this process is very much dependent on the season; it may be very effective at the beginning of the growing period in spring, but much less important in autumn when the growth rates generally decline.

4. IMPORTANCE OF INTERCEPTION FOR DOSE ASSESSMENT

Interception and post-deposition retention are key parameters for dose assessment. In order to demonstrate the impact of the element dependence of the interception of wet deposited radionuclides (Fig. 4), the contamination of grass is calculated with the model ECOSYS-87 [31] for a normalized deposition of 10 kBq/m$^2$ for $^{131}$I, $^{137}$Cs and $^{90}$Sr during a rainfall of 5 mm on 1 June, when the grass is fully developed. This approach considers the element dependence of interception. Although the depositions are the same for all three nuclides, the initial values of contamination differ by a factor of four.
TABLE III. SUMMARY OF WEATHERING HALF-LIVES Tₜ

<table>
<thead>
<tr>
<th>Experimental conditions</th>
<th>Weathering half-life (d)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>mean (range)</td>
<td>Per unit mass</td>
</tr>
<tr>
<td>I₂ vapour on herbaceous vegetation</td>
<td>6.8 (4.8-7.9)</td>
<td>7.2 (4.5-14)</td>
</tr>
<tr>
<td>I particles on herbaceous vegetation</td>
<td>8.2 (2.8-16)</td>
<td>8.8 (2.8-16)</td>
</tr>
<tr>
<td>Particles on herbaceous vegetation</td>
<td>12 (9-19)</td>
<td>17 (9-34)</td>
</tr>
<tr>
<td>Particles on woody vegetation</td>
<td>20 (12-28)</td>
<td>20 (12-28)</td>
</tr>
<tr>
<td>Fine spray of Sr-89 on pasture</td>
<td>9</td>
<td>14</td>
</tr>
<tr>
<td>Spray of radionuclides on barley</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Week 2-11 after sowing</td>
<td></td>
<td>18 (14-22)</td>
</tr>
<tr>
<td>Week 1-16 after sowing</td>
<td></td>
<td>46 (32-54)</td>
</tr>
<tr>
<td>Be-7 on vegetation</td>
<td></td>
<td>17</td>
</tr>
<tr>
<td>Be-7 on dormant vegetation</td>
<td></td>
<td>130</td>
</tr>
<tr>
<td>Pu-238 on corn</td>
<td></td>
<td>12</td>
</tr>
<tr>
<td>I-131, Be-7 and particles on grass</td>
<td>7.2-15</td>
<td>7.5-18</td>
</tr>
<tr>
<td>Cs-137 on lettuce</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>I-131 on pasture</td>
<td>7.5-8.9</td>
<td></td>
</tr>
<tr>
<td>I-131 on pasture</td>
<td>9.8</td>
<td></td>
</tr>
<tr>
<td>Cs-137 on pasture</td>
<td>14</td>
<td></td>
</tr>
<tr>
<td>Cs-137 on pasture</td>
<td>8-14 (95%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>40-60 (5%)</td>
<td></td>
</tr>
<tr>
<td>I-131 on pasture</td>
<td>12</td>
<td></td>
</tr>
<tr>
<td>Cs-137 on pasture</td>
<td>14</td>
<td></td>
</tr>
<tr>
<td>I-131 on pasture</td>
<td>5.9-14</td>
<td></td>
</tr>
<tr>
<td>Cs-137 on pasture</td>
<td>11-15</td>
<td></td>
</tr>
<tr>
<td>I-131 on pasture</td>
<td>11</td>
<td></td>
</tr>
<tr>
<td>Cs-137 on pasture</td>
<td>11</td>
<td></td>
</tr>
<tr>
<td>I-131 on pasture</td>
<td>9.6</td>
<td></td>
</tr>
<tr>
<td>Cs-137 on pasture</td>
<td>14</td>
<td></td>
</tr>
<tr>
<td>Cs-137 on pasture</td>
<td>9-14 (92%)</td>
<td></td>
</tr>
<tr>
<td>Cs-137 on pasture</td>
<td>49-52 (8%)</td>
<td></td>
</tr>
<tr>
<td>Cs-137 on pasture</td>
<td>10-11</td>
<td></td>
</tr>
<tr>
<td>I-131 on pasture</td>
<td>9.1 ± 0.6&lt;sup&gt;a&lt;/sup&gt;</td>
<td></td>
</tr>
<tr>
<td>Cs-137 on pasture</td>
<td>11 ± 0.8&lt;sup&gt;a&lt;/sup&gt;</td>
<td></td>
</tr>
</tbody>
</table>

<sup>a</sup> Standard error of the mean.
FIG. 4. Activity concentration in pasture grass as a function of time after deposition of 10 kBq $^{131}$I, $^{137}$Cs and $^{90}$Sr per square metre during a rainfall of 5 mm on 1 June, estimated according to Ref. [32].

In Fig. 5, the thyroid doses to adults after a normalized deposition of 10 kBq $^{131}$I/m$^2$ on 1 June are compared for different deposition modes. In the first case, only dry deposition is assumed, which leads to a total deposition of 10 kBq/m$^2$. In the second case, only wet deposition is assumed during a light shower of 1 mm. For this scenario, the thyroid dose is about a factor of two lower than that for dry deposition. If deposition occurs during a heavy shower of 10 mm, as in the third case, the thyroid dose is an additional factor of eight lower than in the case of a shower of 1 mm.

These results also underline the importance of initial retention of radionuclides for the reconstruction of ingestion doses for past contamination events. In such cases, the data sets are often incomplete, and the only reliable data available are the total deposition on the ground. However, if the deposition mode is not known or is very uncertain, the uncertainties associated with the resulting food activities and doses are considerable.

The same is true when data about initial contamination of vegetation are used to estimate inhalation doses. Going backwards from activity in vegetation to air concentration, modelling of deposition is the controlling factor for the accuracy of the resulting dose.
5. CONCLUSIONS

From the review of the processes quantifying initial and post-deposition retention of radionuclides, the following conclusions can be drawn:

— The approach of Chamberlain [2] is only appropriate for describing the initial retention of radionuclides by plants in the beginning of the growing period, when there is a correlation between biomass and leaf area. At later stages, when the biomass is increasing whereas the leaf area is already decreasing, this approach cannot be applied successfully without modification.

— Assuming a situation where the approach of Chamberlain is applicable, for dry deposits of small particles, a mass interception factor of $3 \text{ m}^2/\text{kg}$ might be a typical value. For particle sizes above 40 $\mu$m, the mass interception factors are considerably lower. The mass interception factors are significantly higher on wet plant surfaces than on dry surfaces.

— The initial retention of wet deposits depends on the element. The retention of cations is much higher than that of anions. Furthermore, the initial retention decreases with increasing amounts of rainfall. This effect is more pronounced for anions.
— Radionuclides deposited on plants are lost owing to removal by wind, rain, fog and surface abrassion.
— Growth of vegetation has a diluting effect on retention, measured in terms of activity per unit mass. This effect is most pronounced for growing herbaceous vegetation, is less obvious for woody plants and is negligible for dormant vegetation.
— The weathering half-lives observed during the Chernobyl fallout are in the same range as the values of pre-Chernobyl fallout. Typical values for the weathering half-lives, including growth dilution, are in the range of 10–20 days; for iodine the values are slightly lower.

REFERENCES


RADIONUCLIDE TRANSFER
IN THE FRESHWATER ENVIRONMENT

(Session 5)

Chairman

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ESTIMATION OF ECOLOGICAL HALF-LIVES OF CAESIUM-137 IN LAKES CONTAMINATED BY CHERNOBYL FALLOUT

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Abstract

ESTIMATION OF ECOLOGICAL HALF-LIVES OF CAESIUM-137 IN LAKES CONTAMINATED BY CHERNOBYL FALLOUT.

The concept of ecological half-life describes the decrease in radionuclide concentration in natural ecosystems resulting from all processes, including a wide range of physical, chemical and biological processes as well as physical decay. Knowledge of ecological half-lives is important from a radiation protection point of view, since such values can be used for a rapid estimate of recovery times in contaminated areas. Within the scope of the IAEA co-ordinated research programme Validation of Environmental Model Predictions (VAMP), sets of empirical data from seven European lakes contaminated by Chernobyl fallout have been
used to determine the ecological half-lives of radiocaesium for water and different fish species by curve fitting of observed data. For radiocaesium in lake waters, two distinct ecological half-lives were observed: (1) an initial short half-life of a few months due to transfer to the sediments, as well as a short water retention period during spring in several lakes; and (2) the subsequent ecological half-life of radiocaesium in lake waters, which was longer and was related both to the lakes' water retention period and to the characteristics of the secondary load from the catchment. The ecological half-life in fish varied with both species and lake, ranging from 1 year for small perch from IJsselmeer in the Netherlands to 4.6 years for pike from Iso Valkjärvi in Finland. The influence of temperature on the biological half-life in fish was reflected in the values of ecological half-lives, with more southern, warmer lakes showing a faster rate of recovery. Ecological half-lives are not constant but vary with both lake characteristics and fish species. However, the VAMP studies have shown that simple predictive models can account for a wide range of different processes by utilizing the concept of ecological half-life.

1. INTRODUCTION

The concept of ecological half-life describes the decrease in radionuclide (such as $^{137}$Cs) concentration in natural ecosystems resulting from all processes. It includes a wide range of physical, chemical and biological processes in addition to physical decay. Knowledge of ecological half-lives is important from a radiation protection point of view, since such values can be used for a rapid estimate of recovery times in contaminated areas. Ecological half-lives are also important parameters in simple predictive models for assessing the long term dose to man as well as the effectiveness of remedial measures.

2. METHODS AND DATA SOURCES

Within the scope of the IAEA/CEC co-ordinated research programme Validation of Environmental Model Predictions (VAMP), sets of empirical data from seven European lakes contaminated by Chernobyl fallout have been used to test and improve models for the prediction of radionuclide concentrations in lake waters and fish populations [1]. For the lakes with a good database, ecological half-lives for water and different fish species have been determined by curve fitting of these observational data. The sensitivity of values of ecological half-life to the time period considered has also been evaluated.
TABLE I. OBSERVED VALUES FOR CHERNOBYL $^{137}$Cs IN EUROPEAN LAKE WATERS$^a$

<table>
<thead>
<tr>
<th>Lake</th>
<th>Observed peak (Bq/L)</th>
<th>Date of peak</th>
<th>Half-life, &lt;1 year</th>
<th>Half-life, &gt;1 year</th>
</tr>
</thead>
<tbody>
<tr>
<td>Esthwaite Water, United Kingdom</td>
<td>0.3</td>
<td>1986-5-13</td>
<td>0.08</td>
<td>0.2</td>
</tr>
<tr>
<td>Devoke Water, United Kingdom</td>
<td>0.35</td>
<td>July 1986</td>
<td>0.4</td>
<td>2</td>
</tr>
<tr>
<td>Hillesjön, Sweden</td>
<td>6.4</td>
<td>1986-6-12</td>
<td>0.3</td>
<td>1.5</td>
</tr>
<tr>
<td>IJsselmeer, Netherlands</td>
<td>0.2</td>
<td>1986-5-1</td>
<td>0.75</td>
<td>1.7-2.0</td>
</tr>
<tr>
<td>Iso Valkjärvi, Finland</td>
<td>4.6</td>
<td>1987-6-10</td>
<td>—</td>
<td>2.6</td>
</tr>
</tbody>
</table>

$^a$ Peak values, date of observation and calculated ecological half-lives (in years) for the first year after contamination and for subsequent years.

3. RESULTS AND DISCUSSION

Although the database covered a wide range of lake types, they all showed two distinct phases in the decline of radiocaesium in water. During the first months after deposition of fallout from Chernobyl there was a rapid rate of removal of radiocaesium from lake waters (Table I, Figs 1 and 2). This rapid decline was probably due to effective removal to sediments, as the water retention periods in most lakes are too long to explain such a rapid decline. From about two months onwards, however, the fall in the concentration of radiocaesium in water in most lakes was of the order of two years; however, in Esthwaite Water it was much shorter. The short half-life in Esthwaite Water may be, at least in part, explained by its high productivity and high sedimentation rate compared with that of the other lakes. The fit to an exponential decrease for water concentrations was good for most lakes, but for Hillesjön the correlation coefficient was low (Fig. 2). This was probably due to resuspension of caesium rich sediments in this shallow, wind exposed lake [2]. A study of large Finnish drainage basins contaminated by Chernobyl fallout [3] showed a similar pattern of decline in radiocaesium concentrations in water, with two components, a short term one of about two months and a long term one of about two years. However, the decline was much slower in drainage basins with a high percentage of bogs. This can also partly explain the much longer half-lives of radio-nuclides in Devoke Water compared to Esthwaite Water [4].
FIG. 1. Concentration of $^{137}$Cs in the waters of Esthwaite Water in the Lake District (United Kingdom) after the Chernobyl accident in 1986. The best fit to the observed data is given by two regression equations, with a short term rapid decrease followed by a long term slower decline.

FIG. 2. Concentration of $^{137}$Cs in the waters of Hillesjön (Sweden) after the Chernobyl accident in 1986. The best fit to the observed data is given by two regression equations, with a short term rapid decrease followed by a long term slower decline.
TABLE II. CALCULATED MEAN ECOLOGICAL HALF-LIVES FOR CHERNOBYL $^{137}$Cs IN DIFFERENT FISH SPECIES IN CONTAMINATED EUROPEAN LAKES

<table>
<thead>
<tr>
<th>Lake</th>
<th>Fish species</th>
<th>Ecological half-life (years)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Devoke Water, United Kingdom</td>
<td>Brown trout, <em>Salmo trutta</em></td>
<td>1.6</td>
</tr>
<tr>
<td>Øvre Heimdalsvatn, Norway</td>
<td>Brown trout, <em>Salmo trutta</em></td>
<td>2.9</td>
</tr>
<tr>
<td>Hillesjön, Sweden</td>
<td>Roach, <em>Rutilus rutilus</em></td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>Perch, <em>Perca fluviatilis</em> (mixed)</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>Perch, <em>Perca fluviatilis</em> (&lt;20 cm)</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>Pike, <em>Esox lucius</em></td>
<td>2.4</td>
</tr>
<tr>
<td>IJsselmeer, Netherlands</td>
<td>Perch, <em>Perca fluviatilis</em> (10-20 cm)</td>
<td>1</td>
</tr>
<tr>
<td>Iso Valkjärvi, Finland</td>
<td>Pike, <em>Esox lucius</em></td>
<td>4.6</td>
</tr>
</tbody>
</table>

The fish compartment is much more complicated. Although many models only consider uptake from water, this is only one of the pathways for radiocaesium, and many studies (e.g. Refs [5-7]) have demonstrated the importance of uptake via food organisms. Mean ecological half-lives have been calculated for different fish species from selected lakes studied in the VAMP programme (Table II). The results for brown trout, *Salmo trutta*, from Heimdalsvatn were very consistent and showed an ecological half-life of Chernobyl $^{137}$Cs of 3 years (Fig. 3). In contrast, $^{137}$Cs in trout from Devoke Water had an ecological half-life of only 1.6 years, although the empirical data are rather scattered. Shorter half-lives of $^{137}$Cs were found for perch, *Perca fluviatilis*, in IJsselmeer compared with the same species in the Nordic lakes (Fig. 4, Table II). This is a reflection of the influence of temperature on the biological half-life in fish, with the more southern, warmer lakes showing a faster rate of recovery [8, 9]. Intensive fishing may also reduce the ecological half-life of radiocaesium in fish.

The extent and nature of the secondary load will also influence the decline in radiocaesium concentrations, both in lake waters and in fish, and uncertainty analyses have shown the importance of secondary inputs from lake catchments, especially in the long term [1, 10, 11]. Factors influencing the nature and extent of the secondary load include the water retention period, the catchment area in relation to the lake area, and the percentage of bogs within the catchment [1, 4, 12].
FIG. 3. Decline in $^{137}$Cs in brown trout from lake Øvre Heimdalsvatn (Norway) after contamination by Chernobyl fallout in 1986.

FIG. 4. Decline in $^{137}$Cs in small perch from IJsselmeer (Netherlands) after contamination by Chernobyl fallout in 1986.
4. CONCLUSIONS AND RECOMMENDATIONS

Simple predictive models can utilize the concept of ecological half-lives, thus accounting for a wide range of different processes. However, it has to be borne in mind that, in contrast to physical half-lives, ecological half-lives are not constant but vary with both lake characteristics and fish species. They may also be time dependent, as observed for radiocaesium concentrations in lake waters. Nevertheless, the VAMP studies have clearly shown that it is possible to develop simple models which take into account such differences between lake systems [1, 12].

ACKNOWLEDGEMENTS

We are grateful to the IAEA/CEC for their support and encouragement throughout this study. Several persons and organizations supplied empirical data for our studies. In particular, we wish to thank R. Saxén, STUK, Finland, and D. Woodhead, MAFF, United Kingdom, for making data available.

REFERENCES


IN SITU DETERMINATION OF THE DISTRIBUTION OF $^{110}$Ag, $^{58}$Co, $^{60}$Co AND $^{54}$Mn BETWEEN FRESH WATER AND SUSPENDED MATTER

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Abstract

IN SITU DETERMINATION OF THE DISTRIBUTION OF $^{110}$Ag, $^{58}$Co, $^{60}$Co AND $^{54}$Mn BETWEEN FRESH WATER AND SUSPENDED MATTER.

In order to obtain data on the equilibrium conditions and the exchange kinetics of radionuclides between water and suspended matter in natural conditions, an in situ experiment was carried out in the nuclear power plant of Golfech (France). In this plant, an experimental physical model of river systems, represented by a channel, was built. Before being discharged into the Garonne river, part of the radioactive effluents, previously mixed with water originating from the cooling system, are diverted to a channel where different phenomena generally occurring in a river can be simulated. The experiment presented in the paper was undertaken during a scheduled discharge of low activity wastes; the suspended matter was sampled at four points along the channel using sequential filtration. The results give information about:

1. The influence of the particle size on the process of radionuclide adsorption by suspended matter. The uptake of radioactivity is generally higher for the smallest particles (size $< 2.5 \mu m$ for $^{110}$Ag, size $< 10 \mu m$ for $^{58}$Co, $^{60}$Co and $^{54}$Mn).
2. The exchange kinetics between water and suspended matter. Generally, radioactive equilibrium is rapidly attained. Consequently, it is possible to calculate the global distribution coefficient ($K_d$) value of each radionuclide ($^{110}$Ag, $^{58}$Co, $^{60}$Co and $^{54}$Mn) and to determine exactly the specific $K_d$ value for each granulometric class (class 1 $> 10 \mu m$, 10 $\mu m$ < class 2 $< 2.5 \mu m$; 2.5 $\mu m$ < class 3 $< 0.5 \mu m$).

1. INTRODUCTION

It is well known that the migration of many radionuclides in freshwater streams is strongly affected by the interaction of these radionuclides with suspended solids. Partitioning of radionuclides between water and suspended matter is usually described in terms of distribution coefficients ($K_d$), expressed as the concentration...
ratio between the dissolved phase and the particulate phase. The $K_d$ values are generally obtained by two major methodological approaches:

(a) *In situ experiments.* These experiments are based on field measurements (or estimations) of the radioactivity in the dissolved and particulate phases; they are generally undertaken downstream of nuclear power plants during scheduled releases of low activity wastes [1-5]. The measurements are used to calculate the ratio between sorbed and soluble nuclide fractions. However, like other thermodynamic constants, the $K_d$ values should be characteristic of equilibrium systems. Because of metrological limitations, it is often impossible to verify if equilibrium is actually reached under in situ experimental conditions; it is therefore difficult to know if the calculated ratios can really be considered as $K_d$ values [6].

(b) *In vitro experiments.* These experiments are undertaken in laboratories under controlled conditions; high level radioactivity is added to samples of river water and $K_d$ values are calculated using measurements made under equilibrium conditions [7]. However, speciation of the radionuclides added to the medium can be quite different from that of radionuclides released by nuclear power plants. Therefore, it is difficult to know if laboratory $K_d$ values can be extrapolated to in situ conditions [4].

In order to obtain more detailed and realistic data on the equilibrium conditions and the exchange kinetics between water and suspended matter in natural conditions, an in situ experiment was carried out in the nuclear power plant of Golfech (France). In this plant, an experimental physical model of river systems, represented by a channel, was built; before being discharged into the Garonne river, part of the effluents produced by the nuclear installation are diverted to a channel where different phenomena generally occurring in a river can be simulated. This paper presents the principal points of an experiment performed in July 1994 and part of the results obtained.

2. DESCRIPTION OF THE PHYSICAL MODEL

Routine operation of the nuclear power plant of Golfech produces liquid radioactive effluents which are treated and finally released into the Garonne river [8]. Apart from $^3$H, the main radionuclides identified in liquid wastes are generally $^{58}$Co, $^{110}$Ag, $^{60}$Co and $^{54}$Mn. Before being released into the river, the radioactive effluents are pre-diluted and mixed with the water originating from the cooling system; part of this mixture flows through a channel, the geometry of which is shown in Fig. 1.
A radioactive waste tank with a flow rate of 16 m$^3$/h and a volume of 680 m$^3$ is connected to an experimental physical model of the Garonne river. The effluent input has a flow rate of 3 m$^3$/h. Sampling points (1), (2), (3), and (4) are indicated on the diagram. The pH of the effluent is 8.3.

FIG. 1. Description of the physical model.
3. EXPERIMENTAL PROCEDURE

3.1. Location of the sampling points

Four sampling points have been chosen along the channel (Fig. 1). As the channel is supposed to be a plug-flow reactor, the contact time between the radionuclides, coming from the waste tank, and the suspended solids, originating from the cooling system, is different for each selected point; according to the characteristics of the experimental installation, the contact time is 1.5, 6, 9 and 20 hours, respectively.

3.2. Sampling of suspended material for gamma spectrometry

Sampling of large quantities of suspended matter is performed by means of a sequential filtration equipment using Nucleopore cartridges; for each sample, the particulate phase is separated by three successive filters, the porosity of which is 10, 2.5 and 0.5 \( \mu m \), respectively. After sampling, the filters are dried and mineralized at 550°C; the ash samples thus obtained are analysed by gamma spectrometry. (Measurements at the fourth sampling point must be considered carefully because of problems observed during the mineralization phase.)

3.3. Analyses of \(^{3}\)H and calculation of the dilution factor

The \(^{3}\)H released by the plant is supposed not to interact with the particulate phase. Consequently, this element can be used as a tracer: the analyses undertaken at each station under equilibrium conditions allow a good estimation of the dilution factor of the radioactive effluent in the cooling water to be made. This dilution factor is extrapolated to the other radionuclides; it is therefore possible to estimate the total concentration of each radionuclide in the channel.

3.4. Suspended solid concentrations and size distribution

Because of low longitudinal velocities, a high sedimentation rate is generally observed in the channel. It is therefore necessary to measure regularly the suspended solid concentrations at each station; these measurements are undertaken every 1.5 hours using filters of 1.2 \( \mu m \) pore size.

For size distribution analysis, water samples of 20 L are concentrated to a volume of 1 L, using tangential flow microfiltration on membranes of 0.1 \( \mu m \) pore size. The granulometric distribution is measured by laser beam diffraction, using a Coulter particle size analyser.
4. RESULTS

4.1. Characteristics of the water

The characteristics of the water flowing through the channel are as follows: mean temperature = 27°C; pH = 8; conductivity = 360 μS/cm; mean total suspended solids at station 1 = 25 mg/L; mean organic matter = 10 mg/L; granulometric distribution at station 1 (percentage of the total particulate volume): [particle size < 2.5 μm] = 15%; [2.5 μm < particle size < 10 μm] = 52%; [particle size > 10 μm] = 33%.

4.2. Dilution factor and sedimentation rate

For a good interpretation of the measurements of the radioactivity adsorbed on suspended matter, it is first necessary to calculate the total concentration of the radionuclides in the channel and the sedimentation rate.

The measurements of $^3$H in the channel under equilibrium conditions allow the dilution factor of the effluent in the cooling water to be determined and the 'total concentration' of the radionuclides at each station to be calculated. The 'total concentration' $(R)_{tot}$ (in Bq/m$^3$) is defined as follows:

\[
(R)_{tot} = (R)_{dis} + (SS) (R)_{SS} + \sum_{i=0}^{St.i} (R)_{SS} d(SS) \tag{1}
\]

where $(R)_{dis}$ is the dissolved radionuclide concentration (in Bq/m$^3$) at station $i$; $(R)_{SS}$ is the concentration of the radionuclide adsorbed on suspended solids (in Bq/kg dry weight) at station $i$; $(SS)$ is the suspended solid concentration (in kg/m$^3$) at station $i$; and

\[
\sum_{i=0}^{St.i} (R)_{SS} d(SS)
\]

is the concentration (in Bq/m$^3$) of the radionuclides adsorbed on suspended solids which have settled upstream of station $i$.

According to this definition, the 'total concentrations' of the radionuclides in the channel are as follows: $^{110}$Ag = 275 Bq/m$^3$; $^{58}$Co = 220 Bq/m$^3$; $^{60}$Co = 44 Bq/m$^3$; $^{54}$Mn = 6.3 Bq/m$^3$.

The sedimentation rate SED is calculated using the measurements of the suspended solid concentrations obtained at each station. The following value has been determined: SED = 1.6 mg·L$^{-1}$·h$^{-1}$. The specific sedimentation rate $S E D_i$ of each granulometric class (class 1 > 10 μm; 2.5 μm < class 2 < 10 μm;
FIG. 2. Total radioactivity of the suspended matter at each station.
FIG. 3. Radioactivity of suspended matter sampled on each filter (10, 2.5 and 0.5 μm) at station 3.
class 3 < 2.5 μm) can be estimated using the evolution of the granulometric distribution of the suspended matter. The following values have been found: \( \text{SED}_1 = 0.86 \, \text{mg} \cdot \text{L}^{-1} \cdot \text{h}^{-1} \), \( \text{SED}_2 = 0.56 \, \text{mg} \cdot \text{L}^{-1} \cdot \text{h}^{-1} \), \( \text{SED}_3 = 0.18 \, \text{mg} \cdot \text{L}^{-1} \cdot \text{h}^{-1} \).

4.3. Behaviour of \(^{110}\text{Ag}^m\)

The predominant radionuclide in the released effluent is \(^{110}\text{Ag}^m\). The measurements of \(^{110}\text{Ag}^m\) adsorbed on the total suspended matter at each station are presented in Fig. 2. The radioactivity of the suspended matter increases significantly between the first and the third stations; equilibrium seems to be reached downstream of the third station. The influence of particle size on the adsorption process is shown in Fig. 3 (considering the equilibrium conditions obtained at station 3); the observations prove that the fixation of \(^{110}\text{Ag}^m\) is much more important for the smallest particles.

As equilibrium is attained at the third station, it is possible to calculate the \(K_d\) value, expressed as the concentration ratio between the radioactivity of the soluble phase and that of the sorbed phase at this sampling point. The dissolved part of \(^{110}\text{Ag}^m\) at station 3 is calculated using balance equation (1). To solve this equation, it is necessary to determine the radioactivity settled upstream of station 3.

The value of \(\int_{\text{St.0}}^{\text{St.1}} (R)_{\text{SS}} \, d(\text{SS})\) is estimated as follows:

\[
\sum_{\text{St.} \, j = 0}^{\text{St.} \, j = 2} \sum_{\text{class} \, i = 1}^{\text{class} \, i = 3} (R)_{\text{SS}_{i \, j} + 1} \cdot \text{SED}_i \cdot \Delta t_{i, \, j + 1}
\]

where \((R)_{\text{SS}_{i \, j + 1}}\) is the mean particulate radioactivity (in Bq/kg dry weight) between stations \(j\) and \(j + 1\); \(\text{SED}_i\) is the sedimentation rate (in kg·m\(^{-3}\)·h\(^{-1}\)) of granulometric class \(i\); and \(\Delta t_{i, \, j + 1}\) is the transport time between stations \(j\) and \(j + 1\).

According to this methodology, the calculated \(K_d\) value is:

\[
K_d \left( ^{110}\text{Ag}^m \right) = 210 \, \text{m}^3/\text{kg}
\]

This global \(K_d\) value can be broken up into \(K_d\) values for the different granulometric classes:

\[
K_d \left( ^{110}\text{Ag}^m, \text{ particle size } > 10 \, \mu\text{m} \right) = 80 \, \text{m}^3/\text{kg}
\]
\[
K_d \left( ^{110}\text{Ag}^m, 10 \, \mu\text{m} < \text{ particle size } < 2.5 \, \mu\text{m} \right) = 260 \, \text{m}^3/\text{kg}
\]
\[
K_d \left( ^{110}\text{Ag}^m, 2.5 \, \mu\text{m} < \text{ particle size } < 0.5 \, \mu\text{m} \right) = 560 \, \text{m}^3/\text{kg}.
\]

The \(K_d\) values calculated for \(^{110}\text{Ag}^m\) show that this radionuclide has a high affinity for particles; this result confirms those reported in the literature [1, 3]. Besides, the
fixation is directly proportional to the specific surface of the particles; this observation suggests that the transfer of $^{110}\text{Ag}^m$ to the solid phase is governed by an ion exchange mechanism and/or direct adsorption.

4.4. Behaviour of $^{58}\text{Co}$, $^{60}\text{Co}$ and $^{54}\text{Mn}$

The measurements of $^{58}\text{Co}$, $^{60}\text{Co}$ and $^{54}\text{Mn}$ in the particulate phase at each station are presented in Fig. 2. For these radionuclides, equilibrium is attained rapidly.

In the case of radiocobalt, these observations seem to be at variance with some results described in the literature: in vitro experiments have shown that radioactive equilibrium between water and particles is not reached in less than a few days [8–10]. However, these results indicate that the uptake of radiocobalt has a two-step kinetics; the rapid initial uptake of most of the radiocobalt is followed by a slower increase of radioactivity in the particulate phase. Because of the relatively short residence time of the water in the experimental channel of Golfech, it will probably not be possible to observe the slower adsorption mechanism. Nevertheless, the measurements describe the consequences of the rapid uptake stage, which is the most important in terms of adsorbed amount of radioactivity.

The influence of particle size on the adsorption process is shown in Fig. 3. The largest particles adsorb less radioactivity than the intermediate and the smallest ones. Unlike the analyses for $^{110}\text{Ag}^m$, the measurements of $^{58}\text{Co}$, $^{60}\text{Co}$ and $^{54}\text{Mn}$ in the intermediate class and the smallest class are equivalent.

For the calculation of the global $K_d$ value and the specific $K_d$ values of $^{58}\text{Co}$, $^{60}\text{Co}$ and $^{54}\text{Mn}$, the method previously described has been applied. The results are summarized in Table I.

The global $K_d$ value of $^{54}\text{Mn}$ thus calculated is higher than those reported in the literature [1–3]. The high transfer of $^{54}\text{Mn}$ to solid particulate forms may result from precipitate formation; in fact, the total concentration of $^{54}\text{Mn}$ ($10^{-16}$ mol/L) in the channel corresponds to the solubility of $\text{MnO}_2$ at pH8.

<table>
<thead>
<tr>
<th>K_d (particle size &gt; 10 µm)</th>
<th>$^{58}\text{Co}$</th>
<th>$^{60}\text{Co}$</th>
<th>$^{54}\text{Mn}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>K_d (10 µm &lt; particle size &lt; 2.5 µm)</td>
<td>36</td>
<td>81</td>
<td>238</td>
</tr>
<tr>
<td>K_d (2.5 µm &lt; particle size &lt; 0.5 µm)</td>
<td>36</td>
<td>73</td>
<td>231</td>
</tr>
<tr>
<td>K_d (all particles)</td>
<td>25</td>
<td>48</td>
<td>164</td>
</tr>
</tbody>
</table>

TABLE I. $K_d$ VALUES (m$^3$/kg) OF $^{58}\text{Co}$, $^{60}\text{Co}$ AND $^{54}\text{Mn}$
Large differences may be found between the radiocobalt $K_d$ values obtained from field measurements in various localities or for various seasons [1–5]. Indeed, $K_d$ for radiocobalt depends significantly on parameters such as pH [10], speciation of cobalt in the water [9], and concentration and composition of the particulate phase [5, 10]. In order to obtain more detailed information about radiocobalt behaviour, and especially to determine precisely its speciation in the particulate phase (fractions associated with carbonates, organic matter, etc.), an experiment will be carried out with the Golfech experimental physical model.

5. CONCLUSIONS

The experimental physical model described in this paper is useful for simulating the behaviour of radionuclides in freshwater streams. The measurements undertaken during radioactive discharges of low radwastes reflect the situation of a natural ecosystem better than laboratory experiments. Besides, the radioactive effluents are much less diluted in the experimental channel than in the river; consequently, it is possible to carry out a larger number of analyses in different compartments of the ecosystem. The only disadvantage is that some radionuclides may exhibit specific chemical reactions (precipitation, etc.) at these high concentrations.

The results of the experiment presented in this paper give information about the following points:

(1) The influence of the particle size on the process of adsorption of radionuclides by suspended matter. The uptake of radioactivity is generally higher for the smallest particles (size < 2.5 μm for $^{110}$Ag, size < 10 μm for $^{60}$Co, $^{60}$Co and $^{54}$Mn).

(2) The exchange kinetics between water and suspended matter. Generally, radioactive equilibrium is rapidly reached. Consequently, it is possible to calculate the global $K_d$ value of each radionuclide and to determine exactly the specific $K_d$ value for each granulometric class.

In future, the experimental physical model of the nuclear power plant of Golfech could be used to study other compartments of the ecosystem (speciation of the bottom sediments, uptake/release by aquatic plants, etc.).

REFERENCES


MODELLING OF THE TRANSFORMATION OF SPECIATION PROCESSES OF CHERNOBYL ORIGIN $^{137}$Cs AND $^{90}$Sr IN THE SOIL AND IN BOTTOM SEDIMENTS

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Abstract

MODELLING OF THE TRANSFORMATION OF SPECIATION PROCESSES OF CHERNOBYL ORIGIN $^{137}$Cs AND $^{90}$Sr IN THE SOIL AND IN BOTTOM SEDIMENTS. The paper presents the results of systematic studies of physical and chemical processes associated with the behaviour of Chernobyl origin $^{137}$Cs and $^{90}$Sr in soil and water. The short term behaviour of radionuclides is governed by the ratio of the speciations in the fallout, which differs significantly for different regions. The long term behaviour of radionuclides is controlled by leaching of fuel particles in the zone around the nuclear power plant and by transformation of radionuclide speciations in the soil and in water bodies. A scheme for transformation of radionuclide speciations is proposed. Rate constants for the relevant physico-chemical processes have been obtained for different conditions. Major environmental parameters controlling migration characteristics of radionuclides are identified. A procedure for estimating the distribution coefficient $K_d$ is recommended.

1. HOT PARTICLES AS A SPECIFIC FEATURE OF THE CHERNOBYL ACCIDENT

During the explosion and the fire at Unit 4 of the Chernobyl nuclear power plant (NPP) in April–May 1986, large amounts of dispersed nuclear fuel (fuel hot particles), structural materials, and substances dumped into the reactor and formed in it (condensation hot particles) were released into the atmosphere. The fuel particles were either of a dense or of a loose structure and consisted of uranium oxides. Their sizes ranged from hundreds of micrometres to fractions of a micrometre. The radionuclide composition of the fuel particles was similar to the fuel make-up in the damaged unit, with some depletion of volatile nuclides ($^{134,137}$Cs, $^{106}$Ru, etc.). Fuel particles account for more than 90% of the total amount of hot particles. The condensation particles are generally characterized by smaller size and regular form. They include either a wide spectrum of radionuclides or only one to two radionuclides (for example, $^{134,137}$Cs, $^{106}$Ru, $^{144}$Ce + $^{95}$Zr, $^{144}$Ce + $^{106}$Ru).

Release of these fuel and condensation particles into the environment was the main distinguishing feature of the contamination following the Chernobyl accident.
Fallout from nuclear weapons testing had more than 90% of $^{90}$Sr and $^{137}$Cs in water soluble and exchangeable forms (i.e. extractable by neutral salt solutions). The fraction of water soluble and exchangeable forms in the Chernobyl fallout was much lower because of the presence of water insoluble hot particles and depended on the distance from the damaged unit. For example, the fraction of non-exchangeable $^{137}$Cs in the fallout near Chernobyl was about 75% [1], in the Bryansk region it was 50–60% and in Cumbria (United Kingdom) it was about 10% [2]. As a result, the proportion of exchangeable radionuclides in soils in the zone near the plant during the first years after the accident was much lower than that after nuclear weapons testing [3]. For this reason, the Chernobyl radionuclides had higher values of the distribution coefficient in the “soil-water” system and, hence, slower migration. Wash-off of dissolved $^{90}$Sr with surface run-off in the 30 km zone in 1986–1987 was much lower than that at sites where $^{90}$Sr was originally in the form of a soluble salt [1].

Apart from the differences in radionuclide speciation between Chernobyl fallout and fallout from nuclear weapons testing, the radionuclides also differ by the direction of their transformation in the soil. The mobility of radionuclides from nuclear weapons testing decreases with time because of fixation by soil components, while in the zone near the Chernobyl NPP in the first years after the accident the predominant process was leaching of radionuclides from hot particles, which led to increased migration [4].

Thus, because of the peculiar features of the Chernobyl fallout, it was not possible to use directly the results of studies of radionuclides from nuclear weapons testing, and this is why the behaviour of hot particles in the environment needs in-depth investigation.

2. KINETICS OF RADIONUCLIDE LEACHING FROM HOT PARTICLES

Physically based modelling of leaching processes is not easy because of the non-uniformity of sizes and forms and the chemical nature of particles. Besides, for using the results of such modelling for practical purposes, it is necessary to have experimental data on the distribution of particles of different composition and size over a significant area and data on the characteristics of each type of particle. In view of this, it seems reasonable to use for forecasting the integral parameters characterizing the rate of radionuclide leaching in different parts of the contaminated zone. The first order rate constant $k_j$ (leaching) could serve as such a parameter. As the hot particles dissolve, their sizes decrease and the proportion of radionuclides in particles of highest solubility increases. The first process results in an increase of $k_j$ and the second process results in its decrease. The assumption of a balance between the above processes makes it possible to use first order equations to describe the kinetics of radionuclide leaching from hot particles. Values of $k_j$ may be obtained.
TABLE I. RATE CONSTANTS FOR $^{90}$Sr LEACHING FROM HOT FUEL PARTICLES

<table>
<thead>
<tr>
<th>Distance from the NPP (km)</th>
<th>Location</th>
<th>Rate constant ($10^{-3}$ day$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4–10</td>
<td>Floodplain of the river Pripyat</td>
<td>0.15–0.87</td>
</tr>
<tr>
<td></td>
<td>Kopachi</td>
<td>0.40–0.48</td>
</tr>
<tr>
<td>10–20</td>
<td>Benevka</td>
<td>1.4–1.7</td>
</tr>
<tr>
<td></td>
<td>Chernobyl</td>
<td>1.5</td>
</tr>
<tr>
<td></td>
<td>Korogod</td>
<td>1.0–3.6</td>
</tr>
</tbody>
</table>

from data on the transformation of radionuclide forms in soils. The proportion of a radionuclide in fuel particles is assumed to be equal to the fraction of a radionuclide in non-exchangeable form minus the fraction fixed by the soil. In such a calculation it is advantageous to use data on $^{90}$Sr, which is weakly fixed by soils. The results of calculations of rate constants for $^{90}$Sr leaching from hot fuel particles are given in Table I. The indicated constants were used for estimating the proportion of the exchangeable form of radionuclides in the soils of the 30 km zone of the Chernobyl NPP. The predicted values and measurements were in good agreement.

The leaching rate constants given in Table I agree with the values of $k_t$ calculated from the dissolution rate of fuel particles [5]. These constants ranged from $10^{-3}$ to $10^{-4}$ d$^{-1}$ for particles of 50–500 μm (1 km zone) and from $10^{-2}$ to $10^{-3}$ d$^{-1}$ for particles of 5–50 μm (Chernobyl region).

Calculations and measurements of the proportion of the water soluble and exchangeable forms of $^{90}$Sr indicate that, at present, the amount of radionuclides in fuel particles is considerable only in the 10 km zone of the Chernobyl NPP and that at larger distances the hot particles have mostly disintegrated.

3. BASIC TRANSFORMATION PROCESSES OF CHEMICAL SPECIATIONS OF RADIONUCLIDES IN THE SOIL AND IN BOTTOM SEDIMENTS

The most expedient way to classify speciations of radionuclides is to divide them into water soluble, exchangeable and non-exchangeable (fixed) chemical forms in soil [3, 4]. Different chemical forms can be separated by the technique of sequential extraction with solutions of different compositions. The equilibrium between
water soluble and exchangeable forms sets in fairly quickly and in many cases it is reasonable to consider them as a single mobile form. In terms of chemical forms, fixation of radionuclides is the transfer from their mobile form to their fixed form. For a mathematical description of this process, models have been proposed in which fixation is treated as an irreversible process [4, 6]. The data available in the literature suggest that at the initial stage of fixation and for processes of a relatively short time-scale, this assumption is warranted. However, the data for long term transformation of chemical forms of radionuclides in soil indicate the existence of a remobilization process that is the reverse of fixation. After application of $^{137}\text{Cs}$ and $^{90}\text{Sr}$ solutions to the soil, the fraction of their exchangeable form does not decrease to zero, as is supposed to happen during irreversible fixation, but it decreases to a certain stationary level, independent of the amount of radionuclide applied and not changing significantly, at least for several years [7]. Allowing for fixation reversibility and assuming two mechanisms of fixation selective adsorption and diffusion into the solid phase of the soil, the transformation of chemical forms of radionuclides in the soil can be illustrated schematically (see Fig. 1).

Analytical solution of differential equations corresponding to the schematic diagram shown in Fig. 1 gives the following time dependence of mobile forms of Chernobyl radionuclides after their deposition on soil:

\[ k, f, r \text{ are first order rate constants for radionuclide leaching from fuel particles, fixation and remobilization, respectively.} \]
\[ m = (1 - K_s) \left[ \left( \frac{k_f m_0 + k_r - k_f}{k_f + k_r - k_f} - \frac{k_r}{k_f - k_r} \right) e^{-(k_f + k_r)t} \right] \\
+ (1 - m_0) \left( \frac{k_f}{k_f + k_r - k_f} e^{-k_r t} + \frac{k_r}{k_f + k_r} \right) \]

where \( m \) and \( m_0 \) are the current and the initial fractions of the mobile forms in soil, respectively; \( K_s \) is the equilibrium constant of selective sorption; and \( k_f, k_r \) and \( k_r \) are the first order rate constants of radionuclide leaching from fuel particles, fixation and remobilization, respectively.

Experimentally determined transformation parameters of \(^{137}\text{Cs}\) for different soil types in the 30 km zone of the Chernobyl NPP are presented in Table II. Figure 2 illustrates the calculated dependence of the mobile part of \(^{137}\text{Cs}\) and \(^{90}\text{Sr}\) in various areas of the contaminated territory after the Chernobyl accident, as well as the influence of the composition of the original chemical forms on the direction of the transformation processes and the current ratio of the chemical forms of \(^{90}\text{Sr}\) and \(^{137}\text{Cs}\).

4. DIFFUSION MODEL OF RADIONUCLIDE FIXATION BY SOILS AND SEDIMENTS

The models available account for fixation as a usual first order chemical reaction [4, 6, 8]. For modelling of caesium fixation during the first month after its introduction into the soil suspension, three parameters are sufficient; however, in order to achieve satisfactory agreement between calculations and experimental data for longer interaction times, a model with four to six parameters is required. The large number of required parameters makes it difficult to use the models in practice.

<table>
<thead>
<tr>
<th>Soil type</th>
<th>( k_f \times 10^2 ) (d(^{-1}))</th>
<th>( k_r \times 10^3 ) (d(^{-1}))</th>
<th>( K_s )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sod-podzol, clay</td>
<td>1.1 ± 0.3</td>
<td>2.9 ± 1.0</td>
<td>0.41 ± 0.04</td>
</tr>
<tr>
<td>Sod-podzol, sand</td>
<td>1.7 ± 0.2</td>
<td>4.4 ± 0.5</td>
<td>0.54 ± 0.02</td>
</tr>
<tr>
<td>Peat bog soil</td>
<td>3.9 ± 0.4</td>
<td>5.2 ± 0.8</td>
<td>0.70 ± 0.02</td>
</tr>
</tbody>
</table>
Moreover, these parameters are an unknown function of the chemical composition of the liquid phase and there are no methods to determine them independently or to assess them theoretically. The principal cause of these difficulties is that first order kinetics is not quite adequate to the mechanism of radiocaesium fixation.

In this connection, we have started to develop a model which can more adequately describe the transformation of radiocaesium species in soils and sediments. The model describes fixation as diffusion of radionuclides in the surface layer of the soil and in sediment particles. If the thickness of the diffusion layer is much smaller than the characteristic particle sizes, fixation can be regarded as the diffusion of dissolved matter from a limited volume of a well mixed solution into a plane.

In experimental studies of the kinetics of radiocaesium fixation cited in the literature, the radionuclide was introduced into an initially uncontaminated suspension. The solution of the diffusion equation for these initial conditions is given in Ref. [9]. It is rather awkward for practical calculations, but, for a long time period and a small equilibrium portion of radionuclide in solution, it can be simplified to

$$W_t = A + Bt^{-0.5}$$

where \(A = W_\infty\), \(B = W_\infty/((\pi D)^{0.5})\), \(W_t\) and \(W_\infty\) are the fractions of the radionuclide in solution at time \(t\) and at equilibrium, respectively, and \(D\) is the diffusion coefficient.
When using Eq. (2), one should take into account the heterogeneity of the solid phase of soils and bottom sediments comprising complicated multicomponent systems. The simplest way to do this is to divide the adsorption sites of soils and sediments capable of interacting with caesium into two groups: exchangeable sites and fixed sites.

Such a simplification allows a system with inhomogeneous parameters to be described by the same equation as is used for a homogeneous system, using the sum of fractions of exchangeable and dissolved forms of radionuclides instead of the fraction of the radionuclide in solution. This can be done because adding the system with inhomogeneous parameters to the exchange/sorption fraction, for which equilibrium with the solution sets in instantaneously, is mathematically equivalent to an increase in the initial volume of the liquid phase:

$$M_t = A^* + B^* \cdot t^{-0.5}$$

(3)

where $A^* = M_\infty$, $B^* = M_\infty/\left(\pi D\right)^{0.5}$, and $M_t$ and $M_\infty$ are the fractions of the fixed and dissolved forms at time $t$ and at equilibrium, respectively. $M_t$ is by definition at equilibrium with $W_t$ and their relation can be written as

$$M_t = W_t \left(1 - K_{d}^{eq} S/L\right)$$

(4)

where $K_{d}^{eq}$ is the distribution coefficient of the exchangeable form and $S/L$ is the ratio of the solid phase weight to the solution volume.

By combining Eqs (3) and (4), we obtain

$$W_t = \frac{(A^* + B^* \cdot t^{-0.5})}{(1 + K_{d}^{eq} S/L)}$$

(5)

To test the suitability of the model for predicting radionuclide fixation in sediments, Eq. (5) was used to describe the kinetics of $^{137}$Cs fixation in soils [7, 10] and sediments [11]. The results presented in Refs [10, 11] have been obtained for suspensions of soils and sediments, whereas Ref. [7] gives data for radiocaesium fixation in soils with a natural moisture content.

A comparison shows that Eq. (2) describes fairly accurately the kinetics of radionuclide fixation in a time interval of several hours to several months, both in sediment and soil suspensions and in soils with a natural moisture content. Thus, the proposed model seems to reflect adequately the actual mechanism of radiocaesium fixation in soils and bottom sediments and can be recommended for practical use. Its major advantage is the small number of parameters, which have a clear physical meaning and can, in principle, be determined in an independent experiment or assessed theoretically from the properties of the solid and liquid phases of the natural soil–water system.
5. ESTIMATING THE RADIONUCLIDE DISTRIBUTION COEFFICIENT

The need of a specific distribution coefficient for modelling of a specific system is due to the fact that, on the one hand, this parameter governs the migration rate of radionuclides in natural water systems and their buildup in fish and vegetation, and, on the other hand, there are no all-purpose $K_d$ values for use in modelling the behaviour of radionuclides in any natural water ecosystem. For example, the variance of $K_d$ values for $^{137}$Cs in surface water is known to be four orders of magnitude. Moreover, the difference in $K_d$ at one site for different layers of lake bottom sediment can be as high as two orders of magnitude [6].

The $K_d$ value is also dependent on the time after radionuclide entrance into soil or bottom sediments (ageing effect). Therefore, a reliable prediction of the behaviour of radionuclides is possible only if the distribution coefficients for a given soil–water system at a given time are assessed on the basis of knowledge of the mechanisms and kinetics of sorption/desorption processes.

A prediction of the radionuclide distribution in soil–water systems should, generally, be performed in two stages. The first step is the prediction of the chemical transformation of speciations and the determination of the portion of radionuclides in exchangeable form. This prediction is based on modelling of the transformation processes as described in Sections 3 and 4.

The goal of the second step is to assess $K_d$ values for the exchangeable form of a radionuclide. For estimating this distribution coefficient, two approaches can be used. Both assume that $^{137}$Cs and $^{90}$Sr in solution occur in the cation form and their exchangeable sorption is described by the equation for cation exchange. The major difference between the two approaches is the way in which the non-uniformity of adsorption sites of soils and sediments is accounted for. In the first approach, the external non-homogeneities of adsorption sites are minimized by using a cation analogue of the radionuclide as a competing cation $M$ having sorption properties as close as possible to those of the radionuclide [4, 12, 13]. For determination of the required parameters, a standard procedure of sequential extractions is used [14, 15]. The concentrations of radionuclides and competing cations are determined in a water extraction, and the proportion of exchangeable forms of radionuclides in the solid phase is calculated from their concentrations in an acetate extraction. The data obtained are used for calculation of the selectivity coefficient:

$$K_c = \frac{[R]_{ex} [M]_w}{[R]_w [M]_{ex}}$$

(6)

where $M = K, NH_4$ for $^{137}$Cs, and $M = Ca, Mg$ for $^{90}$Sr.

The calculated selectivity coefficient is then used to predict the distribution coefficient of the exchangeable form of the radionuclide:
Among the advantages of the procedure is the simplicity of the experimental determination of parameters. Besides, parameters can also be assessed from literature data. In particular, data on the proportion of exchangeable forms of radio-nuclides and competing cations in soils and sediments, determined by a standard method, are available for many natural soil-water systems.

The method has been used successfully for predicting the distribution of $^{90}$Sr and $^{137}$Cs in lake bottom sediments [4] and their concentrations in surface run-off [13].

A limitation of the method is that the $K_c$ value may, strictly speaking, depend on the composition of the liquid phase, and, in those cases where this composition differs significantly from the composition at which $K_c$ was found, the value should be recalculated. This adds uncertainty to the prediction.

In the approach proposed by Cremers and co-workers [16], the non-uniformity of adsorption sites is taken into account by dividing them into selective sites (FES) and regular sites (RES). The distribution coefficient is then calculated as a sum of $K_d$ values for FES and RES:

$$K_d = K_d^{(FES)} + K_d^{(RES)} = \frac{K_c^{(FES)} Z_K}{[K]_w} + \frac{K_c^{(RES)} [K]_w}{[K]_w}$$

where $[FES]$ is the concentration of FES in the solid phase (μeq/g), $Z_K$ is the part of FES occupied by potassium, and $K_c^{(FES)}$ is the selectivity coefficient for exchange of caesium by potassium on FES. The distribution coefficient for FES can also be found by dividing the specific radiocaesium interception potential $K_d^{mK}$ by the concentration of potassium in the water phase.

The advantage of this method is the constant values of $K_c^{(FES)}$ for different soils [8] and better accounting for the sorption non-uniformity of soils.

The main disadvantage of the method is the complexity of the experimental procedure for parameter determination. Moreover, the border between exchangeable sorption and fixation is rather blurred and, as a result, the prediction does not give the distribution coefficient of the exchangeable form, but the total amount of the radionuclide $K_d^{(tot)}$ after its ageing under the conditions of the experimental procedure during 24 h.

On the whole, it can be concluded that application of both procedures will make it possible to obtain satisfactory estimates of the distribution coefficient of the exchangeable form of the radionuclide; the choice of method in each particular case should be dictated by the availability of the necessary parameters.
6. CONCLUSIONS

It has been shown that the transformation of chemical speciation of radionuclides in soils and sediments can be described by first order kinetic equations. A three-box model is adequate to describe experimental data on radiocaesium fixation in soils. An additional box is needed when a significant fraction of radionuclides is incorporated into the volume of fuel particles. Values of the rate constants of radionuclides leaching from fuel particles in the Chernobyl area have been determined as a function of the distance from the NPP. Radionuclide fixation has been modelled as a two-step reversible process. The initial, very fast, step of fixation was assumed to be instantaneous. This is followed by a slower process with a time-scale of several months for radiocaesium. Caesium fixation parameter values have been determined for typical soils of the Chernobyl area. A more accurate diffusion model of $^{137}\text{Cs}$ fixation is proposed. The distribution of radiostrontium and radiocaesium in exchangeable form in soil-water systems is governed by cation exchange, with $K_{Ca}$ about 1 and $K_{K}$ about 10. Cation exchange equations have been used for the prediction of the distribution of these radionuclides between solid and liquid phases.

REFERENCES


ENVIRONMENTAL MODEL TESTING

(Session 6)

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USE OF CHERNOBYL DATA TO TEST PREDICTIONS AND UNCERTAINTY ESTIMATES FROM EXPOSURE ASSESSMENT MODELS

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Abstract

USE OF CHERNOBYL DATA TO TEST PREDICTIONS AND UNCERTAINTY ESTIMATES FROM EXPOSURE ASSESSMENT MODELS.

Measurements of radioactive fallout from the Chernobyl accident have provided an opportunity for environmental assessors to test model predictions and uncertainty estimates against independent data sets. The Multiple Pathways Assessment Working Group of the IAEA/CEC Co-ordinated Research Programme on Validation of Environmental Model Predictions (VAMP) has organized the development of several test scenarios using data collected after the Chernobyl accident. Model testing exercises have been completed for two sites, Central Bohemia in the Czech Republic (Scenario CB) and southern Finland (Scenario S). These exercises have shown that, in general, multiple pathways assessments and predictions of time averaged or time integrated concentrations were more accurate than predictions of concentrations for single exposure pathways and specific time periods. Model predictions were generally within an order of magnitude of the observed or estimated values. When site specific data and element specific transfer coefficients were available, experienced analysts achieved accuracy to within a factor of two. Several conclusions have been drawn to date: (1) Compensatory effects are commonplace and are revealed only when models are tested for both mid-points and end-points of the calculation. (2) Increased model complexity does not necessarily lead to increased accuracy. However, flexibility in model structure is important for addressing site specific situations. (3) The training and experience of the analyst are important factors in determining the accuracy of the prediction produced by any code. (4) Interpretation of imperfect data requires judgement, which varies from investigator to investigator. There will be differences among the results provided by independent groups of risk assessors, even when the same computer code is used. (5) Important assessments requiring a high level of credibility should be assigned to more than one group, and resources should be allocated to resolve discrepancies between groups. Multiple independent assessments should be encouraged in addition to peer review. (6) Models should be tested for a variety of sites and sets of conditions.

* Chairman, Multiple Pathways Assessment Working Group of the IAEA/CEC Co-ordinated Research Programme on Validation of Environmental Model Predictions (VAMP).
1. INTRODUCTION

Environmental radiological assessments are strongly dependent on the use of mathematical models to extrapolate beyond the realm of direct observations. Mathematical models can be used to project into the future in order to assist decisions concerning the acceptability of planned facilities. Mathematical models can also be used to reconstruct past situations, given the existence of partial or incomplete information. Models may also be used in current situations, for instance in an emergency response mode to direct monitoring activities and help determine the need for intervention to reduce exposures of populations or contamination of foodstuffs.

For models intended to produce realistic estimates, it is important to assess the accuracy of the models; for models intended to produce screening calculations, it is important to assess the extent of conservatism in the model. Where large discrepancies between model predictions and observations exist, it is necessary to resolve the reasons for these discrepancies and, where justified, implement the required corrections to the models.

Measurements of radioactive fallout from the Chernobyl accident have provided an opportunity for environmental assessors to test model predictions and uncertainty estimates against independent data sets. As a part of the IAEA/CEC Coordinated Research Programme on Validation of Environmental Model Predictions (VAMP), the Multiple Pathways Assessment Working Group has organized the development of several test scenarios using data collected in several countries following the Chernobyl accident.

Model testing exercises have been completed for two European sites, Central Bohemia in the Czech Republic (Scenario CB) and southern Finland (Scenario S). This paper describes the findings from these studies. Three new multiple pathways scenarios have also been developed and distributed: Scenario H (Hanford), which describes an $^{131}$I release in the United States of America in 1963; Scenario I (Iput River), which is based on data from the Bryansk region of Russia following the Chernobyl accident and which is the first multiple pathways scenario to consider the effects of countermeasures; and Scenario T (Techo River), which uses data from historic releases of radioactive substances into the Techa River in Russia. An additional scenario using Chernobyl data is under development for the Polesskoe region in Ukraine. These scenarios will be used in future model validation programmes sponsored by the IAEA.

2. METHODS

Each test scenario was initiated with a detailed description of the site during the time of arrival of the Chernobyl plume and of the amount of radionuclides measured in air, soil and water in 1986. This information was given to participants in
The test exercise; they were then challenged to predict time dependent quantities of $^{137}\text{Cs}$ in various types of food and fodder, time dependent concentrations of $^{137}\text{Cs}$ in the whole body for humans, the statistical distribution describing variability among individual whole body concentrations, and estimates of mean doses to adult humans from external and internal radiation exposure. For Scenario S, the foodstuffs included foods derived from the natural and semi-natural environment, such as mushrooms, berries, wild game and freshwater fish. Predictions were made for the years 1986–1989 for Scenario CB and 1986–1990 for Scenario S. Quantitative estimates of uncertainty were required for both the test data and the model predictions.

Scenarios CB and S constitute the first example of model testing we know of to require estimates of uncertainty on both model predictions and the observations. Uncertainties are placed on the test data to indicate that the data may be only partially relevant for the true but unknown values. The originators of the test data were asked to use judgement in assigning uncertainties when the data were not obtained from a purely randomized statistical design.

Access to the test data was restricted until after predictions and uncertainty estimates were submitted to the test co-ordinator. In Scenario S, as opposed to Scenario CB, the modellers were told the name of the test area and were permitted to ask questions of the authors of the scenario description. The test was blind only in that the actual test data for the scenario were not revealed until after predictions had been submitted. In addition, the participants were asked not to consult the published literature for the site.

Quantitative measurements of model performance were sought to facilitate objective comparisons of predictions and observations. In order to accommodate simultaneous comparison of several different aspects of model performance (e.g. peak values, the dynamic behaviour of the entire time series and overlapping uncertainty estimates), it was decided to use graphical comparisons. Comparisons across models were made by placing on the same page multiple graphs of similar scale and containing the same test data, with each individual graph containing the results from a single model. For end-points which did not involve a time series (e.g. deposition, dose estimates), the results for each end-point, with uncertainties, were displayed on a single graph.

3. RESULTS FROM THE TEST EXERCISES

The predictive capabilities of the various models were demonstrated for a large number of end-points, including estimates of internal and external doses. Complete results from all models and modellers are described in the reports for Scenarios CB and S [1, 2]. Model performance in many cases was excellent, with predictions within a factor of two of the observations; discrepancies between the test data and the estimates of annual average concentrations produced by most participants seldom
exceeded a factor of ten. In most cases, these discrepancies were biased high (i.e. the model predictions were overestimates).

Selected model predictions for the time variation of $^{137}$Cs concentrations in humans in the CB region are shown in Fig. 1 (left). The overestimates in the model predictions are due primarily to overestimates of $^{137}$Cs concentrations in foodstuffs and of amounts of foods consumed; the best results were obtained by modellers familiar with the region. Following disclosure of the test data, modellers were given an opportunity to improve their models. Revised predictions for the same modellers are also shown in Fig. 1 (right). Changes to models were based on improved understanding of the processes being modelled (e.g. metabolic transfer of $^{137}$Cs into meat and milk), as well as corrections to errors in the codes or parameter values. Predicted dose estimates for external and ingestion exposure over 3 years (CB) or 4.5 years (S) are shown in Fig. 2.

Scenario S has been the most comprehensive test of multiple pathways exposure assessment models conducted to date. This scenario has been the first of its kind to include portions of the human diet originating from the natural and seminatural ecosystem. It is also the first to test for the importance of the consumption of freshwater fish in the determination of human whole body concentrations of $^{137}$Cs initially deposited from the atmosphere. Examples of model predictions for $^{137}$Cs concentrations in fish and mushrooms in southern Finland are shown in Fig. 3; a comparison of predictions with estimates for the three major contributors to the ingestion dose in southern Finland is shown in Fig. 4. Selected predictions of the $^{137}$Cs concentrations in adult males in the same region are shown in Fig. 5, together with selected predictions of the distribution of the individual whole body concentrations.

The test data for Scenario S illustrate the differences in the dietary uptake and whole body concentrations of $^{137}$Cs for individuals due to differences in sex and age. The test data also demonstrated that different pathways dominated the exposure of the residents of southern Finland at different times following the contaminant release. In the first year or the first two years, milk was the major dietary contributor to exposure of the population, followed by meat (domestic beef) and fish; later on, fish became much more important than beef. Over a human lifetime, the most significant dietary components for the average resident of southern Finland are expected to be fish, milk and forest mushrooms, in that order.

Changes with time in the dominant pathways of human exposure were affected by soil composition and hence soil fixation of $^{137}$Cs. Food products derived from the natural ecosystem were particularly important because the bioavailability of $^{137}$Cs in the soils, especially the forest soils, did not decline much with time, and the soils therefore provided a continuing source of $^{137}$Cs to the food-chain. The $^{137}$Cs concentrations in barley and oats, much of which was grown in soils where $^{137}$Cs remained bioavailable, remained at relatively constant levels even several years after deposition; this trend was reflected in pork produced from these feeds.
FIG. 1. Comparison of selected model predictions with observations for whole body concentrations of $^{137}$Cs in humans in Central Bohemia. Initial (left) and revised (right) predictions are shown. Vertical bars indicate the 95% confidence intervals for the mean value of observations (filled circles); dashed lines indicate the 95% confidence intervals for the mean prediction (solid line).
The availability of two independent data sets for model testing provided an opportunity to compare the behaviour of the contaminant concentrations in two different locations over a period of several years. Differences between the test data from Scenarios CB and S were reduced to within a factor of two to three once the data were time integrated and normalized to the total amount of $^{137}\text{Cs}$ deposited within each region (Fig. 6). The largest difference observed between these locations was that the time integrated concentrations (normalized for total deposition) of $^{137}\text{Cs}$ did not level off as fast for Scenario S as for Scenario CB. The major explanations for this observation are thought to be differences between the two regions in the rates of caesium fixation in soil and the importance in southern Finland of food products from the natural ecosystem.

**FIG. 2.** Comparison of model predictions with estimated values for external dose and ingestion dose from $^{137}\text{Cs}$ after 3 years in Central Bohemia (left) and after 4.5 years in southern Finland (right).
FIG. 3. Comparison of selected model predictions with observations for mean concentrations of $^{137}\text{Cs}$ in fish and mushrooms in southern Finland. Vertical bars indicate the 95% confidence intervals for the mean value of observations (filled circles); dashed lines indicate the 95% confidence intervals for the mean prediction (solid line).

FIG. 4. Comparison of predictions with estimates for the three major contributors (in percentages) to the effective dose from ingestion in southern Finland for April 1986 to December 1990.
FIG. 5. (A–D) Comparison of selected model predictions with observations for whole body concentrations of $^{137}$Cs in adult males in southern Finland. Vertical bars indicate the 95% confidence intervals for the mean value of observations (filled circles); dashed lines indicate the 95% confidence intervals for the mean prediction (solid line).

(E) and (F) Comparison of selected model predictions with observations for the distribution of concentrations of $^{137}$Cs in individual adult males in southern Finland at the end of 1990.
FIG. 6. (A–D) Comparison of cumulative, time integrated $^{137}$Cs concentrations in milk, beef, pork and adult humans, respectively, for Central Bohemia (CB) and southern Finland (S). Concentrations are normalized for total deposition; the 95% confidence limits are indicated as bars.

(E) and (F) Comparison of distributions of observed whole body concentrations of $^{137}$Cs for adults (males and females) in Central Bohemia (CB) and southern Finland (S) for 1987 and 1990, respectively. Concentrations are normalized for total deposition; confidence limits represent only the uncertainty in the total deposition.
4. CONCLUSIONS

The use of data from the Chernobyl accident to test the predictions of exposure assessment models provided an opportunity for participants in the Multiple Pathways Working Group to intercompare, evaluate and improve their estimates of $^{137}$Cs in foods and humans. Through the process of a blind test and intercomparison of results, participants detected previously unidentified errors in computer codes, compensatory effects in assumptions of parameter values and dominant processes, and principal sources of uncertainty in their model predictions. In addition, extensive discussion among participants served to clarify issues regarding the methods for propagating and interpreting uncertainty, as well as separating uncertainty due to variability among individuals from uncertainty due to lack of knowledge about fixed but unknown quantities.

The primary factors affecting the accuracy of model predictions in these exercises were the experience of the model user, the ability of the user to change the structure of the model to address site specific processes and issues, and the availability of abundant site specific data. The least accurate model predictions tended to occur as a result of undetected mistakes, inexperience of the user, or application of generic default values rather than parameter values derived specifically for the conditions of the region. In many ways, exercises that were designed to test model predictions became tests of modellers and assessors.

The test exercises identified or re-emphasized a number of important aspects of successful modelling in general:

— The experience of the modeller, especially with respect to understanding the processes being modelled, is perhaps the single most important factor in determining successful model performance.
— Computer codes should permit flexibility in model structure to allow adaptation for site specific conditions.
— Compensatory effects in intermediate steps of an exposure pathway can give good results for the wrong reason.
— Inclusion of estimates of uncertainty for both the test data and the model predictions is absolutely essential for a sound interpretation of the results.
— Uncertainty estimates are highly dependent on judgement and thus differ among investigators.
— Uncertainty estimates produced by individual assessors frequently do not encompass the observed values; they are thus statements of overconfidence.
— As previously concluded by other model testing exercises (e.g. Refs [3, 4]), critical assessments should be performed by more than one assessor or modelling group. Multiple independent assessments are effective in disclosing discrepancies in user judgement and differences in interpretation of input data. In these cases, resources should be allocated to resolving these differences before drawing final conclusions.
The evaluation of model complexity and model performance must be considered in the light of the intended purpose and level of accuracy of the model in question. For this reason, participants were required to provide a detailed analysis of the performance of their own models or codes.

The process of testing independent model calculations against independent data sets provided useful information to the originators of the test data as well as to the modellers. The discussions led to rethinking of interpretations of some measurements by the developers of the test scenarios and to in-house revisions of some estimates of concentrations, dietary intakes and doses.

The data sets from the CB and S test exercises remain available both for additional modellers who wish to use some or all of the information to test their own models and for future modellers for use in the development or testing of new models. Inexperienced modellers can use these data sets to gain experience with their models or with the modelling process. The detailed IAEA reports about the CB and S test exercises include documentation of the test data and of each model (including model structure, equations, assumptions and parameter values), graphical presentations of model results compared both with other model results and with the test data, estimates of uncertainty for both the test data and most model predictions, and the modellers' own evaluations of their performance in the test exercise [1, 2].

The task of model validation has only begun — it is not finished. Participants in the Multiple Pathways Assessment Working Group have identified a number of needs for the future:

— More testing at the process level;
— More testing for pathway mid-points;
— Testing for end-points of critical population groups (e.g. agricultural workers, dairy farmers, hunters, fishermen and harvesters of forest mushrooms);
— Testing at more sites;
— Testing for a wide variety of radionuclides;
— Extending testing to include non-radioactive trace contaminants.

REFERENCES


MODELLING OF RADIOCAESIUM IN LAKES
Results from the VAMP project

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Abstract

MODELLING OF RADIOCAESIUM IN LAKES: RESULTS FROM THE VAMP PROJECT.

The paper provides an overview of the main results from the IAEA VAMP (Validation of Environmental Model Predictions) project, which will be presented in a comprehensive technical report that will be published by the IAEA. Seven models for radiocaesium in lakes have been critically examined. The predictions ($^{137}$Cs in water, sediments and biota) have been validated with data from seven European lakes covering very wide ranges in most limnological characteristics and Chernobyl fallout. The models differ widely in terms of size (number of variables and processes accounted for) and set-up (definitions of fundamental model...
variables, such as retention of water and radiocaesium, bio-uptake rates, internal loading, seasonal variability, etc.). Much work has been done on critical evaluations of the models (e.g. sensitivity and uncertainty tests). An important objective has been to identify the most fundamental processes regulating the distribution and bio-uptake of radiocaesium in lakes. A new model for predicting radiocaesium in water and predatory fish has been developed. A definition of the important concept of ‘predictive power’ is presented. The paper discusses the ‘sensitivity’ of lakes to radiocaesium as well as various remedial strategies.

1. INTRODUCTION

The results presented here emanate from the IAEA VAMP (Validation of Environmental Model Predictions) project. The work carried out by the Lake Sub-Group will be presented in a comprehensive technical report that will be published by the IAEA. That report will give a thorough description of seven lakes (Iso Valkjärvi, Finland; Bracciano, Italy; Øvre Heimdalsvatn, Norway; IJsselmeer, Netherlands; Hillesjön, Sweden; Devoke Water and Esthwaite Water, United Kingdom), which differ in most limnological characteristics (size, trophic level, food-web, etc., see Table I). The concentration of radiocaesium in the fallout after the Chernobyl accident varies from less than 1 kBq/m$^2$ to more than 100 Bq/m$^2$ among these lakes.

Eight models have been tested. They differ in size (i.e. number of compartments and processes accounted for) and structure (i.e. conceptual set-up), but all of them focus on predictions for radiocaesium in water (used for drinking, irrigation, etc.) and in predatory fish (used by man for consumption). The technical report mentioned above evaluates the data and the models. It presents different approaches for critical evaluations, such as sensitivity and uncertainty analyses. It also discusses important aspects of predictive, generic lake ecosystem models, such as the optimal size of predictive models, modelling of special climatological conditions (e.g. transport in connection with snow and ice), model comparisons, modelling of dietary shifts, and ecological and biological half-lives. The report also defines the concept of ‘predictive power’ and presents a new state-of-the-art model for radiocaesium in lakes — the VAMP model.

The VAMP Aquatic Working Group has also made a review of remedial strategies for lakes and rivers, with special focus on how ‘sensitive’ aquatic systems are to radiocaesium, and approaches to quantify lake sensitivity. The objective of this paper is not to summarize all these results or to give an overview of the results. Instead, the aim is to highlight some interesting and important topics: (a) Experiences of general interest gained during the project. (b) Definition of the predictive power, because this is of great general importance in all types of modelling and especially in model validations. This part will also focus on the optimal size
of dynamic models. (c) The practical use of these new modelling techniques, because many of them may be used not only for lakes but also for other types of ecosystem and for substances other than radiocaesium.

2. GENERAL EXPERIENCE

The first part of the VAMP project focused on the collection and structuring of the empirical data, and the second part focused on the models and critical evaluations of the models. At an early stage, it was decided that this project should not repeat what had already been done by, e.g., the BIOMOVS project. So, the idea was not to make blind tests of the various models for different scenarios and then to compare the model results. Instead, the emphasis was on critical evaluations of the data, the technical details (algorithms, sub-models, definitions, concepts, etc.) of the models, which determine the predictive power and the usefulness of the models, and on model validations. The work was carried out in a small group, openly, informally and very constructively. Initially, it was also decided that all models should focus on predictions of radiocaesium in lake water and in predatory fish, and the basic task was to identify the most important processes regulating the transport of radiocaesium. This is easy to state but difficult to accomplish. It was also decided that the project should not stop at this, but that it should also try to build a new model based on those processes and the experience gained during the project. So, the new VAMP model is meant to be a state-of-the-art model. It is, however, not intended to be a general, comprehensive ecosystem model which accounts for all possible processes regulating the transport and bio-uptake of radiocaesium in lakes, but a predictive and practically useful model based on the most important processes and factors.

This section gives information on the new VAMP model. It has nine specific components, which are meant to increase the predictive accuracy of the model and to make it easy to apply for lakes in general. These components will be presented in greater detail in the above mentioned report.

(1) The seasonal variability moderator for water discharge. This is a new technique for predicting seasonal (monthly) variability in tributary discharge from simple estimates of the mean annual water discharge, which is estimated by traditional methods based on mean annual precipitation, size of catchment and specific run-off.

(2) The dimensionless moderator for the water retention rate. This is a new, general technique for estimating a time dependent value of the water retention rate rather than a constant value. Instead of defining the retention rate as $1/T$, where $T = V/Q$ ($T$ — water retention time, $V$ — volume, $Q$ — mean annual water discharge), this retention rate is given as $1/T(t)$ or $Q(t)/V$. 
**TABLE I. DATA FOR THE SEVEN VAMP LAKES**

<table>
<thead>
<tr>
<th>Lake</th>
<th>Altitude (m.a.s.l.)</th>
<th>Latitude (°N)</th>
<th>Lake area (km$^2$)</th>
<th>Mean depth (m)</th>
<th>Catchment (km$^2$)</th>
<th>Precipitation (mm/a)</th>
<th>Retention time (years)</th>
<th>pH</th>
</tr>
</thead>
<tbody>
<tr>
<td>Iso Valkjärvi, Finland</td>
<td>126</td>
<td>61</td>
<td>0.042</td>
<td>3.1</td>
<td>0.168</td>
<td>600</td>
<td>3</td>
<td>5.1</td>
</tr>
<tr>
<td>Bracciano, Italy</td>
<td>164</td>
<td>42</td>
<td>57</td>
<td>89.5</td>
<td>91.2</td>
<td>900</td>
<td>137</td>
<td>8.5</td>
</tr>
<tr>
<td>Øvre Heimdalsvatn, Norway</td>
<td>1090</td>
<td>61</td>
<td>0.78</td>
<td>4.7</td>
<td>23.4</td>
<td>800</td>
<td>0.17</td>
<td>6.8</td>
</tr>
<tr>
<td>IJsselmeer, Netherlands</td>
<td>0</td>
<td>52</td>
<td>1147</td>
<td>4.3</td>
<td>114 700</td>
<td>750</td>
<td>0.41</td>
<td>8.5</td>
</tr>
<tr>
<td>Hillesjön, Sweden</td>
<td>10</td>
<td>61</td>
<td>1.6</td>
<td>1.7</td>
<td>19.2</td>
<td>650</td>
<td>0.36</td>
<td>7.3</td>
</tr>
<tr>
<td>Devoke Water, UK</td>
<td>233</td>
<td>54</td>
<td>0.34</td>
<td>4.0</td>
<td>3.06</td>
<td>1840</td>
<td>0.24</td>
<td>6.5</td>
</tr>
<tr>
<td>Esthwaite Water, UK</td>
<td>66</td>
<td>54</td>
<td>1</td>
<td>6.4</td>
<td>14.0</td>
<td>1800</td>
<td>0.19</td>
<td>8.0</td>
</tr>
<tr>
<td>Lake</td>
<td>Potassium production (mg/L)</td>
<td>Primary production rate (g C·m⁻²·a⁻¹)</td>
<td>Suspended load (mg/L)</td>
<td>Sedimentation rate (g·m⁻²·a⁻¹)</td>
<td>Deposition of $^{137}$Cs (kBq/m²)</td>
<td>Fish</td>
<td>Max. $^{137}$Cs (Bq/kg ww)</td>
<td></td>
</tr>
<tr>
<td>---------------------</td>
<td>-----------------------------</td>
<td>----------------------------------------</td>
<td>-----------------------</td>
<td>---------------------------------</td>
<td>-----------------------------------</td>
<td>-------------------------------</td>
<td>---------------------------</td>
<td></td>
</tr>
<tr>
<td>Iso Valkjärvi</td>
<td>0.4</td>
<td>25</td>
<td>0.5</td>
<td>70</td>
<td>70</td>
<td>Whitefish &amp; perch</td>
<td>11650</td>
<td></td>
</tr>
<tr>
<td>Bracciano</td>
<td>40</td>
<td>0.8</td>
<td>0.5</td>
<td>70</td>
<td>0.9</td>
<td>Whitefish</td>
<td>14</td>
<td></td>
</tr>
<tr>
<td>Øvre Heimdalsvatn</td>
<td>0.4</td>
<td>27</td>
<td>0.3</td>
<td>60</td>
<td>130</td>
<td>Minnows &amp; trout</td>
<td>5250</td>
<td></td>
</tr>
<tr>
<td>IJsselmeer</td>
<td>7</td>
<td>350</td>
<td>40</td>
<td>500</td>
<td>2.2</td>
<td>Smelt, roach &amp; perch</td>
<td>21</td>
<td></td>
</tr>
<tr>
<td>Hillesjön</td>
<td>3</td>
<td>100</td>
<td>5</td>
<td>0</td>
<td>100</td>
<td>Roach &amp; perch</td>
<td>4750</td>
<td></td>
</tr>
<tr>
<td>Devoke Water</td>
<td>0.55</td>
<td>0.5</td>
<td>300</td>
<td>17</td>
<td>2</td>
<td>Perch &amp; trout</td>
<td>1750</td>
<td></td>
</tr>
<tr>
<td>Esthwaite Water</td>
<td>0.9</td>
<td>350</td>
<td>1</td>
<td>700</td>
<td>2</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* All lakes are oligohumic, except Iso Valkjärvi and Hillesjön, which are mesohumic. IJsselmeer, Hillesjön Water and Esthwaite Water are eutrophic, and the remainder are oligotrophic.
The seasonal variability moderator for epilimnetic and hypolimnetic temperatures, which influence stratification and internal loading of radiocaesium from sediments induced by the redox potential. This technique may also be used to define the active volume, i.e. the volume that participates in active transport and mixing of the lake water. The active volume is a small part of the entire volume during stratified conditions. This means that the retention rate may be given as \( Q(t)/V(t) \).

A sub-model to calculate the relationships between biomasses in lakes of different trophic character. The empirical calibrations for the VAMP lakes indicate that the transfer coefficient (phytoplankton biomass/prey biomass) may be given by:

\[
\text{Transfer\_coefficient} = (\text{Prim\_production} + 1)^{0.6}
\]

The primary production is in g C \( \cdot \) m\(^{-2}\) \( \cdot \) a\(^{-1}\). The choice of the transfer coefficient (TC\(_i\)) is important for the radiocaesium concentration of predatory fish because that concentration is calculated as the amount of \( ^{137}\text{Cs} \) per predatory biomass, and the predatory biomass is calculated from the ratio plankton\_biomass/transfer\_coefficient.

This definition of TC\(_i\) gives a value of about 1/7.5 for an oligotrophic lake with a primary production of 25-30 g C \( \cdot \) m\(^{-2}\) \( \cdot \) a\(^{-1}\), about 1/34 for a very productive lake (e.g. Lake IJsselmeer) with a primary production of 350 g C \( \cdot \) m\(^{-2}\) \( \cdot \) a\(^{-1}\), and about 1/1.5 for a lake with an extremely low production (the Italian Lake Bracciano).

The outflow rate function for the transport of radiocaesium from the catchment to the lake. This is a time dependent function which gives a reduced transport of radiocaesium from land to lake with time after the fallout event. The function is given by:

\[
\text{Outflow\_rate\_function} = (0.04/12) \text{Seasonal\_moderator}/(\text{time}_{FM} + 1)^{0.5}
\]

where FM is the fallout month (5 indicates May). The seasonal moderator gives an increased transport of radiocaesium from land to lake during spring and autumn peaks. The rate decreases with time from the month of the fallout (given by \( 1/\sqrt{\text{time}} \)). A delay function triggers the run-off to the month of the fallout plus one month (\( \text{time}_{FM} + 1 \)). The seasonal moderator influences the seasonal variability in the outflow rate.

The dimensionless moderator for planktonic uptake of radiocaesium. This moderator sets the bio-uptake as a function of the potassium (K) concentration of the water and of the pH. It has been derived empirically, using the data from the VAMP lakes. The moderator (Mod) operates on the phytoplankton uptake rate, which is set at 0.005 (1/month) as a default value. If this value is higher, the biological uptake (\( = 0.005 \times \text{Mod} \)) increases. The moderator is given by:

\[
\text{Mod} = 2/((\text{pH} + K) - 3)^2 + 0.01
\]
For a very acid lake, e.g. when the pH is 5 and the potassium concentration is 0.5 mg/L, the moderator is 0.33. For a lake with very hard water (pH = 7.5, K = 40 mg/L), the moderator is 0.011. This means that the bio-uptake rate of radiocaesium from the dissolved phase in the water is 30 times lower (0.33/0.011 = 30) in such a hard-water lake compared to a soft-water lake.

(7) The algorithm for the lake partition coefficient ($K_d$). The $K_d$ value depends on the pH. The distribution (partition or partitioning) coefficient $K_d$ is used to differentiate the caesium in the lake water into a particulate phase and a dissolved phase. The lower the pH, the more radiocaesium is in the dissolved phase. The pH could influence both the affinity of radiocaesium to the carrier particles (humus, clays, etc.) and the aggregation of the carrier particles and hence the sedimentation rate. The following algorithm (a dimensionless moderator) has been derived for lakes with pH values between 4 and 9:

$$K_d = \frac{1}{(1.04 + (1.75 \times \left(\frac{pH}{4}\right) - 1)^2)}$$

Thus, 96% of the radiocaesium in the lake water is in the dissolved phase at pH = 4 and 26% is at pH = 9.

(8) The sub-model for biological half-lives of radiocaesium in lake fish, which depend on temperature and fish weight:

$$RR = \frac{0.693}{(0.5 \times BHL)} = 0.136 \times W^{0.139} \times e^{-temp \times 0.0693}$$

where RR is the retention rate (RR = 1/BHL), BHL is the biological half-life (in months), W is the mean weight (g) of fish in the population, and temp is the mean annual lake temperature (°C); the cooler the water, the longer is the half-life of radiocaesium.

(9) The approach to quantifying internal loading, based on the lake dynamic ratio:

Internal loading = Amount_in_active_sediments 0.05 DR

where 0.05 is the transport rate for sediment to water (1/month) and DR is the dynamic ratio, given by:

$$DR = \sqrt{\text{area}}/D_m$$

where area is the lake area (in $m^2$) and $D_m$ is the mean depth (m).

3. DEFINING PREDICTIVE POWER

One way of quantifying the fit between modelled values and empirical data is to make a regression. If the model predicts well, the $r^2$ value and the slope should be as close to unity as possible and the intercept should be at the origin. One can
safely assume that the $r^2$ value will never be equal to one. There will, however, be situations when the model will yield a poor prediction, a low $r^2$ and a slope that is far from one. For each validation, one can determine $r^2$ and slope, and the coefficient of variation (CV = SD/MV) for the calculated $r^2$ values. If the model has a high predictive power (PP), then CV should be small. A general definition of PP is:

$$PP = \frac{R^2}{((1.1 - \alpha) \cdot CV)}$$

where $R^2$ is the mean $r^2$ of all model validations: the higher $R^2$, the higher is PP. One could instead use the median $r^2$ value. This is a matter of definition. The slope, $\alpha$, of the regression line may be smaller or larger than 1. If the slope is smaller than 1, one can quantify the influence on PP by means of the factor $1.1 - \alpha$. Since $\alpha$ may equal 1 and since division by zero is impossible, this approach uses 1.1 instead of just 1. Constants other than 1.1 may be used, but 1.1 will cause the PP values to vary between 0 and 100 (Fig. 1). This means that the slope factor is always larger than 0.1. If the slope is larger than 1, one simply sets $\alpha = 1/slope$, and uses the same factor. This means that a slope of 0.5 will give the same factor as a slope of 2,

![FIG. 1. Nomograms illustrating predictive power: (a) curves for a constant CV of 0.1, and (b) curves for a constant slope of 0.8.](image-url)
namely 1.1 - 0.5 = 0.6 or 1.1 - 1/2 = 0.6. One could also account for the intercept in this approach, but that would add very little, since the slope and the $r^2$ value are already used. CV is the coefficient of variation for the $r^2$ values obtained in the validations. It is a measure of model uncertainty. Similar CV values may be determined in other ways, e.g. by Monte Carlo simulations.

Figure 1 shows two nomograms illustrating how $R^2$, slope and CV influence PP. Using this definition, PP will generally be lower than 100. Models yielding PP above 10 would be very good. Models giving PP below 1 would be useless for all practical purposes. Such models have a poor fit and great uncertainties.

This expression of predictive power should be regarded as a tool accounting not for every conceivable factor but for the most important factors in a simple and useful manner. All expressions related to $r^2$ would depend on the number of data pairs (n), and on the range and the transformation of the x- and y-variables.

It should be noted that the uncertainty (CV) is determined independently of the fit. One should not use, e.g., expressions related to the confidence interval of the regression line for the uncertainty, since such measures are directly related to $r^2$. In this approach, model uncertainty is expressed in two ways, using either Monte Carlo simulations or repeated validations.

4. PREDICTIVE POWER OF DYNAMIC MODELS

Three dynamic models for radiocaesium in lakes have been tested:

(i) A small, mixed (dynamic and empirical) model. This model has only three compartments: water, prey and predatory fish; it has six model variables and five lake specific variables. The total number of driving variables (x) is thus 11. Note that there is no catchment area, no sediments, no food-web and no partition coefficient ($K_d$) in this model.

(ii) The VAMP model is presented in Fig. 2. It has 10 compartments, 21 model variables and 13 lake specific variables. The model size is given by $n = 34$.

(iii) The generic model, a traditional model with 9 compartments, 27 model variables and 9 lake specific variables, which gives $n = 36$.

These models have been tested using the data for the VAMP lakes (Table I). The results are summarized in Table II and in Fig. 3.

There are large uncertainties in some of the empirical data. In this project, the uncertainty of all time series of data for radiocaesium concentrations in water and fish were handled in a special way to facilitate comparisons between modelled values and empirical data. First, the data for a given time series in one column were copied and then pasted into a new column so that data for April were compared with data from March, etc. By doing so, we obtained two empirical sets of data, Emp1 and
The VAMP model

Panel of driving variables

Model variables

Lake-specific variables
Emp2. Ideally, they should be identical. Note, however, (1) that all data from the first half year after the Chernobyl accident (i.e. data up to September 1986) were excluded in this test, since the conditions during this period were very variable, and significant changes took place, especially in lake water and in plankton eating fish, from one month to the next; and (2) that, in order to obtain enough data for this comparison, we accepted for 1986 and 1987 only data from two consecutive months, but, for 1988, we accepted data taken two months apart (but not more); and, for 1989 and the following years, we accepted data taken three months apart (but not more). This gives data for a very interesting comparison. One cannot expect that models would yield better correlations with the empirical data than those obtained from this comparison between Emp1 and Emp2.

Table II gives a compilation of many model runs for the VAMP model, for the seven VAMP lakes and for radiocaesium concentrations in water, whitefish, trout, small perch, large perch, roach and pike. The $r^2$ values are given when empirical data are compared with modelled values. Note that there is a large variation in the $r^2$ values for the two empirical data sets, Emp1 and Emp2, from 0 to 0.99. This indicates the uncertainty in some of the empirical data.

Another interesting result is given in Table II. There is something 'strange' in Lake Bracciano. The correspondence between the two empirical sets of data is very good, but the VAMP model prediction is poor ($r^2 = 0.31$ for water and $r^2 = 0.41$ for whitefish). The reason for this is, in fact, that this lake has high concentrations of 'old' radiocaesium. The VAMP model assumes that all radiocaesium emanates from the Chernobyl fallout, and there is no factor accounting for old radiocaesium from weapons tests during the 1950s and 1960s. Therefore, this does not illustrate a weakness of the VAMP model, but its strength. The model provides very good to adequate predictions for most lakes, for water and most species of fish, and when this is not the case, then the difference between the model prediction and the empirical value could be discussed in quantitative terms. In the case of Lake Bracciano, it is evident that a model which does not account for old radiocaesium should provide poor predictions. If the VAMP model were to give good predictions, it would be for the wrong reason!

The VAMP model provides poor predictions also for small perch ($r^2 = 0.01$) and large perch ($r^2 = 0.08$) for Devoke Water. Is this due to deficiencies in the

---

FIG. 2. VAMP model for radiocaesium in lakes.
Sub-models for biomass and the seasonal moderators for Q (and T), and temperature, and the panel of driving variables are also shown. This panel is divided into two parts: model variables and lake specific variables. The environmental variables change for every lake, but, ideally, the model variables do not change, unless there are excellent reasons to do so. This conservative rigour about ad hoc adjustments minimizes the ‘art’ and maximizes the science in building predictive models.
HÅKANSON et al.

**TABLE II. EMPIRICAL $r^2$ VALUES**

<table>
<thead>
<tr>
<th>Lake</th>
<th>VAMP $r^2$</th>
<th>Emp1 versus Emp2 $r^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>Bracciano</td>
<td>0.31</td>
</tr>
<tr>
<td>Whitefish</td>
<td>Bracciano</td>
<td>0.41</td>
</tr>
<tr>
<td>Water</td>
<td>Iso Valkjärvi</td>
<td>0.84</td>
</tr>
<tr>
<td>Water</td>
<td>IJsselmeer</td>
<td>0.89</td>
</tr>
<tr>
<td>Water</td>
<td>Esthwaite Water</td>
<td>0.66</td>
</tr>
<tr>
<td>Water</td>
<td>Devoke Water</td>
<td>0.79</td>
</tr>
<tr>
<td>Water</td>
<td>Hillesjön</td>
<td>0.52</td>
</tr>
<tr>
<td>Whitefish</td>
<td>Iso Valkjärvi</td>
<td>0.89</td>
</tr>
<tr>
<td>Trout</td>
<td>Heimdalsvatn</td>
<td>0.86</td>
</tr>
<tr>
<td>Trout</td>
<td>Devoke Water</td>
<td>0.39</td>
</tr>
<tr>
<td>Smelt</td>
<td>IJsselmeer</td>
<td>0.81</td>
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<td>Small perch</td>
<td>Hillesjön</td>
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<td>Small perch</td>
<td>IJsselmeer</td>
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</tr>
<tr>
<td>Small perch</td>
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<td>Roach</td>
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<td>Roach</td>
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<td>Pike</td>
<td>Hillesjön</td>
<td>0.94</td>
</tr>
<tr>
<td>Pike</td>
<td>Iso Valkjärvi</td>
<td>0.69</td>
</tr>
<tr>
<td>Large perch</td>
<td>Iso Valkjärvi</td>
<td>0.98</td>
</tr>
<tr>
<td>Large perch</td>
<td>Hillesjön</td>
<td>0.55</td>
</tr>
<tr>
<td>Large perch</td>
<td>IJsselmeer</td>
<td>0.30</td>
</tr>
<tr>
<td>Large perch</td>
<td>Devoke Water</td>
<td>0.08</td>
</tr>
</tbody>
</table>

*The $r^2$ values have been obtained from comparisons between empirical and modelled $y$ for the VAMP model, and for the test series from comparisons between the two empirical samples (Emp1 versus Emp2) for different lakes, and different concentrations of radio-caesium in water and in different species of fish.*
From Table II, it can be seen that the answer to that question is "no", since there is also an equally poor correspondence between the two empirical data sets (small perch, \( r^2 = 0.02 \); large perch, \( r^2 = 0 \)). It is evident that these empirical data are uncertain and that they are a poor basis for model validations.

The table in Fig. 3 gives mean \( r^2 \) values, CV related to the \( r^2 \) values, mean slope and PP for the three models, and the corresponding data for the comparison between the two empirical samples (Emp1 versus Emp2). Note that the smallest model, the mixed model, yields the highest PP; the largest model, the generic model, yields the lowest PP. So, in this example, the best PP is obtained for the smallest model, accounting for the most important processes, no more and no less. One can also see that the lowest PP value is obtained when the two empirical samples are compared. This indicates the empirical uncertainties associated with some of the VAMP data.

![Graph showing predictive power vs model size](image-url)
5. USE OF THE SUB-MODELS

The sub-models for prediction of seasonal variability for water discharge (Q) and theoretical water retention time (T), lake water retention rate, and surface and bottom water temperatures applied in the VAMP model may be used in different contexts. For example, they can be used to reconstruct or to predict the seasonal pattern of lake variables when only annual or super-annual means are given. Such annual mean values may in turn be predicted from simple characteristics of the catchment or the lake [2].

It is of the utmost importance that the VAMP model accounts for seasonal differences, since the next accident cannot be expected to happen at the same time of year and under the same weather conditions. Figure 4 shows the concentrations of radiocaesium in trout in Lake Øvre Heimdalsvatn which would have been observed if the accident had happened in another season. The simulation is therefore a sensitivity analysis of the VAMP model, with (Fig. 4(a)) and without (Fig. 4(b)) the seasonal moderator for Q and T, where the month of the fallout changes, while everything else remains the same. The question of interest is whether the timing of the event affects the peak value and the shape of the recovery ‘tail’.

The model indicates that May — the very beginning of the growing season — was one of the worst possible months for an accident of this type. The peak values would have been lower if the accident had occurred in January (curve 1), in March (curve 3) or in late autumn. In winter, a significant part of the fallout would have quickly escaped from the lake in the spring flood. In late summer and autumn, the plankton would have accumulated less fallout than it did just after the spring flood.

The predicted peak value of radiocaesium for trout with the seasonal moderator is 5300 Bq/kg wet weight (ww); without the moderator it is 2900 Bq/kg ww. The empirical peak value is 4600 Bq/kg. The timing of the peak is little affected by the presence or absence of the moderator for Q and T. However, when the month of the fallout is changed, the model without this seasonal variability moderator simply transposes the same curve in simple steps, depending on the month of the fallout. This does not seem to be very probable. These predictions concerning different fallout months have not been validated, and one hopes they will never be tested.

6. EMPIRICAL TESTS

Since the seven VAMP lakes vary in most lake characteristics (Table I), it is a great challenge to model the effects of the Chernobyl ‘spike’ in water and biota. Figure 5 shows the very close correspondence between the empirical data and the model values for trout (Fig. 5(a)) and minnows (Fig. 5(b)) in Lake Øvre Heimdalsvatn, in model simulations with and without the seasonal moderator for Q and T. Figure 6 shows the even more remarkable result that the $r^2$ value for measured and
predicted peak values is 0.996! All lakes lie almost directly on the regression line, the slope is about 1, i.e. close to the ideal $y = x$, and the 95% confidence interval for the predicted $y$ value is close to the regression line. The Norwegian lake is the worst case.

It must be pointed out that these fine results are not derived from blind tests. The model required a considerable amount of ‘tuning’ before it behaved so well. But — and this is important — this tuning did not involve the model variables illustrated in Fig. 2; it involved only the lake specific, environmental variables. Consequently, the results are really very good, for both the peak and the ‘tail’.

FIG. 4. Sensitivity analyses of the effect of the timing of the acute fallout dose on radio-cesium concentrations in predatory fish (trout) for Lake Øvre Heimdalsvatn, predicted using the VAMP model, (a) with and (b) without the seasonal variability moderators for $Q$ and $T$. Curve 1 gives the results for an acute deposition of 130 kBq/m$^2$ in January, curve 3 represents the results for March, etc.
FIG. 5. Comparison of the observed levels of radiocaesium in (a) trout and (b) minnows in Lake Øvre Heimdalsvatn with those predicted by the VAMP model with and without the seasonal variability moderators for $Q$ and $T$.

7. CONCLUSIONS

The experience gained in the VAMP project demonstrates clearly that dynamic models should always be calibrated against reliable empirical data, that it is very important to validate models against independent data sets, and that it is important to control the representativity of the empirical data. It is also always important to make sensitivity analyses, by varying one variable or parameter within given, reasonable or unreasonable, limits while keeping the others constant. It is also important to conduct uncertainty analyses in cases where several related variables are varied simultaneously.
Different types of sub-models could be connected to form a large ecosystem model, but that large model would compound the weaknesses of the sub-models and might even reveal new problems that are not apparent in the sub-models. Large ecosystem models are needed to study complex relationships in dynamic ecosystems. But a model is a model, not the real world. Models without biological inputs, such as physical and chemical models, can often be constructed entirely 'scientifically', by logical deductions and causal analyses. Models that include biological variables and interactions among biotic and abiotic variables on the ecosystem level are not so easily built! Every model, even the largest conceivable ecosystem model, is a gross simplification.

REFERENCES

MODELLING OF RADIONUCLIDE TRANSFER IN RIVERS AND RESERVOIRS

Validation study performed within the IAEA/CEC VAMP programme

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MODELLING OF RADIONUCLIDE TRANSFER IN RIVERS AND RESERVOIRS: VALIDATION STUDY PERFORMED WITHIN THE IAEA/CEC VAMP PROGRAMME.

The comprehensive data obtained for different rivers after the Chernobyl accident, primarily the data from the heavily contaminated Pripyat river-Dnieper river-reservoir system, provide a unique basis for validations and improvements of transport models for aquatic radionuclides. These data were used in two scenarios by the River Sub-Group of the Aquatic Working Group of the IAEA/CEC co-ordinated research programme Validation of Environmental Model Predictions (VAMP). For one model validation scenario, the Clinch River-Tennessee River system, contaminated by releases from the Oak Ridge National Laboratory, was chosen. The data collected during the post-Chernobyl period, related to the Pripyat river and to the Dnieper river and reservoirs, were used in the other scenario. Different kinds of models were applied, and the data from the two scenarios were used to simulate the dynamics of radionuclides in water, suspended sediments and bottom sediments. Analytical models, two kinds of box models and two kinds of one-dimensional (cross-sectionally averaged) models were used for the simulation of radionuclide concentrations in water and sediments. These models represent different levels of model complexity for the description of interactions between radionuclides, suspended sediments and sediments deposited on the bottom layer. The vertical profiles of radionuclides in bottom sediments were simulated for the Tennessee River scenario by the multilayer bottom model and by two kinds of vertical diffusion models. For this scenario, the modelling of radionuclide distribution in the aquatic food-chain, mainly in fish, was based on static approaches. For the other scenario, the temporal changes in $^{137}$Cs concentrations in bream, trench and Carpio carpius in the Kiev reservoir (Dnieper river) were simulated by the dynamic model. The doses to the population via aquatic pathways were calculated on the basis of simulated concentrations in an environmental compartment and were used in analyses of the role of aquatic pathways in the formation of the total radiation dose after large releases.

1. INTRODUCTION

Studies of the environmental impact of radionuclide releases demonstrate that, after the initial fallout, large river systems are the main pathways for radionuclide transport from the point of discharge to distances of hundreds to thousands of kilometres. Some of these modelling methods have been reviewed by the IAEA [1] and by different authors (see, e.g., Refs [2, 3]). The spread of computer technology
during the last decade and the urgent need to increase the predictability of models in order to provide adequate information for decision making concerning remedial measures in the most contaminated water bodies after the Chernobyl accident have led to an intensive development of river–reservoir modelling. The comprehensive data obtained for different river–reservoir systems after the Chernobyl accident provide an excellent basis for model validation and improvement. To provide a mechanism for validation of assessment models by using environmental data on transfer of radionuclides from the Chernobyl accident was the main aim of the IAEA/CEC co-ordinated research programme Validation of Environmental Model Predictions (VAMP) [4], which was established in 1988 and finished in 1994. In the framework of the VAMP Aquatic Working Group, a Sub-Group for Rivers and Reservoirs was established in 1992 [5]. This paper gives a brief description of its activities.

2. SCENARIOS

The data collected during the post-Chernobyl period relating to the Pripyat river and the Dnieper river and reservoirs, for water, fish and irrigated products, were used in scenario R2, prepared by the Ukrainian Hydrometeorological Institute (UHI), Kiev. The Clinch River–Tennessee River system, contaminated by releases from Oak Ridge National Laboratory, was chosen for the model validation scenario R1 (developed by SENES Oak Ridge Inc.) for a comparison of the predictability of models in different hydrological and physico-chemical conditions. The validation study could not be done on the basis of direct dose measurements; however, inter-calibration of the dose simulation approaches could be used for analysing the role of aquatic pathways in the formation of the total radiation dose after large releases.

2.1. Clinch River–Tennessee River (Scenario R1)

The Oak Ridge National Laboratory, located in the south-eastern part of the United States of America, has discharged radionuclides into the White Oak Creek since 1943. Significant releases took place from 1949 to 1968. Maximum annual amounts of selected radionuclides were discharged in the following years: $^{137}$Cs: 6290 GBq in 1956; $^{106}$Ru: 74 000 GBq in 1961; $^{90}$Sr: 5550 GBq in 1949; $^{60}$Co: 2849 GBq in 1956. The White Oak Dam, which is located 1 km upstream of the mouth of the creek, controls the release of radionuclides into the Clinch River at kilometre CRK 33.5. The Clinch River joins the Tennessee River at kilometre TRK 913.4. The Clinch River–Tennessee River system is a highly regulated river–reservoir system (Fig.1) that is used for hydroelectric power generation, flood control, navigation and recreation.
For model testing, the following tasks for scenario R1 have been proposed [5]:

1. Given the concentrations of $^{137}$Cs and $^{90}$Sr in water at the White Oak Dam, to predict the monthly concentrations of these radionuclides from December 1960 until November 1962 at the following locations: Centers Ferry, CRK 8.8; Watts Bar Dam, TRK 529.9; and Chickamauga Dam, TRK 471.0.

2. To predict the profile of a sediment core taken from the Tennessee River just below its confluence with the Clinch River at TRK 567.5, and to provide the concentration of $^{137}$Cs for each 4 cm section of the core to a depth of 96 cm, representing the years 1949 to 1986.

3. To predict the annual average concentrations of $^{137}$Cs, $^{106}$Ru and $^{90}$Sr in the water column and in fish in the Clinch River at CRK 23.3 and in the Tennessee River at TRK 758.0 for 1961-1963. Using the predictions for these two loca-
tions, to estimate the radiation dose to an individual from the aquatic pathway, i.e. from swimming in the sea and consumption of fish.

2.2. Dnieper reservoirs (scenario R2)

The watershed of the Dnieper river in Ukraine, Russia and Belarus was heavily contaminated by radionuclides accidentally released from reactor Unit No.4 of the Chernobyl nuclear power plant (NPP) in Ukraine on 26 April 1986. The Chernobyl NPP is located on the west bank of the Pripyat river, approximately 30 km from its mouth at the Kiev reservoir on the Dnieper river. A significant portion of the radionuclides released were deposited on the watershed of the Pripyat river in Ukraine and Belarus. Other areas that were heavily contaminated by $^{137}$Cs are in the upper Dnieper watershed in Russia and Belarus. As a result of this surface contamination, there was a long term influx of $^{137}$Cs and $^{90}$Sr into the Dnieper river, which passes through reservoirs before discharging into the Black Sea (Fig. 2). The modern Dnieper river system downstream of the mouth of the Pripyat river includes six large reservoirs: the Kiev, Kanev, Kremenchug, Dnieprodzerzhinsk, Zaporozhie and Kakhovka reservoirs. The average capacity of the reservoirs varies from $2.45 \text{ km}^3$ (Dnieprodzerzhinsk reservoir) to $18.2 \text{ km}^3$ (Kakhovka reservoir). The population of the Dnieper basin region is 32.5 million. Water from the Dnieper river that is used in the municipal tap water supply is consumed by more than 8.1 million people in ten regions and in the Crimean Republic. The Dnieper reservoirs are also used for commercial fishing. Water from the Dnieper river is used for irrigation of more than 1.8 million hectares in Ukraine, for 1.3 million hectares of which (72%) water from the Kakhovka reservoir is used.

The modelling scenario was prepared on the basis of monthly averaged data from May to December 1986 and ten-day averaged data during 1987–1993 on water discharge, suspended sediment discharge and influx of radionuclides ($^{137}$Cs and $^{90}$Sr) in solute and suspended sediments into the Kiev reservoir through cross-sections of the Pripyat river at Chernobyl and the Dnieper river near the village of Nedanchichi (points 1 and 2, respectively, in Fig. 2) [5, 6]. To validate the models of water, sediment and fish contamination, the $^{137}$Cs and $^{90}$Sr data for the Dnieper reservoir were used as a starting point for modelling (1 January 1987). The initial version of this scenario includes data on radionuclide fallout on the surface of the Kiev reservoir and estimated data on the monthly averaged concentrations of $^{137}$Cs and $^{90}$Sr in the Dnieper tributaries in the year 1986. It was proposed that the starting point of modelling should be May 1986. However, later, more detailed analyses of the situation were performed, which demonstrated that the uncertainties in the fallout data and especially in the data on the contamination of the Pripyat river in 1986 were so large that they had an impact on the water transport models and deformed the simulated results. Therefore, the data for the period until 1 January 1987 were made available for modelling.
FIG. 2. Schematic diagram of the Dnieper reservoirs.
The main tasks for Scenario R2 are as follows:

1. To predict the monthly and annual average concentrations of $^{137}\text{Cs}$ and $^{90}\text{Sr}$ in water, suspended particles and bottom sediments of all six Dnieper reservoirs over the period 1987–1993; and to provide uncertainty estimates for the predictions. The 1986 data were used as a starting point for the simulations.

2. To estimate the $^{137}\text{Cs}$ concentrations in commercial species of fish during the post-accident period.

3. To estimate the radiation dose for individuals and for the population due to consumption of drinking water and fish, and of products from areas irrigated with water from the Dnieper reservoirs.

3. MODELLING OF RADIONUCLIDE CONCENTRATIONS IN WATER, SEDIMENTS AND FISH

Modelling of radionuclide dispersion in rivers and reservoirs has some peculiarities compared with modelling of dispersion in lakes. The radionuclide dispersion in rivers and reservoirs is affected by different flow velocities, short retention times and large variability in water discharge during the year. As a result, there are strong temporal variations in the sedimentation–resuspension rates. Suspended and bottom sediments interact with water, whereby radionuclides are exchanged by physical, chemical and biological processes.

The main physical exchange mechanisms are sedimentation of contaminated suspended matter in the river bed and resuspension of the bottom sediment. These mechanisms are controlled by hydraulic factors (e.g. river flow and sediment transport) and are strongly dependent on the sediment size fractions (e.g. clay, silt, sand, gravel). Radionuclide diffusion through interstitial water is the physical process that accounts for migration phenomena which are not related to sediment transport. Adsorption and desorption of radionuclides by surface bed sediments are the main chemical processes. They are not always completely reversible and are controlled by geochemical reactions of dissolved radionuclides with sediment. Uptake and subsequent excretion of radionuclides by aquatic biota and, in general, perturbation of the sediment due to the action of living organisms represent biological processes that are responsible for the exchange of radionuclides between water and bottom sediment. Different levels of complexity of transfer processes of radionuclides between the water phase and the solid phase have been described and different averaging scale models have been used in the validation study.

The following models have been used:

(a) The analytical model describing contamination in water and sediments (AI, HC [5]);
(b) A box-type (compartmentally averaged) model which does not simulate the suspended sediment dynamics, but which accounts for three bottom layers: the first, thin top layer in which radionuclides are in chemical equilibrium with the overlying water; the second layer (just below the first one) in which radionuclides mix with overlying water through the top layer; and the deep layer which acts as an ultimate radionuclide sink [7];

(c) Box-type and one-dimensional (cross-sectionally averaged) models simulating the dynamics of the concentrations of suspended sediments of one dominant grain size, and the concentrations of radionuclides in dilution, on suspended sediments and in the upper part of the bottom deposition layer (CC-WATOX and CC-RIVTOX models [8]);

(d) A one-dimensional model describing the dynamics of the concentrations of sand, silt and clay in water and simulating the radionuclide dynamics for each of these three typical grain sizes (PNL-TODAM model [3]).

The AI model, the analytical HC model and the RIVTOX and TODAM models were applied on the basis of data on releases from Oak Ridge and hydrological data for the Clinch River-Tennessee River scenario, to simulate radionuclide concentrations in water and in the upper part of the bottom deposition layer in the Clinch River and the Tennessee River. All models except HC were used for the Dnieper river scenario.

Some results of simulations of the $^{137}$Cs and $^{90}$Sr concentrations for scenario R2 are presented in Figs 3 and 4. The distributions were analysed for each of the listed models for each of the six Dnieper reservoirs. The simulated data for scenario R1 were validated with measured data averaged over a much larger time-scale. Sensitivity and uncertainty analyses were performed for ENEA and for the CC-WATOX and CC-RIVTOX models on the basis of data for scenario R2.

The vertical profiles of $^{137}$Cs in bottom sediments in the Clinch River-Tennessee River system were simulated with the TODAM model using a multiple bed layer approximation; with the TYPHOON DIFF model [9], which explicitly accounts for exchangeable and non-exchangeable processes of radionuclide sorption by sediments and which solves two sets of one-dimensional mass balance equations for the exchangeable and non-exchangeable radionuclide components; and with the CC vertical diffusion model BOTOX, which describes only one component — the total $^{137}$Cs concentration in interstitial water [5]. It was demonstrated that estimates of the key model parameter $K_d$ — the distribution coefficient for $^{137}$Cs — using data from the literature and from the two scenarios, allowed good agreement to be achieved between calculated and experimental results. The vertical distribution of $^{137}$Cs in bottom sediments under non-uniform entry of radionuclides into the river is determined by the type of the relationship between sedimentation rate and time. Thus, in this case, the use of complicated physical and chemical models of vertical migration does not lead to significant improvement in predictions.
FIG. 3. Concentrations of $^{137}$Cs in the Kiev reservoir (scenario R2).

FIG. 4. Concentrations of $^{90}$Sr in the Kakhovka reservoir (scenario R2).
Modelling of radionuclide distribution in the aquatic food-chain, mainly fish (tasks of scenario RI), was based on static approaches, which are characterized by mathematical relations independent of time, using the concentration factor (HC, AI and CC models). Comparison with measured data shows an overprediction of fish contamination, as a result of which the estimated dose to people would be too high. The dynamic model TRANSAQUA [10, 11], which describes the migration of radionuclides through the compartments of the food-chain using 'transfer rates', was applied to simulations of temporal changes in $^{137}$Cs concentrations in bream and roach in the Kiev and Kakhovka reservoirs. On the basis of annual comparisons, TRANSAQUA gives quite good results, only taking into account a simple pelagic food-chain.

4. DOSE CALCULATIONS

Doses from the aquatic pathways were calculated on the basis of modelled concentrations in an environmental compartment (by teams from AI, HC, UCRM, UHI and CC) in order to analyse the role of aquatic pathways in the formation of the total radiation dose after large releases. Pathways such as consumption of drinking water and fish, irrigation, swimming in the sea and external irradiation on beaches were considered for both scenarios.

The assessment of the dose to the population via Dnieper water intake is not of purely scientific interest but is also a problem of great practical significance. Dose calculations were made on the basis of experimental data for 1986–1993 (which could be simulated with reasonable accuracy, as demonstrated [5]) and on the basis of simulations of $^{90}$Sr concentrations in water, obtained with the WATOX model for stochastically simulated hydrological data until the year 2056. The dose calculations were done at UCRM, using input data from UHI, HC and CC. The radiation dose which the public could have potentially received as a result of intake of radionuclides from Dnieper water was calculated in terms of the annual committed effective dose (ACED, in Sievert) — the dose resulting from intake over one year, committed to age 70; and the collective cumulative (for 70 years) committed effective dose (CCCED$_{70}$, in man-Sievert) — the age dependent ACED, integrated over 70 years of intake and over the age structure of the population. A very important characteristic of dose formation is the ACED to the maximally exposed member of the general public (MEMGP). This dose was calculated by assuming an 'average' diet for a particular region, which includes only products from irrigated land and tap water from the Dnieper river. The consumption rate of Dnieper fish is determined by the commercial catch.

The basis of the software module, which simulates irrigation and radionuclide transport in food-chains, is the ECOSYS-87 radioecological model. The simulation of the metabolic processes in the human body under chronic intake of radionuclides
is based on the metabolic models of caesium and strontium from ICRP-56. Nuclear decay data from ORNL and the Specific Absorbed Fraction files of the ICRP recommendations (1990) are used. For dose calculation the specially adopted multipurpose computer software internal dosimetry support system (IDSS) [12] is used, which has been developed specifically for the needs of the ICRP.

In the simulation of long term processes of dose formation, only doses due to the intake of $^{137}$Cs and $^{90}$Sr were considered. The collective dose in the Kiev region in 1986 exceeded the current dose level by factors of five to seven. A diametrically opposite effect has been observed in the Crimean Republic. This is due to the low level of contamination of the water of the lower Dnieper river from primary fallout.

For 1986, the ACEDs to the MEMGP due to the use of water in the Kiev region are $1.7 \times 10^{-5}$ Sv and $2.7 \times 10^{-5}$ Sv for $^{90}$Sr and $^{137}$Cs, respectively. The assessible contributions of irrigation with Dnieper water, of its use in the municipal tap water supply and of fish consumption to the CCCED$_{70}$ are, respectively, 18%, 43%, 39% in the Kiev region, 8%, 25%, 67% in the Poltava region, and 50%, 50%, 0% in the Crimean Republic (no consumption of fish from the Dnieper river).

The predicted average contribution of $^{90}$Sr to the CCCED$_{70}$ to the population of the Dnieper regions resulting from the use of water is 80%. The CCCED$_{70}$ to the population of the Dnieper regions (32.5 million people) resulting from the use of water is about 3000 man-Sv.

5. CONCLUSIONS

The main output of the study was the practical validation of the applicability of different radionuclide dispersion models for simulating large scale processes in river-reservoir systems. It was demonstrated that even simple models could be tuned (calibrated) for the specific case studies. However, it was also shown that only models the structure of which reflects the main hydrological, physical and chemical processes are valid for different situations with different sets of generic parameters.

The modelling results confirm the important influence of water body processes such as reversible and non-reversible kinetics of radionuclide exchange in the system water-suspended sediments-bottom depositions, radionuclide transfer through algae as an amplification factor for oscillations of radionuclide concentrations during summer, and the governing role of sedimentation in the formation of bottom sediment profiles. The similar results obtained for different models with a more or less complicated structure (one-layer bottom or multilayer bottom, one or three grain sizes of typical suspended sediments) were helpful in finding a balance between the complexity of the models and their applicability.

The study has decreased the uncertainty range for the main model parameters (distribution coefficient, time-scales of the transfer rate) for $^{90}$Sr and $^{137}$Cs in large river systems.
The study has demonstrated the significant contribution of aquatic pathways connected with the use of water from the Dnieper river (mainly through drinking water and irrigation of land) to the doses received by the population of southern Ukraine from the Chernobyl accident. Taking into account that the $^{90}$Sr contamination of the Dnieper river continues to be a problem that is not only of scientific interest, a very important output of the study is the increasing predictive possibilities of modelling, which can be used for estimating the radioactive contamination of the Dnieper river water in cases of future high floods.

REFERENCES


Abstract

A TEST OF RESUSPENSION FACTOR MODELS AGAINST CHERNOBYL DATA.

After the accident at Unit 4 of the Chernobyl nuclear power plant (NPP), stationary air samplers were operated at Chernobyl and Baryshevka, cities which are 16 km and 150 km, respectively, from the NPP. Other air samplers were operated simultaneously, but intermittently, at locations within the 30 km zone at distances of 4–25 km from the NPP. These data were used to check the validity of time dependent models of the resuspension factor $K (m^{-1})$.

Seven different models were examined, three of which are discussed in the paper. Data from the stationary air samplers were averaged over one day or one month; data from the intermittent air samplers were averaged over three days in 1986 and over four hours in 1991. The concentrations of eight radionuclides were measured at ten points during the same time period (14-17 September 1986). The calculated resuspension factors range from $6 \times 10^{-9} m^{-1}$ to $3 \times 10^{-6} m^{-1}$. Data for the spatial means of $K$ are given for certain time periods in 1986 and 1991; also presented are the calculated values according to the models. The experimental data and the calculated values differ by up to more than one order of magnitude. Also analysed was the temporal change in experimental values of $K$ and these values were compared with model predictions. The annual means of the resuspension factor as determined experimentally and as calculated with the models are presented. The model derived from empirical data measured in Neuherberg after the Chernobyl accident agrees best with the data. The Garland model systematically gives results lower than the experimental values, and the calculated values of $K$ from the Linsley model are consistently conservative. Also considered were the uncertainty of $K$ due to fluctuations in air concentrations and possible biological effects of episodic exposures.
Several approaches are available for estimating secondary contamination of the surface boundary layer by resuspension of radioactive aerosols. In the present study, we use new data from Ukrainian scientists in an approach that is based on the resuspension factor as a function of time.

After the accident at Unit 4 of the Chernobyl nuclear power plant (NPP), stationary air samplers were operated at Chernobyl and Baryshevka, cities which are 16 km and 150 km, respectively, from the NPP [1]. Other air samplers were operated simultaneously, but intermittently, at locations within the 30 km zone, at distances of 4–25 km from the NPP [2]. The samplers, designed by the Scientific Production Association "Typhoon" (Institute of Experimental Meteorology, Obninsk, Russia), operated at a flow rate of 100,000 to 120,000 m$^3$/d, with a sampling height of about 1.5 m. The filter was of the Russian Petrynov type cloth FPP-15-1.5, with an area of 1.05 m$^2$. The exposed filters were pressed into tablets, ashed at 300–400°C and then analysed by gamma spectrometry. Data from the stationary samplers were averaged over a day or a month; data from the intermittent samplers were averaged over three days in 1986 and four hours in 1991. The data were used to check the validity of time dependent models of the resuspension factor $K$ (m$^{-1}$), defined as follows:

$$K (m^{-1}) = \frac{q \text{ (Bq/m}^3\text{)}}{c \text{ (Bq/m}^2\text{)}}$$  \hspace{1cm} (1)

where $q$ is the airborne concentration and $c$ is the surface concentration of deposited material.

The resuspension factor is useful for local contamination for characterizing the relationship between surface contamination and airborne contamination; however, its use implies an equilibrium between the resuspended aerosol and the deposited aerosol [3]. In practice, the contamination on the surface is not homogeneous and the airborne concentration of radioactivity is a sum of local resuspended contamination and contamination that is advected from upwind sources of resuspension [3]. The airborne concentration and the surface deposition of radioactivity are interrelated functions of the co-ordinates, time and soil properties. Therefore, $K$ is an integral function with many variables. The dependence of this function on the different variables is not addressed here. However, we do want to examine various sources of uncertainty of the applied models of resuspension, with the resuspension factor as the characteristic parameter of resuspension, as well as to estimate the limits of the model applications. In the light of the recent review work on modelling of resuspension [4], the present work focuses particularly on testing the widely applied simple models for $K$ on the basis of data obtained in the vicinity of the Chernobyl NPP.

One empirical predictive resuspension model describes the time dependence as
\[ K(t) = K(0) \exp(-\lambda t) \]

where \( \lambda \) is the half-time [5]. Such a formulation appears to simulate reasonably well the available observations for time periods of up to several weeks after deposition.

Another simple model is used to approximate the time dependence in the following manner:

\[ K(t) = K(0) \exp(-\lambda \sqrt{t}) + 10^{-9} \]  \hspace{1cm} (2)

The second term of Eq. (2) was developed on the basis of data obtained during 17 years from the observations of plutonium in air, and the initial value of \( K(0) \) was equal to \( 10^{-4} \) m\(^{-1}\).

The liquid metal fast breeder reactor study and the reactor safety study [6] used the following formulas for \( K \):

\[ K(t) = 10^{-5} \exp(-0.0139t) + 10^{-9} \]  \hspace{1cm} (3)

\[ K(t) = 10^{-5} \exp(-0.00185t) + 10^{-9} \]  \hspace{1cm} (4)

A similar formula is:

\[ K(t) = 10^{-6} \exp(-0.01t) + 10^{-9} \]  \hspace{1cm} (5)

where \( t \) is the time in days [6] and the initial value of \( K \) was selected by discounting the relatively damp climate in the United Kingdom. With this formula, the background value of \( K \) can be obtained after two years. Thus, all of these expressions are empirical interpolation formulas describing the process of transition from an initial high level of \( K \) to the background level.

In contrast to Eqs (2)-(5), the following expression does not have the low background level for \( K \) [4]:

\[ K(t) = 1.2 \times 10^{-6} t^{-1} \]  \hspace{1cm} (6)

where \( t \) is the time in days. Empirical equations accounting for changes of \(^{137}\)Cs activity in air with time have been given for Neuherberg from mid-May 1986 to the end of 1988 [7] and from June 1986 to the end of 1990 [8]. Normalizing these equations for the density of \(^{137}\)Cs contamination in the 0-1 cm soil layer [8] gives the following formulas:

\[ K(t) = 3.4 \times 10^{-6} \exp(-0.152t) + 18.4 \times 10^{-9} \exp(-0.003t) \]  \hspace{1cm} (7)

\[ K(t) = 2.67 \times 10^{-6} t^{-1.07} \]

where \( t \) is the time in days.
TABLE I. ANNUAL MEAN RESUSPENSION FACTORS K FOR $^{137}$Cs MEASURED AT CHERNOBYL CITY IN 1986 (AFTER 20 MAY 1986) AND IN 1987–1991, COMPARED WITH MODEL CALCULATIONS

<table>
<thead>
<tr>
<th>Year</th>
<th>Lower limit of K ($10^{-7}$ m$^{-1}$)</th>
<th>Mean value of K ($10^{-7}$ m$^{-1}$)</th>
<th>Upper limit of K ($10^{-7}$ m$^{-1}$)</th>
<th>$K_{Gar}$ ($10^{-7}$ m$^{-1}$)</th>
<th>$K_{Hoet}$ ($10^{-7}$ m$^{-1}$)</th>
<th>$K_{Lin}$ ($10^{-7}$ m$^{-1}$)</th>
<th>$K_{Gar}$ / $K_{exp}$</th>
<th>$K_{Hoet}$ / $K_{exp}$</th>
<th>$K_{Lin}$ / $K_{exp}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1986</td>
<td>0.053</td>
<td>0.330</td>
<td>0.830</td>
<td>0.270</td>
<td>0.510</td>
<td>3.90</td>
<td>0.82</td>
<td>1.54</td>
<td>11.8</td>
</tr>
<tr>
<td>1987</td>
<td>0.013</td>
<td>0.082</td>
<td>0.170</td>
<td>0.030</td>
<td>0.042</td>
<td>0.27</td>
<td>0.36</td>
<td>0.51</td>
<td>3.30</td>
</tr>
<tr>
<td>1988</td>
<td>0.004</td>
<td>0.032</td>
<td>0.070</td>
<td>0.014</td>
<td>0.021</td>
<td>0.016</td>
<td>0.44</td>
<td>0.66</td>
<td>0.50</td>
</tr>
<tr>
<td>1989</td>
<td>0.002</td>
<td>0.014</td>
<td>0.034</td>
<td>0.010</td>
<td>0.014</td>
<td>0.010</td>
<td>0.71</td>
<td>1.00</td>
<td>0.71</td>
</tr>
<tr>
<td>1990</td>
<td>0.0008</td>
<td>0.006</td>
<td>0.015</td>
<td>0.008</td>
<td>0.010</td>
<td>0.010</td>
<td>1.33</td>
<td>1.66</td>
<td>1.66</td>
</tr>
<tr>
<td>1991</td>
<td>0.001</td>
<td>0.008</td>
<td>0.018</td>
<td>0.006</td>
<td>0.008</td>
<td>0.010</td>
<td>0.75</td>
<td>1.00</td>
<td>1.25</td>
</tr>
</tbody>
</table>

* $K_{Gar}$ — calculated according to the Garland et al. model [4];
* $K_{Hoet}$ — calculated according to the Hoetzl et al. model [8];
* $K_{Lin}$ — calculated according to the Linsley model [6];
* $K_{exp}$ — derived from experimental measurements.
In Eqs (2)-(6) the first general uncertainty is the selection of the initial value of the resuspension factor, which can change from $10^{-6}$ to $10^{-4}$ m$^{-1}$ during the first ten days, producing a difference among model estimates of $K$ of two orders of magnitude. The second general source of uncertainty is connected with the rate of decrease of $K$ with time; the time-scale $\lambda$ in Eqs (2)-(6) may change by two to three orders of magnitude. The background term causes errors in the estimates of $K$ from Eqs (2)-(5) for a very long time after the initial starting point.

When comparing model predictions with empirical data, a question arises about the correspondence of time or space averaging of calculated and measured values of $K$. Since for model values of $K$, homogeneity of external conditions (soil, contamination, character of the underlying surface) and stationary meteorological conditions for the measuring period are assumed, we opted to use empirical data averaged over the whole year.

The annual averages of the resuspension factor, as determined experimentally and as calculated by models [4, 6, 8], are shown in Table I. This table also shows lower and upper limits of $K$ from 1986 to 1991, and the ratio of calculated values of $K$ to experimental ones. The model of Hoetzl et al. [8], derived from empirical data measured in Neuherberg after the Chernobyl accident, agrees best with the data. The model of Garland et al. [4] agrees well with the data, but it systematically underpredicts the experimental values. The calculated values of $K$ from the Linsley model [6] are conservative, especially for the first two years. If the calculated values are compared with the upper limits of the measured values, as is important in practice, the data of the Garland model [4] and the Hoetzl model [8] are lower than the upper limits of $K$ by a factor of two to three. It is important to note that lower and upper limits are placed on the monthly mean resuspension factors. The actual ratio of $K$ for the daily data will be even larger.

It is also interesting to compare modelled values of concentrations with measured ones. For this purpose, we selected concentration data for two months (Fig. 1). The concentrations calculated with two models [4, 6] are presented as the upper and lower models, respectively, with the daily concentrations of $^{137}Cs$ obtained in September 1987 and in September 1990. For 1987, these model estimates yielded insufficient values for many days, especially in the case of the Garland model [4]. The data for 1990 are in accordance with the data of Table I. There is a certain amount of overestimation in both the annual and the daily concentrations.

Finally, we have compared the model $K$ values with the experimental data obtained at different points in the 30 km zone. Air concentrations and surface deposition of eight radionuclides were measured at ten points during the same period (14-17 September 1986) [2]. The calculated resuspension factors range from $6 \times 10^{-9}$ m$^{-1}$ to $3 \times 10^{-6}$ m$^{-1}$. Data on the spatial means of $K$ are presented in Table II for time periods in 1986 and 1991. Also shown are the calculated values according to the three models [4, 6, 8]. The experimental data and the calculated results differ by up to more than one order of magnitude. The large range of the
resuspension factor reflects the differences in the underlying surface soil and the effects of anthropogenic actions in the 30 km zone. The large range of values is especially important for locations near the NPP and for general roads. The advection effects from upwind sources have been described in the literature [1, 9] and the K values for concrete may increase by two to three orders of magnitude. All of these
TABLE II. COMPARISON OF CALCULATED RESUSPENSION FACTORS K WITH THOSE EXPERIMENTALLY DETERMINED AFTER THE CHERNOBYL ACCIDENT

<table>
<thead>
<tr>
<th>Model/data</th>
<th>$K \text{ (m}^{-1}\text{)}$</th>
<th>$K \text{ (m}^{-1}\text{)}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$t = 117$ days</td>
<td>$t = 1957$ days</td>
</tr>
<tr>
<td>Linsley [6]</td>
<td>$31 \times 10^{-8}$</td>
<td>$10 \times 10^{-10}$</td>
</tr>
<tr>
<td>Garland et al. [4]</td>
<td>$1.1 \times 10^{-8}$</td>
<td>$6.1 \times 10^{-10}$</td>
</tr>
<tr>
<td>Hoetzl et al. [8]</td>
<td>$1.6 \times 10^{-8}$</td>
<td>$8.0 \times 10^{-10}$</td>
</tr>
<tr>
<td>Experimental data</td>
<td>$^{137}\text{Cs}^a$: $(17 \pm 19) \times 10^{-8}$</td>
<td>First point: $^{137}\text{Cs}$: $(4.3 \pm 2.2) \times 10^{-10}$</td>
</tr>
<tr>
<td></td>
<td>$^{106}\text{Ru}$: $(23 \pm 16) \times 10^{-8}$</td>
<td>$^{137}\text{Cs}$: $(4.3 \pm 2.2) \times 10^{-10}$</td>
</tr>
<tr>
<td></td>
<td>$^{144}\text{Ce}$: $(19 \pm 24) \times 10^{-8}$</td>
<td>Second point: $^{95}\text{Zr}$: $(20 \pm 23) \times 10^{-8}$</td>
</tr>
<tr>
<td></td>
<td>$^{137}\text{Cs}$: $(4.3 \pm 2.2) \times 10^{-10}$</td>
<td>$^{137}\text{Cs}$: $(2.4 \pm 1.0) \times 10^{-10}$</td>
</tr>
</tbody>
</table>

$^a$ Mean over ten points and three days.

$^b$ Mean of six values of $K$ measured during one day.

Factors put severe limitations on the practical application of $K$ and require new elaboration on a more physical base.

In conclusion, we note that it is necessary to take into account the fluctuations of the air concentration of radioactivity in order to obtain more correct estimates of the uncertainty of $K$. An empirical expression has been given [10] for the standard deviation of the air concentration with different observation times:

$$
\sigma(T_2) = \sigma(T_1) \times \left( \frac{T_2}{T_1} \right)^{-m}
$$

where the exponent $m$ is $0.33 \pm 0.08$. This formula permits estimation of more realistic limits of the variation of the experimental values of $K$. For example, the standard deviation of the daily air concentrations will be three times higher than the standard deviation of the monthly air concentrations.

REFERENCES


RADIOLOGICAL IMPACT ASSESSMENT

(Session 7)

Chairman

F.O. HOFFMAN
United States of America
EXPOSURE OF THE POPULATION OF COUNTRIES WITHIN THE EUROPEAN UNION TO RADIOACTIVITY IN THE MEDITERRANEAN SEA: Project MARINA-MED

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Abstract

EXPOSURE OF THE POPULATION OF COUNTRIES WITHIN THE EUROPEAN UNION TO RADIOACTIVITY IN THE MEDITERRANEAN SEA: PROJECT MARINA-MED.

The discharges examined in the Project MARINA-MED were from nuclear installations in France, Italy and Spain, over the period 1980-1991; those from the Marcoule fuel reprocessing plant were found to contribute most to the collective dose arising from radioactivity in the Mediterranean Sea. Over 90% of beta-gamma discharges were of tritium and the remainder comprised some 80% $^{106}$Ru + $^{106}$Rh, 7% $^{90}$Sr + $^{90}$Y, 6% $^{137}$Cs, and 1% each of $^{134}$Cs, $^{58}$Co and $^{54}$Mn. The alpha emitters were dominated by $^{241}$Am, $^{239+240}$Pu and natural uranium, each representing 30% of the total. The contribution from Chernobyl had largely disappeared by 1990, except in the northern Aegean Sea, where continuing input came from the Black Sea. The corresponding exposure of the critical group in the Mediterranean Sea area from $^{137}$Cs (for an annual consumption of 73 kg fish and 35 kg shellfish) was estimated to be 7.5 $\mu$Sv in 1990; the exposure of the critical group in the Black Sea area would have been about 40 $\mu$Sv. These values are much lower than that estimated for $^{210}$Po, which corresponds to about 0.5 mSv. Data were obtained on catches and consumption rates of fish, crustaceans and molluscs for the relevant countries within the European Union, including import and export. Using data from the Food and Agriculture Organization of the United Nations, it was possible to extend the data to other countries bordering the Mediterranean Sea. Peak annual collective doses were estimated at less than 0.2 man-Sv for the period considered. For discharges over the entire period (1980-1991) the total collective dose commitment, truncated at 500 years, was estimated to be about 2 man-Sv. The most significant radionuclides, especially in the first few years, were found to be $^{106}$Ru (0.76 man-Sv) and $^{241}$Am (0.25 man-Sv).

1. INTRODUCTION

The project MARINA-MED [1, 2] was established subsequent to an analogous project [3] concerning radioactivity in the north-east Atlantic. The two projects adopted the same approach.

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2. SOURCES OF RADIOACTIVITY IN THE MEDITERRANEAN SEA

Both natural radionuclides, such as $^{14}$C, $^3$H, $^{10}$Be, $^7$Be, $^{26}$Al, $^{238}$U, $^{235}$U and $^{232}$Th, with their decay series, and man-made radionuclides are found in the marine environment. Detonation of nuclear weapons in the atmosphere, controlled discharges of low level liquid effluents from nuclear power plants, fallout from accidents such as the Chernobyl reactor accident in 1986 and the Transit 5BN-3 satellite accident in 1964, discharges from laboratories at universities and research centres, and phosphate ore processing are the main sources of man-made radionuclides.

The impact of nuclear installations in the Mediterranean marine environment is particularly felt in coastal regions near the mouths of rivers, such as Rhone, Ebro, Júcar and Po, as well as Dnieper and Don, which flow into the Black Sea — linked to the Mediterranean.

The nuclear power plants taken into account are located in Spain, France and Italy. Other plants located outside these countries discharged either directly or indirectly into the Black Sea. By far the main sources of radionuclide releases are the discharges from the Marcoule complex. These releases account for 76.7% of the total beta-gamma discharges and for more than 99.9% of the total alpha discharges (Table I).

Aside from $^3$H, which corresponds to 92.26% of the global beta-gamma releases, the remaining radionuclides are subdivided as shown in Table II.

Other releases from the industry, research centres and hospitals are negligible compared with those mentioned above.

One officially confirmed accident with losses of radioactive material at sea occurred on 17 January 1966, when a US B-52 bomber crashed over the Almanzora river, between Palomares and Cuevas de Almanzora (Spain). On the continental shelf, the estimated percentages were 78% for Pu and 52% for Am with respect to fallout radioactivity in marine sediments in this area (Vera Gulf).

Another source of radioactivity is phosphate rock, which is extensively used to gain phosphorus for fertilizers. It contains trace amounts of $^{40}$K and $^{232}$Th, with

<p>| TABLE I. DISTRIBUTION OF DISCHARGES INTO THE MEDITERRANEAN SEA |
|---------------------------------|-----------------|--------|---------------|</p>
<table>
<thead>
<tr>
<th>Discharges (1980-1991)</th>
<th>Marcoule</th>
<th>Other</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beta–gamma radionuclides</td>
<td>0.533 PBq</td>
<td>0.013 PBq</td>
<td>0.546 PBq</td>
</tr>
<tr>
<td>(without $^3$H)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Tritium</td>
<td>4.87 PBq</td>
<td>1.63 PBq</td>
<td>6.50 PBq</td>
</tr>
<tr>
<td>Alpha radionuclides</td>
<td>894 GBq</td>
<td>0.26 GBq</td>
<td>894.26 GBq</td>
</tr>
</tbody>
</table>
its decay products, as well as the radionuclides of the $^{238}\text{U}$ decay series, which are 5-50 times higher than in normal soil.

3. RADIOACTIVITY IN THE MEDITERRANEAN SEA

From data on $^{137}\text{Cs}$ and $^{210}\text{Po}$ in surface water, fish, shellfish and coastal surface ($\approx$0-5 cm) sediments from the seas and basins comprising the Mediterranean, the individual mean doses to the population of the Mediterranean countries belonging to the European Union (EU) from the consumption of seafood were calculated.

Since the main sources of radioactive contamination of the Mediterranean Sea are global fallout and debris from Chernobyl, the dose calculations reported here comprise only these sources. A special study was performed for the Gulf of Lions, which receives radioactivity from the nuclear plants located along the Rhone river.

The individual mean doses from intake of radionuclides with marine food were based upon the intakes of $^{137}\text{Cs}$ and $^{210}\text{Po}$ in 1990. Dose commitments were calculated for consumption of 5 kg fish and 2 kg shellfish pro capite per year. The critical group was assumed to eat 73 kg fish and 35 kg shellfish per year. The dose factors used for the dose calculation were the same as those applied in the MARINA Project [3], i.e. $1.2 \times 10^{-8}$ Sv per Bq for $^{137}\text{Cs}$ and $4.3 \times 10^{-7}$ Sv per Bq for $^{210}\text{Po}$.

The $^{137}\text{Cs}$ concentrations in the Black Sea are one order of magnitude higher than those in the remaining part of the Mediterranean Sea. In 1993, at the exit of the Bosporus Channel, a value of 26 Bq $^{137}\text{Cs}/m^3$ was measured, while the concentration in the Mediterranean Sea was a few Bq $^{137}\text{Cs}/m^3$. The measured $^{210}\text{Po}$ concentration in the western Mediterranean was $1.1 \pm 0.2$ Bq/m$^3$. 

<table>
<thead>
<tr>
<th>Beta-gamma radionuclides</th>
<th>Percentage ($^3\text{H}$ excluded)</th>
<th>Alpha radionuclides</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ru-106 + Rh-106</td>
<td>79.69</td>
<td>Am-241</td>
<td>31.4</td>
</tr>
<tr>
<td>Sr-90 + Y-90</td>
<td>7</td>
<td>Pu-239 + Pu-240</td>
<td>27.87</td>
</tr>
<tr>
<td>Cs-137</td>
<td>5.63</td>
<td>Natural U</td>
<td>24.79</td>
</tr>
<tr>
<td>Cs-134</td>
<td>1.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Co-58</td>
<td>0.93</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mn-54</td>
<td>0.8</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
In fish and shellfish the $^{137}\text{Cs}$ concentration was less than 1 Bq/kg (sometimes much less). The mean concentration of $^{210}\text{Po}$ in fish was $7 \pm 4$ Bq/kg, but this value was obtained from very few measurements. The mean level of $^{210}\text{Po}$ in shellfish was $21 \pm 4$ Bq/kg. The mean concentration of $^{137}\text{Cs}$ in surface sediments in 1990 was $6.5 \pm 4.4$ Bq/kg.

The $^{137}\text{Cs}$ data from the Gulf of Lions are rather close to those from the Liguro-Provençal Basin. In general, only the available data of $^{137}\text{Cs}$ concentrations were used; these data were collected for radiation protection purposes. However, the evaluation of these data suggests that the actual mean values are probably less than a factor of two of the present estimates.

The mean concentration factor for fish ($0.8 \times 10^2$), calculated from corresponding seawater values, was in good agreement with the value recommended by the IAEA [4], i.e. $10^2$. However, the value found for shellfish ($2 \times 10^2$) was higher than the IAEA value of 30. This difference is primarily due to the very localized distribution of the Chernobyl fallout.

The mean $K_d$ value for $^{137}\text{Cs}$ (Bq/kg dry weight per Bq/L sea water) for sediments ($1.4 \times 10^3$) was found to be in reasonable agreement with the recommended IAEA values, which are $1.7 \times 10^3$ for deep ocean sediments and $3 \times 10^3$ for coastal sediments.

The concentration factor of $^{210}\text{Po}$ was calculated to be $(6 \pm 4) \times 10^3$ for fish and $(19 \pm 5) \times 10^3$ for shellfish — not incompatible with the recommended IAEA values, which are $2 \times 10^3$ and $(1-5) \times 10^4$, respectively.

After the Chernobyl accident, the radionuclide concentrations in sea water and fish increased by an order of magnitude. The Chernobyl signal was, in general, less evident in shellfish but was particularly evident in sediments. In 1990, the $^{137}\text{Cs}$ levels in sea water and fish were again at the pre-Chernobyl values, except in the Black Sea and the northern Aegean Sea, which in 1993 still showed enhanced levels.

The input of Chernobyl $^{137}\text{Cs}$ to the Mediterranean Sea (except for the Black Sea) was estimated to be 25-40% of the inventory of $^{137}\text{Cs}$ from previous global fallout, estimated for 1986 by the United Nations Environment Programme (UNEP)/IAEA [5] to be $12 \pm 2$ PBq. Hence, the Chernobyl input is estimated at 3-5 PBq $^{137}\text{Cs}$. The inflow of Chernobyl $^{137}\text{Cs}$ from the Black Sea is less than the decay of the 3-5 PBq $^{137}\text{Cs}$ already present in the Mediterranean Sea. Therefore, a general increase of $^{137}\text{Cs}$ in the Mediterranean Sea due to this inflow is not expected.

The input from nuclear power plants discharging liquid waste into the rivers flowing into the Mediterranean Sea was estimated by the UNEP/IAEA [5] to be approximately 5 TBq $^{137}\text{Cs}$/a. This is less than 1% of the inventory of $^{137}\text{Cs}$ and is more than compensated for by its radioactive decay.

In 1990 the overall mean concentration of $^{137}\text{Cs}$ in fish from the Mediterranean Sea alone was $0.37 \pm 0.27$ Bq/kg and that in shellfish was $0.30 \pm 0.09$ Bq/kg. For a mean consumption rate of fish and shellfish, as reported before, the total
$^{137}$Cs intake per person is 2.5 Bq/a. By applying the $^{137}$Cs concentration factors recommended by the IAEA for fish (100) and shellfish (30), the estimation from seawater concentrations gives an intake of 3.2 Bq $^{137}$Cs/a.

If an effective half-life of $^{137}$Cs in the Mediterranean of 10 years is assumed, the effective dose from $^{137}$Cs in marine foods for 1990 and the following years is 0.5 µSv; the same value was found when the dose due to $^{137}$Cs from consumption of fish and shellfish for the first years after Chernobyl (1986–1989) was calculated as above from the time integrated mean seawater concentrations.

The annual intake of $^{210}$Po from fish and shellfish is 77 Bq, corresponding to an effective dose of 33 µSv from the intake in 1990, i.e. three orders of magnitude higher than the dose from $^{137}$Cs; however, the very few data available make the dose estimate for $^{210}$Po less reliable.

For a hypothetical critical group, the doses from the 1990 intake are 0.5 µSv from $^{137}$Cs and 0.54 mSv from $^{210}$Po. The latter dose is half of the annual dose from natural background radiation (excluding the radon dose). The critical Mediterranean group may receive an effective dose from Chernobyl $^{137}$Cs in marine food of about 7.5 µSv. A corresponding critical group in the Black Sea would receive a five times higher dose, i.e. 40 µSv.

4. SURVEY OF THE QUANTITIES AND UTILIZATION OF MARINE PRODUCTS

Data from the Food and Agriculture Organization of the United Nations (FAO) and from national sources were used to allocate nominal catches for the different compartments of the Mediterranean Sea to France, Greece, Italy and Spain. For completeness, the Black Sea was considered as an additional compartment. Import and export data on fish, crustaceans and molluscs of EU Member States during 1988 and 1990 were obtained from EUROSTAT C.

For the collective dose calculations, the nominal landings were converted into amounts of marine products actually available for human consumption by taking half of the wet weight for fish, one third for crustaceans and one sixth for molluscs.

It is assumed that EU Member States not bordering the Mediterranean Sea do not have any significant fisheries in this area and that marine products from the Mediterranean Sea imported by any EU Member State are not exported again by the receiving country.

From different food basket studies, roughly half of the net amount of marine products available for human consumption is actually eaten by humans. Therefore, by taking the net available amounts of marine products from the Mediterranean Sea as the basis for the collective dose calculations, some overestimation may result.
5. RADIOLGICAL IMPACT ON EU MEMBER STATES OF ROUTINE DISCHARGES INTO THE MEDITERRANEAN SEA

The radiological impact on EU Member States was evaluated using data on liquid discharges from European civil nuclear plants in the period 1980–1991. The radiological impact of $^{137}$Cs which entered the Mediterranean Sea from the Black Sea as a result of the Chernobyl accident and which was measured in 1990 was also estimated.

A mathematical model, ATOMED [6], was developed to predict the dispersion of radionuclides in the Mediterranean Sea and to estimate the exposure of the population of EU countries in this area. This model represents the Mediterranean Sea by a number of compartments which simulate the horizontal and vertical structure of the sea. Instantaneous uniform mixing within the compartments was assumed and the movement of water masses between various sea areas was modelled using rates of transfer between adjacent compartments.

The dynamics of the Mediterranean is in many places highly baroclinic and a vertical structure has been adopted in the dispersion model. A 100 m deep upper layer represents an annual thermocline. Two deep compartments represent the intermediate and deep waters of the eastern and western Mediterranean Levantine. Separate compartments representing the bottom boundary layer have been included to model in detail the processes at the interface of the water column—bottom sediments.

A model for the Rhone river [6, 7] was also used to model more accurately the dispersion of radionuclides discharged by the Marcoule plant. For this plant, radioactive decay of the nuclides discharged was ignored because it takes only a few days for the river water to reach the sea from the point of discharge.

The time-scale for transport of unconsolidated river bed sediments containing adsorbed radionuclides from Marcoule to the Mediterranean Sea is of the order of 30 years. The concentrations of the short and medium lived radionuclides absorbed onto such sediments before they reach the sea decrease considerably owing to radioactive decay.

Collective doses were calculated for four exposure pathways: consumption of fish, crustaceans and molluscs, and external exposure from contaminated beach sediments. Doses due to ingestion of seafood were calculated using activity concentrations in marine biota, amounts of seafood consumed by people living in EU countries in this area, edible fractions of each type of seafood, and the committed effective dose equivalents per unit intake of each radionuclide.

The collective doses resulting from external exposure to radioactivity deposited in beach sediments were calculated, assuming a beach occupancy rate of $75 \text{ h} \cdot \text{m}^{-1} \cdot \text{a}^{-1}$. The total collective dose commitment, truncated at 500 years, delivered to the population of the EU countries, following the discharge of liquid radioactive effluents from nuclear installations of EU Member States in the period 1980–1991, was estimated to be $1.96 \text{ man} \cdot \text{Sv}$. 
TABLE III. COLLECTIVE DOSES FROM THE MOST IMPORTANT RADIONUCLIDES

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Dose (man • Sv)</th>
<th>Mostly delivered before</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ru-106</td>
<td>0.76</td>
<td>1995</td>
</tr>
<tr>
<td>Am-241</td>
<td>0.25</td>
<td>1994</td>
</tr>
<tr>
<td>Sr-90</td>
<td>0.21</td>
<td>2050</td>
</tr>
<tr>
<td>Cs-137</td>
<td>0.14</td>
<td>2200</td>
</tr>
<tr>
<td>Pu-239</td>
<td>0.13</td>
<td>2200</td>
</tr>
<tr>
<td>Co-60</td>
<td>0.049</td>
<td>1992</td>
</tr>
<tr>
<td>Co-58</td>
<td>0.036</td>
<td>1992</td>
</tr>
<tr>
<td>Cs-134</td>
<td>0.02</td>
<td>1992</td>
</tr>
<tr>
<td>Pu-238</td>
<td>0.019</td>
<td>2100</td>
</tr>
<tr>
<td>Ce-144</td>
<td>0.011</td>
<td>1992</td>
</tr>
</tbody>
</table>

The most important radionuclides contributing to the collective dose are presented in Table III. Ingestion of molluscs is the dominant exposure pathway (60% of the total collective dose) and $^{106}$Ru (62%) is the main contributor to the dose. Ingestion of fish (20% of the total collective dose) and external exposure to radioactivity in beach sediments (18% of the total collective dose) are also significant pathways. The most important contributors to the collective dose associated with the ingestion of fish are $^{90}$Sr and $^{137}$Cs. They account for 51% and 31%, respectively, of the total collective dose due to this pathway. The most significant radionuclides contributing to the collective dose due to external irradiation from beach sediments are $^{60}$Co (27%), $^{58}$Co (27%) and $^{106}$Ru (13%).

The Marcoule nuclear plant is by far the largest contributor to the collective dose arising from man-made radioactivity in the Mediterranean Sea. The total collective dose associated with discharges from Marcoule was 1.6 man • Sv, which is 83% of the total collective dose delivered by all nuclear plants in the EU discharging into the Mediterranean Sea.

The highest collective dose rates were calculated for France in 1982 and 1984 (0.083 man • Sv/a), for Italy in 1987 (0.05 man • Sv/a) and for Spain in 1987: (0.046 man • Sv/a). In 1992, at the end of the period of radioactive release, the collective dose rate was 0.079 man • Sv/a. France and Italy each contributed 35% of this dose rate, while Spain accounted for 24% of it.

The highest collective doses to the populations of the Mediterranean countries are: 0.78 man • Sv in France, 0.6 man • Sv in Italy and 0.46 man • Sv in Spain. The
populations of the EU countries which import large quantities of seafood from the Mediterranean region and Greece are exposed to the following collective doses: 0.02 man·Sv in Greece, 0.02 man·Sv in Germany and 0.017 man·Sv in Portugal. The collective doses to the populations of all other EU nations contribute less than 0.5% each to the total collective dose.

The most important contribution to the total collective dose (49%) is due to the release of liquid discharges from French nuclear plants into the Gulf of Lions. The Liguro-Provençal Basin, which is connected directly with the Gulf of Lions and receives liquid discharges from the Spanish installations, accounts for 32% of the total collective dose. The Adriatic Sea (8%), into which the Italian nuclear plants discharge their liquid effluents, the Tyrrhenian Sea (5%) and the Ionian Sea (3%) also contribute significantly to the total collective dose. The contribution to the total collective dose of all the other compartments is less than 1%.

The total collective dose, truncated at 500 years, to the EU population due to liquid discharges into the northern European waters was calculated from the Project MARINA [3] to be 5200 man·Sv, i.e. more than three orders of magnitude higher than the corresponding value obtained for the Mediterranean Sea.

It was estimated that the input of $^{137}$Cs into the Black Sea from the Dnieper river due to the Chernobyl accident was 2.4 PBq and the collective doses from $^{137}$Cs to the EU population was 3.7 man·Sv. The global inventory of $^{137}$Cs in the Mediterranean Sea was estimated to be 12 PBq and the inflow from the Black Sea 0.14 PBq — the increase in the activity concentration therefore being of the order of 1%. The most important sources of $^{137}$Cs in the Mediterranean Sea and in the Black Sea are fallout from experimental nuclear weapons explosions and releases from the Chernobyl nuclear power plant.

The total collective dose due to $^{137}$Cs resulting from its concentration in surface waters and sea-bed sediments was 2.6 man·Sv, and 91% of this dose was due to consumption of fish. The contribution of fallout from nuclear bombs and from the Chernobyl accident to the collective dose was estimated to be 3.7 man·Sv. The total collective dose due to the discharges of nuclear plants in the EU countries was predicted by the model ATOMED to be 1.6 man·Sv.

REFERENCES


TEMPORAL AND SPATIAL DISTRIBUTION OF THE ENVIRONMENTAL IMPACT OF RADIOACTIVE RELEASES FROM NUCLEAR FUEL CYCLE FACILITIES*

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Abstract

TEMPORAL AND SPATIAL DISTRIBUTION OF THE ENVIRONMENTAL IMPACT OF RADIOACTIVE RELEASES FROM NUCLEAR FUEL CYCLE FACILITIES.

An assessment of the radiological impact of all stages of the nuclear fuel cycle in France has been completed. The fuel cycle is broken down into nine stages, including the transportation of radioactive material between the different stages. For each facility, representative annual environmental release rates and measured exposures of workers are used. The results are presented in a time and space matrix in order to provide a clear perspective for the distribution of the results. A total collective dose of 13 man·Sv/TW·h of electricity generation has been calculated. The local and regional doses contribute less than 8% to this total dose, which is dominated by the global long term collective dose due to $^{14}$C releases from electricity generation and reprocessing (12 man·Sv/TW·h). The relevance of this value must be considered carefully because it is the result of integration of many, extremely small, average individual doses for a large population group over long periods of time. If the global doses are not included, more than half of the remaining 0.9 man·Sv/TW·h is due to occupational doses. The study clearly shows the need to present the results of a complicated assessment including some imperfect methodologies in a transparent manner, so that the potential risks can be properly considered in decision making.

1. INTRODUCTION

As the scope of impact assessments increases, it becomes necessary to make decision makers aware of the temporal and regional distribution of the impacts being reported. The current trend in life-cycle analysis methodologies has expanded the assessment of risks, not only to second and third order processes but also to a global scale. The potential long term impact of activities such as waste disposal has extended the time-scale to hundreds of thousands of years into the future. If such

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large scales of time and space are included in a single value, then an accurate presentation of the risks is not provided. For the decision making process it is necessary to have sufficient information to be able to weigh the key criteria and to balance the available options.

The degree of variation in the range of results reported for a risk assessment project has been illustrated in the Joint European Union/United States Department of Energy project on the external costs of fuel cycles [1]. The general objective of the study was to evaluate the priority impact pathways for each stage of ten different fuel cycles, within the same methodological framework. In the course of this study, it became clear that with the existing methods and data there was a large disparity in the range of time and space impacts being reported for the nuclear fuel cycle compared with other fuel cycles.

In view of this, the results of the radiological impact assessment of specific installations representing each stage of the nuclear fuel cycle in France have been organized in such a way as to provide a clear perspective for use in comparative risk assessment and decision making. This assessment has also re-emphasized some of the key problems that remain unresolved in the comparative assessment methodologies currently being employed.

2. METHODOLOGY

The radiological impacts, normalized to a unit of energy production, have been calculated on the basis of radiological releases from the routine operation of facilities representing nine stages of the nuclear fuel cycle in France (Fig. 1). Transportation and a hypothetical repository for high level waste disposal have been included. The distribution of the radiological impacts in time and space has been assessed, categorized by medium and long time-scales, and by local, regional and global spatial scales.

Deterministic radiological health effects do not occur as a result of routine operations, so no short term (<1 year) impacts are reported. The medium term period is considered to last from 1 year to 100 years after the time of release, and the arbitrary cut-off for the long term period has been set at 100 000 years. The local region is considered to be within a radius of 100 km of the site, and the boundary between regional and global is at a radius of 1000 km. The dispersion of four radionuclides (\(^{85}\)Kr, \(^{129}\)I, \(^{3}\)H and \(^{14}\)C) has been assessed for the global long term category. The definition of these categories was influenced by the dispersion models used and the risks assessed for the other fuel cycles in the overall project.

For each existing facility, representative annual atmospheric and liquid environmental release rates were used as source terms for the radiological impact assessment. The environmental transfer of each radionuclide was considered independently, using generalized pathway models [2, 3] with site specific meteo-
FIG. 1. Sites of nuclear fuel cycle facilities included in the assessment.

1. Mining and milling; 2. transformation of yellowcake; 3. conversion; 4. enrichment; 5. fuel fabrication; 6. nuclear power plant; 7. reprocessing; 8. low and intermediate level waste disposal; 9. proposed site for high level waste disposal.

logical and/or aquatic conditions. For calculation of the collective dose, the site specific geographical distribution of the population was input, assuming a standard man with average rates of food consumption.

The assessment of the impact of a high level waste disposal site was based on a hypothetical geological repository in granite that was completed for a waste disposal project for the European Community [4]. Transportation impacts are based on French statistics and on the actual amounts of radioactive material that would be transported. The INTERTRAN model was used for non-accidental situations [5].
<table>
<thead>
<tr>
<th>Stage of cycle</th>
<th>Local</th>
<th>Local and regional</th>
<th>Global</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Direct exposure</td>
<td>Occupational exposure</td>
<td>Atmospheric releases</td>
<td>Liquid releases</td>
</tr>
<tr>
<td>Mining and milling</td>
<td>0</td>
<td>$1.1 \times 10^{-1}$</td>
<td>$1.8 \times 10^{-1}$</td>
<td>$1.5 \times 10^{-5}$</td>
</tr>
<tr>
<td>Conversion</td>
<td>0</td>
<td>$2.3 \times 10^{-3}$</td>
<td>$3.4 \times 10^{-5}$</td>
<td>$4.6 \times 10^{-7}$</td>
</tr>
<tr>
<td>Enrichment</td>
<td>0</td>
<td>$8.3 \times 10^{-6}$</td>
<td>$2.6 \times 10^{-5}$</td>
<td>$9.3 \times 10^{-8}$</td>
</tr>
<tr>
<td>Fuel fabrication</td>
<td>0</td>
<td>$7.1 \times 10^{-3}$</td>
<td>$4.2 \times 10^{-7}$</td>
<td>$8.8 \times 10^{-6}$</td>
</tr>
<tr>
<td>Electricity generation</td>
<td>0</td>
<td>$3.5 \times 10^{-1}$</td>
<td>$1.2 \times 10^{-5}$</td>
<td>$1.7 \times 10^{-2}$</td>
</tr>
<tr>
<td>Decommissioning</td>
<td>$1.5 \times 10^{-4}$</td>
<td>$2.2 \times 10^{-2}$</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Reprocessing</td>
<td>0</td>
<td>$1.8 \times 10^{-3}$</td>
<td>$8.4 \times 10^{-3}$</td>
<td>$5.3 \times 10^{-2}$</td>
</tr>
<tr>
<td>Disposal of LLW</td>
<td>0</td>
<td>$1.0 \times 10^{-4}$</td>
<td>0</td>
<td>$1.3 \times 10^{-5}$</td>
</tr>
<tr>
<td>Disposal of HLW</td>
<td>0</td>
<td>$6.0 \times 10^{-7}$</td>
<td>0</td>
<td>$1.4 \times 10^{-1}$</td>
</tr>
<tr>
<td>Transport</td>
<td>$1.3 \times 10^{-3}$</td>
<td>$1.2 \times 10^{-3}$</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td><strong>Sub-total</strong></td>
<td>$1.5 \times 10^{-3}$</td>
<td>$5.0 \times 10^{-1}$</td>
<td>$1.9 \times 10^{-1}$</td>
<td>$2.01 \times 10^{1}$</td>
</tr>
</tbody>
</table>

*a The average individual doses are integrated for a time period of 100,000 years.
*b Due to radioactive releases to the environment; assuming a world population of ten billion people.
*c Direct exposure of the public (not due to environmental releases).
*d Due to food products produced within the regional area (radius of 1000 km) but consumed elsewhere.
The occupational doses reported in this study are based on the actual worker exposure records from each facility. A detailed description of the methodology can be found in the main report of this project [6].

3. RESULTS AND DISCUSSION

Table I presents the distribution of the total collective dose of 13 man·Sv/TW·h of electricity generation for each stage of the nuclear fuel cycle by exposure pathway, exposed population and spatial range. Direct exposure of workers and the public from radioactive material (due to routine transportation of material and transportation during decommissioning) contributes about 4% to the total collective dose and is considered to fall into the local category. The local and regional doses to the public, including the population out to a radius of 1000 km, contribute about 3% of the total collective dose.

Ninety per cent of the total dose is due to the modelled global distribution of $^{14}\text{C}$, $^{129}\text{I}$, $^{85}\text{Kr}$ and $^3\text{H}$. Only a very small amount of the global dose is from the distribution of agricultural and marine food products on the worldwide market. The most significant contributor to the global category is the dose from the atmospheric releases of $^{14}\text{C}$. By integrating the average individual doses for 100 000 years into the future, the uncertainty of the results increases significantly. However, it can be assumed that all of the $^{14}\text{C}$ released from current facilities will have been taken into account (the half-life of $^{14}\text{C}$ is 5710 years). If shorter cut-off times are considered, 65%, 22% and 9% of the total dose of 12 man·Sv/TW·h would be accounted for in 10 000 years, 1000 years and 100 years, respectively.

The relevance of this global dose value must be considered carefully because it is the result of integration of many, extremely small, average individual doses (about $2 \times 10^{-9}$ mSv/a per TW·h) for a large population group over a long period of time (100 000 years). Thus, even though the value of the collective dose is high compared with the other values of the assessment, the actual risk to individuals is very small. To put these values in perspective, one can compare the average individual dose due to the naturally occurring atmospheric $^{14}\text{C}$ of 12 µSv/a with the average individual dose from $^{14}\text{C}$ due to the total annual production of nuclear energy in France (350 TW·h) of 0.001 µSv/a. The average annual individual dose from all sources of natural background radiation is 2.4 mSv/a.

Although the effects of radiation have been studied for many decades, large uncertainties about the form and magnitude of the dose response function at these low doses still exist and compound the uncertainty of the environmental model and the input data used. For this reason, it has been proposed to consider these levels of individual doses as being negligible. This consideration must be a part of the decision making process.
4. CONCLUSIONS

The pertinent boundaries of the assessment must be defined initially to take into account the important issues of concern to society. In many cases, this is not done before the risk assessment work is completed. Therefore, a presentation of the distribution of the results as a function of time and space (see Fig. 2) provides a very important input into the process of finding an optimum solution. Direct use of the global dose, as discussed in this paper, clearly represents a dilemma for those decision makers who need to consider all potential risks, but who are faced with imperfect methods that give unbalanced impressions of the actual risks. A method for integration of the uncertainty of the results and the level of individual risk involved is still missing in this type of evaluation, and further work on these issues must continue.

REFERENCES


LONG TERM EXPOSURE OF THE POPULATION OF THE RUSSIAN FEDERATION AS A CONSEQUENCE OF THE ACCIDENT AT THE CHERNOBYL NUCLEAR POWER PLANT

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St. Petersburg, Russian Federation

Abstract

LONG TERM EXPOSURE OF THE POPULATION OF THE RUSSIAN FEDERATION AS A CONSEQUENCE OF THE ACCIDENT AT THE CHERNOBYL NUCLEAR POWER PLANT.

Seventeen regions of Russia were contaminated after the Chernobyl accident in April 1986. Two and a half million persons live in areas contaminated with $^{137}$Cs above 0.04 MBq/m$^2$ (1 Ci/km$^2$). The paper describes the main consequences of the long term exposure of this population. The influence of some natural, human and social factors on the forming of the external and internal doses of rural and urban populations has been studied in Bryansk and Tula, the most contaminated regions of Russia, during 1986-1994. Radioecological processes of migration of caesium and strontium nuclides in the biosphere are considered. A model of their intake into the human body has been developed and validated by large scale measurements of the human body content. Also, a model of external exposure of different population groups has been developed and confirmed by a series of individual external dose measurements with thermoluminescent dosimeters. General dosimetric characteristics of the exposure of the population are given, together with some samples of accumulated external and internal effective doses of inhabitants of some settlements of the Bryansk and Tula regions in 1986–1994.

1. INTRODUCTION

The Chernobyl nuclear power plant (NPP) is located in Ukraine, 150 km south-west of the nearest border of Russia. After the Chernobyl accident in 1986, the western part of the Bryansk region was most intensively contaminated with radio-nuclides. Dose monitoring and a system of countermeasures (supply of non-contaminated food, decontamination of settlements, special agricultural measures, etc.) were implemented in this contaminated area. Contamination was lower in some districts of Tula, Kaluga and Orel, and in 13 other regions of Russia (Fig. 1). In 1991, the area with a contamination of the soil by $^{137}$Cs above 0.04 MBq/m$^2$ (1 Ci/km$^2$) was more than 57 000 km$^2$, with a population of about 2.5 million,
according to data from the Russian Hydrometeorological Service. In the Bryansk region, 112,000 persons lived in 1986 in an area of 2,400 km$^2$ with a contamination of more than 0.6 MBq/m$^2$ (15 Ci/km$^2$).

The Bryansk–Byelorussian 'radioactive spot', centred 200 km north-north-east of the Chernobyl NPP, was created on 28–30 April 1986 as a result of rainfall at the interface of the Bryansk region of Russia and the Gomel and Mogilev regions of Byelorussia. The soil surface activity of $^{137}$Cs ($\sigma_{137}$) in the most contaminated areas reached 3–5 MBq/m$^2$. Another 'caesium spot' — the Kaluga-Tula-Orel 'spot' in Russia, with its centre about 500 km north-east of the Chernobyl NPP — was also due to rainfall from the same radioactive cloud that passed the Bryansk–Byelorussian 'spot'. In the Kaluga–Tula–Orel region the deposition levels from the depleted cloud were lower, and $\sigma_{137}$ was below 0.6 MBq/m$^2$.

The composition of radionuclides released from the Chernobyl NPP to the atmosphere did not only considerably differ from the composition of the nuclear reactor contents but also varied in time, as far as the temperature and the conditions of the release changed. During the release period, the weather conditions changed repeatedly, causing distinctions in the radioactive fluxes in different directions and even in the initial nuclide composition. The basic cause of further separation of the radioactive mixture in the cloud was the different deposition rate of aerosol particles of different dispersivity and density [1–4].

**FIG. 1.** Caesium-137 contamination on the territory of the former Soviet Union: dotted areas >5 Ci/km$^2$ (0.2 MBq/m$^2$), dark areas >15 Ci/km$^2$ (0.6 MBq/m$^2$).
TABLE I. ISOTOPIC COMPOSITION OF RADIOACTIVE RELEASE FROM CHERNOBYL AND FALLOUT IN RUSSIA [1, 2, 4]°

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Total release (Ref. [4])</th>
<th>'Caesium spots' in Russia (&gt;100 km)b</th>
</tr>
</thead>
<tbody>
<tr>
<td>I-131</td>
<td>20</td>
<td>10–14</td>
</tr>
<tr>
<td>Te-132</td>
<td>5</td>
<td>(13)</td>
</tr>
<tr>
<td>Cs-134</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>Cs-137</td>
<td>1.0</td>
<td>1.0</td>
</tr>
<tr>
<td>Sb-125</td>
<td>—</td>
<td>0.03–0.07</td>
</tr>
<tr>
<td>Ag-110m</td>
<td>—</td>
<td>0.005–0.014</td>
</tr>
<tr>
<td>Ru-103</td>
<td>2.0</td>
<td>1.7–2.0</td>
</tr>
<tr>
<td>Ru-106</td>
<td>0.4</td>
<td>0.5–1.4</td>
</tr>
<tr>
<td>Ba-140</td>
<td>2.0</td>
<td>0.7–1.1</td>
</tr>
<tr>
<td>Sr-89</td>
<td>1.0</td>
<td>0.2–0.3</td>
</tr>
<tr>
<td>Sr-90</td>
<td>0.10</td>
<td>0.01–0.03</td>
</tr>
<tr>
<td>Y-91</td>
<td>—</td>
<td>0.06</td>
</tr>
<tr>
<td>Zr-95</td>
<td>2.0</td>
<td>0.03–0.11</td>
</tr>
<tr>
<td>Mo-99</td>
<td>—</td>
<td>(0.11)</td>
</tr>
<tr>
<td>Ce-141</td>
<td>2.3</td>
<td>0.07–0.16</td>
</tr>
<tr>
<td>Ce-144</td>
<td>1.6</td>
<td>0.04–0.15</td>
</tr>
<tr>
<td>Np-239</td>
<td>20</td>
<td>(0.6)</td>
</tr>
</tbody>
</table>

° $\sigma_r/\sigma_{137}$ on 26 April 1986 (relative units).

Figures in brackets are estimated from indirect data.

Table I presents data on the isotopic composition of radioactive release from Chernobyl and fallout in the 'caesium spots' in Russia, giving the ratio of the soil surface activity $\sigma_r$ of the r-th radionuclide to the surface activity $\sigma_{137}$ of the most radiologically significant nuclide of the Chernobyl accident, $^{137}$Cs [1, 2]. The list of radionuclides is divided into three groups: volatile (isotopes and compounds of I, Te, Cs, etc.), intermediate (Ru, Ba, Sr) and refractory non-volatile (Zr, Ce, Pu, etc). It is pointed out that there is a considerable difference in the relative content of refractory radionuclides in the fallout in the far zone, compared with the composition of the release.
TABLE II. MAIN RADIATION FACTORS AND NUCLIDES OF THE CHERNOBYL ACCIDENT AND THEIR IMPACT ON THE RUSSIAN POPULATION

<table>
<thead>
<tr>
<th>Time after the accident (days)</th>
<th>External exposure</th>
<th>Internal exposure</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\beta$</td>
<td>$\gamma$</td>
</tr>
<tr>
<td></td>
<td>$^{106}$Ru/Rh</td>
<td>$^{132}$Te/I</td>
</tr>
<tr>
<td></td>
<td>$^{132}$Te/I</td>
<td>$^{131}$I</td>
</tr>
<tr>
<td></td>
<td>$^{134,137}$Cs</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$^{106}$Ru/Rh</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Transuranium elements (Pu, Am, Cm).

Taking into account the radionuclide composition of the fallout, we present in Table II basic radiation factors having an impact on the Russian population during different periods after the Chernobyl accident. The table presents basic nuclides or groups of nuclides responsible for the given radiation factors. Regarding ingestion, only the radionuclides absorbed in the alimentary tract are given.

The population of the contaminated area has been subjected to exposure during many years. In view of the long term dose accumulation, the contribution to the dose of the exposure from the radioactive cloud is insignificant compared with the external exposure from deposited radionuclides: according to the available measurement data, it is less than 10% of the dose during the first year. The contribution to the dose of inhalation of aerosols and gases from the initial cloud and of resuspended radionuclides from wet deposition is also insignificant compared with that from ingestion of I, Cs and Sr. It is necessary to take into account the external exposure of the skin to high energy beta radiation in the first months after the accident. Later on, as the radionuclides decayed and migrated deeper into the soil, the role of this factor decreased. The leading factor of internal exposure during the first month after the accident was incorporation of $^{131}$I, especially into children drinking milk (not considered here). Later on, caesium radioisotopes ($^{137}$Cs and $^{134}$Cs) played the leading role in internal exposure of the population. Their intake into the body in the first months is determined by surface contamination of the environment and later it depends strongly on the properties of the local soil and on the time after contamination. The contribution of $^{90}$Sr to the internal dose does not exceed 5% and that of transuranium nuclides (Pu, Am, Cm) is less than 0.5%.
2. DOSIMETRY OF EXTERNAL EXPOSURE

2.1. Measurement data

For the evaluation of the doses to the population of the contaminated areas, we have used the results of hundreds of thousands of measurements of the dose rate in different periods after the accident, 1 m above the virgin soil, in typical plots of settlements, including residential, industrial and social buildings; the results of a poll, involving approximately one thousand inhabitants of the Bryansk region in Russia, regarding their mode of behaviour during different seasons; the results of over 300 analyses of $^{137}$Cs and $^{134}$Cs in profiles of virgin soil taken in 1986-1994; and the results of over five thousand measurements of individual doses to inhabitants obtained with the thermoluminescent (TL) method [5, 6]. Golikov [5] exposed anthropomorphous phantoms of people of different ages, ranging from one year to adult, containing TL detectors in many organs, to conditions of exposure to $^{137}$Cs and $^{134}$Cs outdoors and indoors, in order to determine the conversion factors from the exposure to the organ and effective doses.

2.2. The deterministic model

A model of the exposure of people of different ages and social groups has been developed on the basis of experimental investigations in 1986-1994 [5, 6]. The average annual effective dose $E_k$ in the k-th group of inhabitants of a settlement depends on the dose rate in air at a height of 1 m above an open plot of virgin soil in this settlement and in its vicinity, $\dot{D}(t)$; the location factor $LF_i$ is equal to the ratio of the dose rate at the i-th typical plot in the settlement to $\dot{D}(t)$; the behaviour factor $BF_{ik}$ is equal to the time spent during a year at the i-th plot; and the conversion factor $CF_{ik}$ from the absorbed dose rate in air to the effective dose:

$$E_k (\mu Sv) = \int_0^{365} \dot{D}(t)dt \sum_i LF_i BF_{ik} CF_{ik}$$

(1)

In April–June 1986, short lived radionuclides contributed largely to the dose rate. This contribution was about 80% in May 1986, 40% in June 1986 and about 40% over the whole year 1986. Since autumn 1986, in Russia the dose rate was determined by gamma radiation of $^{134}$Cs and $^{137}$Cs. The isotopic composition of fallout and the initial dynamics of the dose rate are well known [1, 2, 5] and have been used in the model. The dose rate in air $\dot{D}(t)$ is considered to be the sum of the dose rate from short lived and medium lived radionuclides and from long lived Cs radionuclides:

$$\dot{D}(t) (\mu Gy/h) = \dot{D}_{sm}(t) + \dot{D}_{Cs}(t)$$

(2)
\[ D_{\text{sn}}(t) = \sigma_{137}^0 \sum_r \sigma_r^0 / \sigma_{137}^0 \, d_r^0 \exp \left(-\ln 2 \, t/T_r\right) \]  

(3)

where \( \sigma_{137}^0 \, (\text{kBq/m}^2) \) is the average surface activity of \( ^{137}\text{Cs} \) on soil in the locality on the day of the accident (1986-04-26); \( \sigma_r^0 \, (\text{kBq/m}^2) \) is the same quantity for a radionuclide \( r \) with a decay half-period \( T_r \); \( d_r^0 \, (\text{pGy/h)/(Bq/m}^2) \) is the dose rate coefficient equal to the initial dose rate in air 1 m above the ground, created by a thin plane source of 1 kBq/m\(^2\) of radionuclide \( r \) in soil for dry or wet fallout.

After decay of the short lived radionuclides and initial intensive migration of \( ^{134,137}\text{Cs} \) in the environment of settlements, the further dynamics of the dose rate is determined mainly by decay and penetration of caesium radionuclides in soil. Golikov and Jacob have investigated about 300 soil samples during eight years (Fig. 2) and used these data in a two-exponential expression for the average dose rate of \( ^{134,137}\text{Cs} \) gamma radiation in an open area:

\[ D_{\text{Cs}}(t) \, (\mu\text{Gy/a}) = \sigma_{137}^0 \left( d_{137}^0 \exp(-\ln 2 \, t/30) + 0.54 \, d_{134}^0 \exp(-\ln 2 \, t/2.1) \right) \, \text{AT}(t) \]  

(4)

\[ \text{AT}(t) \, (\text{rel. units}) = 0.42 \exp(-0.16 \, t) + 0.26 \]

where 0.54 (rel. units) is the average ratio of the \( ^{134}\text{Cs} \) to \( ^{137}\text{Cs} \) activities in the Chernobyl fallout; 30 and 2.1 years are the half-periods of \( ^{137}\text{Cs} \) and \( ^{134}\text{Cs} \) decay, respectively; and \( \text{AT}(t) \, (\text{rel. units}) \) is the attenuation function for the dose rate in air due to migration of Cs radionuclides in soil. The second term of \( \text{AT}(t) \) is obtained with the help of data from Ref. [7].

During eight years after the Chernobyl accident, location factors \( \text{LF}_i \) have been determined with the help of multiple measurements. To estimate behaviour factors \( \text{BF}_{ik} \), the results of a population poll have been used. A set of model (1) parameters for Russia is presented in Table III in terms of generalized dose reduction factors

\[ \text{RF}_k = \sum_i \text{LF}_i \, \text{BF}_{ik} \]

for different population groups. Appropriate conversion factors \( \text{CF}_{ik} \) have been calculated for different age groups and typical locations from the results of the above mentioned experiments with phantoms. These conversion factors are in the range of 0.7–0.8 Sv/Gy for adults, 0.85–0.95 Sv/Gy for children of five years, and about 1 Sv/Gy for children of 1 year. About five thousand measurements of individual doses of inhabitants of the Bryansk region with the TL technique in 1986–1994 [6] allowed the developed model and its parameters to be verified. The results of TL
FIG. 2. Long term time dependence of the dose rate in air 1 m above undisturbed soil, measured in the Bryansk, Orel and Tula regions of Russia (1), according to the UNSCEAR-88 model (2) and according to Gale, 1964 (3). Data from Ref. [7] have been used for calculating the values for curve (1) after 24 years.

measurements showed a linear statistical connection of the dose with the soil contamination by $^{137}\text{Cs}$.

2.3. Actual levels of population exposure

Table IV presents, as an example, values of the accumulated average effective dose E of inhabitants of contaminated areas of Russia, normalized to the average surface activity of $^{137}\text{Cs}$ on soil in a settlement and in its vicinity. These data take into account the isotopic composition of fallout in the 'caesium spots' of the Bryansk-Byelorussian and Kaluga-Tula-Orel regions presented in Table I. The data of Table IV refer to the so-called representative group of the population [5, 6], which reflects the typical age and socio-professional composition of the urban and rural population of the western region of Russia, and the typical structure of housing resources. For more specific groups of inhabitants, the average dose calculated using the data of Table IV should be multiplied by a factor of 0.6 to 1.7.
### TABLE III. DOSE REDUCTION FACTORS RF$_k$ FOR RURAL AND URBAN POPULATIONS IN RUSSIA

<table>
<thead>
<tr>
<th>Population</th>
<th>Time after the accident</th>
<th>Population groups</th>
<th>Representative group</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Outdoor</td>
<td>Indoor</td>
</tr>
<tr>
<td>Rural</td>
<td>First year</td>
<td>0.41$^a$/0.34$^b$</td>
<td>0.32/0.24</td>
</tr>
<tr>
<td></td>
<td>Subsequently</td>
<td>0.36/0.31</td>
<td>0.26/0.22</td>
</tr>
<tr>
<td>Urban</td>
<td>First year</td>
<td>0.29/0.23</td>
<td>0.23/0.15</td>
</tr>
<tr>
<td></td>
<td>Subsequently</td>
<td>0.25/0.20</td>
<td>0.20/0.13</td>
</tr>
</tbody>
</table>

$^a$ Wooden houses.

$^b$ Brick houses.

After evacuation of the inhabitants of the 30 km zone, the villages located in the Bryansk-Byelorussian ‘spot’ became the most contaminated inhabited areas. The average dose to inhabitants of the contaminated area during the past nine years was from 1.4 mSv for $\sigma_{137} = 0.04\text{ MBq/m}^2$ (1 Ci/km$^2$) to 140 mSv for $\sigma_{137} = 4\text{ MBq/m}^2$ (Bryansk region). The dose to separate groups of the population varies by 0.6–1.7 times and the dose to individuals varies by up to a factor of three (up as well as down). According to our prognosis, the dose to the population during 9 years was about 50% of the dose expected for 70 years after the accident.

3. DOSIMETRY OF INTERNAL EXPOSURE

3.1. Radioecological data

The process of dose formation from internal exposure of the population due to intake of long lived $^{137}\text{Cs}$, $^{134}\text{Cs}$ and $^{90}\text{Sr}$ through food-chains can be divided into the stage of surface contamination of vegetation in spring and summer 1986 and the stage of system (root) transfer of these nuclides from soil to plants since summer and autumn 1986. The parameters of deposition and migration at the first stage strongly depend on the state of vegetation and the weather conditions, and those at the second stage depend on the agrochemical properties of soil.

In May 1986, measurements were made which showed that the maximum concentration of $^{134,137}\text{Cs}$ and $^{89,90}\text{Sr}$ in milk was reached in 1–2 weeks after the accidental contamination of the area. The maximum values of the transfer factor (TF) from soil to milk were 0.04 m$^2$/kg for Cs isotopes and 0.006 m$^2$/kg for Sr isotopes.
**TABLE IV. RECONSTRUCTION OF THE AVERAGE EFFECTIVE DOSE IN A REPRESENTATIVE GROUP OF ADULT INHABITANTS OF RUSSIAN SETTLEMENTS AFTER THE CHERNOBYL ACCIDENT**

<table>
<thead>
<tr>
<th>Exposure</th>
<th>Population group and soil type</th>
<th>( E(nSv)/\alpha_{137}^0, (Bq/m^2) )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>2 months</td>
</tr>
<tr>
<td>External</td>
<td>Urban</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.9</td>
</tr>
<tr>
<td></td>
<td>Rural</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>6.4</td>
</tr>
<tr>
<td>Internal</td>
<td>Turf-podzol</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>40</td>
</tr>
<tr>
<td></td>
<td>Black soil</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>27</td>
</tr>
</tbody>
</table>

Later, the TF values decreased with half-periods of 17 and 14 days, respectively, reflecting mainly the rate of natural decontamination of meadow grasses after their surface contamination.

Since autumn 1986, the soil-plant system is the most variable link of the food-chain, which determines protracted incorporation of Cs and Sr radionuclides into the human body from vegetable and animal products. The parameters of Cs and Sr transfer in subsequent links do not vary considerably. The types of dominating soils in various regions of radioactive contamination differ considerably: black earths and grey forest soils in the Tula and Orel regions and turf-podzol soils in the Bryansk region. Depending on the type of soil, the average value of the Cs transfer factor to grass varies by almost three orders of magnitude. The transfer factor for \(^{90}\)Sr from soil to natural grasses varies within narrower limits, namely up to 60 times [8].

Besides the soil properties, the process of natural fixation of Cs radionuclides in soil structures with time after the radioactive contamination of the area considerably influences their content in agricultural products. Figure 3 illustrates the dynamics of the transfer factor for \(^{137}\)Cs from soil to natural grasses and to milk of cows grazing in natural pastures. The specific activity of \(^{137}\)Cs in grass, milk and meat of cattle have decreased from 1987 to 1991-1992 by one to two orders of magnitude. A similar decrease, with a half-period of 0.6-2 years and an average of 1.2 years, has been observed in the content of \(^{137}\)Cs in agricultural products from ploughed soils: cereals, potatoes, etc. Along with the annual decrease, the content of radionuclides in milk and meat varies considerably during the different seasons, depending on the rations of cattle; it increases during the pasture period and decreases when cattle are fed fodder root crops and mixed fodder during the stalled period (see Fig. 3). Since 1991-1992, a tendency of slowing down of the process of natural decontamination of agricultural products has been observed, mainly in the black earth zone.
FIG. 3. Average $^{137}$Cs concentration in milk ($C_m$) from collective farms of the Bryansk region for soil surface activities of (a) 0.2–0.6 MBq/m$^2$; (b) 0.6–1.5 MBq/m$^2$ and (c) >1.5 MBq/m$^2$. (1 Ci = $3.7 \times 10^{10}$ Bq.)

3.2. Measurements of $^{134,137}$Cs in the human body

Besides the use of radioecological data, monitoring of the internal exposure of the population included over 300,000 measurements of the $^{134}$Cs and $^{137}$Cs content in the bodies of inhabitants of all ages in more than 100 Russian settlements; these measurements were made in 1986–1994 by means of stationary and transportable whole body counters. As a rule, a sample consisted of 10–100 persons of the following age groups: children under 7, school children (7–17 years) and adults. Figure 4 shows an example of the time dependent average $^{137}$Cs content in the bodies of adult inhabitants in the village Novye Bobovichi in the Bryansk region.

3.3. Dosimetric model

In general, the expected effective dose of internal exposure E from Cs and Sr radionuclides to different groups of inhabitants has been calculated using their intake with food I(t) during the investigated period ($t_1$, $t_2$) by means of dose factors $d_k$ [9]:
FIG. 4. Average content of $^{137}$Cs in adults (village of N. Bobovichi, Bryansk region).

$$E(t_1, t_2) = \sum_r d_k \int_{t_1}^{t_2} I_r(t) dt$$  \hspace{1cm} (5)

The function of Cs radionuclide intake by adult rural inhabitants of Russia with food, mainly milk, can be modelled as a combination of four exponential components, with half-periods of 2 days, 2 weeks, 1-1.5 years and 10 years (see Fig. 5). The first half-period is characterized by the rate of Cs excretion with milk by the cow; the second one by decrease of surface Cs contamination of grass; the third one by the rate of Cs fixation in soil structures (which is observed at present); and the fourth one by the rate of decrease of Cs transfer to vegetation (this is expected according to previous observations of the consequences of global fallout). The contributions of the different components have been obtained from the results of actual measurements. The intake function is presented in Fig. 5 for regions with turf-podzol sandy soils and black-earth soils. The difference in the intensity of Cs transfer
through roots causes the difference in the effective dose during 70 years by a factor of six [8].

In the most contaminated areas of Russia, measures for radiation protection of the population have been widely taken, beginning in May 1986; these included prohibition of consumption of local food products, mainly dairy, meat and natural products, and supply of non-contaminated food products. These measures considerably decreased the intake of radionuclides into the body. The degree of decrease of intake in different settlements varies widely and is characterized by factors of from 1.5 to 15. Therefore, in the most contaminated area, the correct method of assessing the current dose in inhabitants is measurement of the $^{134}\text{Cs}$ and $^{137}\text{Cs}$ content in the
body, $Q$, and calculation of the actual dose during the period investigated, taking into account seasonal variations:

$$E(t_1, t_2) = \sum_r k_d \int_{t_1}^{t_2} Q_r(t)dt$$  \hspace{1cm} (6)

### 3.4. Actual levels of population exposure

The maximum content of $^{134,137}$Cs in the bodies of inhabitants of the Chernobyl zone was reached in the first months after the accident. In summer 1986, the average content of these nuclides in adult inhabitants of some villages in Russia was 0.3–0.5 MBq and the individual content was up to 4 MBq. In the town of Novozybkov in the Bryansk region, with a population of 46,000, the average $^{134,137}$Cs content in the bodies of adults was about 0.06 MBq in summer 1986. Later, in the settlements where non-contaminated food products were supplied, the content of radiocaesium decreased at first with a period of less than one year and then with a period of 1–1.5 years. This process slowed down considerably after 3–4 years. In the villages where the consumption of local food products continued, the ‘fast’ part of the process was absent, and slowing down of the decrease took place in 3–5 years.

Table IV shows in a generalized form assessments of the average effective dose from internal exposure of adult inhabitants of Russian settlements during different periods after the Chernobyl accident. The assessments are based on the above mentioned radioecological data and the dosimetric model (Eq. (5)) for conditions of wet fallouts without consequent countermeasures, as applied to the areas with dominating turf–podzol sandy and black-earth soils. The contribution of external and internal exposure to the total effective dose in different natural conditions can be seen from these data. According to our data, the dynamics of external exposure actually does not depend on soil type. In the absence of protective measures, the contribution of internal exposure to the total dose during the first year is higher than that of external exposure for both soil types. Already during the first year, the higher Cs transfer factor to vegetation in poor soils influences the value of the dose. Later, internal exposure was higher than external exposure in the areas with turf–podzol soil; in black earth, $^{137}$Cs is strongly fixed and only small amounts are taken up by people through the food-chain.

The actual doses to the population of an area in which countermeasures against internal exposure have been applied are considerably lower than those calculated using the data given in Table IV. As an example, Table V presents the accumulated doses of inhabitants of a number of settlements in Russia, obtained by repeated individual dosimetric monitoring. From an analysis of Table V it is seen that the dependence of the effective dose on the $^{137}$Cs soil surface activity is not definitive.
TABLE V. AVERAGE EFFECTIVE EXTERNAL DOSE (γ), INTERNAL DOSE FROM $^{134,137}$Cs AND $^{89,90}$Sr, AND TOTAL DOSE (Σ)

<table>
<thead>
<tr>
<th>Region (soil type)</th>
<th>Settlement</th>
<th>$\sigma^0$ (MBq/m$^2$)</th>
<th>Doses (mSv) for 1986–1992</th>
<th>External</th>
<th>Internal</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bryansk (Turf-podzol)</td>
<td>Novozybkov</td>
<td>0.7 0.02</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Zaborie</td>
<td>4.4 0.04</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Svyatsk</td>
<td>1.6 0.02</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Usherpie</td>
<td>0.7 —</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Unccha</td>
<td>0.5 —</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Tula (black soil)</td>
<td>Plavsk</td>
<td>0.5 0.01</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Rakhmanovo</td>
<td>0.4 —</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Kamynino</td>
<td>0.3 —</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

An almost linear relationship exists between $\sigma_{137}$ and the dose from external gamma radiation. On the contrary, the dose from internal exposure strongly depends on ecological and social factors. The greatest influence of soil properties is seen from a comparison of the internal dose (Cs) to inhabitants of the Bryansk region, where turf-podzol soil prevails, with the dose to inhabitants of the Tula black-earth region. However, the relation between the doses from internal and external exposure within the Bryansk region is from 15–20% to 50–70%, depending on specific ecological and social conditions.

REFERENCES


ESTIMATION OF RADIATION DOSES TO POPULATION GROUPS IN THE BRYANSK AREA FOLLOWING THE CHERNOBYL ACCIDENT

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Abstract

ESTIMATION OF RADIATION DOSES TO POPULATION GROUPS IN THE BRYANSK AREA FOLLOWING THE CHERNOBYL ACCIDENT.

Since 1990, a joint Russian–Nordic project has been carried out to determine the absorbed doses to selected population groups in the Bryansk region of Russia. The measurement programme involved a total of about 500 persons, living in six different villages, with contamination levels ranging from 0.8 to 2.3 MBq $^{137}$Cs/m². Most of the villages were decontaminated to various degrees, and some villages were not decontaminated. Each September of the years 1990–1994, measurements of individual external doses were carried out using thermoluminescence dosimeters. Since 1991, individual urine samples have been collected annually from about 150 persons and have been used for estimating the body burden of $^{137}$Cs and the internal dose contribution. Adults working outdoors and indoors, and groups of school children were included in the study. Dose measurements using anthropomorphic phantoms were performed in order to determine the relationship between several parameters that are important for determining the external dose in contaminated areas. The reduction of the Chernobyl component of the external dose was 10–30% per year in the period from September 1990 to September 1994. The most heavily contaminated villages were most effectively decontaminated and the effect of this decontamination is reflected in the dose data. The results also reflect the differences between doses to people with indoor occupations and doses to people with outdoor occupations, as well as the differences between doses to persons living in wooden
houses and doses to persons living in brick houses. Individuals with indoor occupations received a lower effective dose from external irradiation than those with outdoor occupations. The doses to people living in brick houses were lower than those to people living in wooden houses. The levels of $^{137}$Cs in urine decreased somewhat between 1991 and 1992, but during 1993 they increased, which is probably due to a larger consumption of locally produced and wild growing food.

1. INTRODUCTION

Because of rainfall in the Bryansk region in Russia at the time of the Chernobyl accident, this region, with a population of about 270 000 inhabitants, was heavily affected by the radioactive fallout from this accident. The population in the area was exposed to both external and internal irradiation, mainly from $^{134}$Cs and $^{137}$Cs, which are the dominating sources of irradiation of people after the decay of short lived radionuclides. Since 1990, a Russian (former Soviet)–Nordic co-operation project has been carried out to determine the individual doses to selected population groups in the Bryansk area. The aim of the present study was to make an independent estimate of the levels of internal and external irradiation of various population groups, to study the effects of decontamination and to determine time variations of dose values.

2. MATERIAL AND METHODS

The measurement programme involved a total of 500 persons, living in six different villages, with various contamination levels, ranging from 0.8 to 2.3 MBq $^{137}$Cs/m$^2$. Most of the villages were decontaminated to various degrees, and some villages were not decontaminated. All villages were supplied with food from distant areas to various extents.

Dose measurements of three anthropomorphic phantoms containing dosimeters were performed in order to determine the relationship between several parameters that are important for determining the external dose in contaminated areas. The phantoms, containing thermoluminescence (TL) dosimeters, were placed in various positions indoors and outdoors (in a field). The phantoms simulated an adult weighing 70 kg, a five year old child and a one year old child. Thermoluminescence dosimeters were also placed on the surface of the phantoms and in free air, close to the phantoms. The investigated parameters were the absorbed dose to various organs, the effective dose (according to ICRP 60), the absorbed dose to the surface (personal dosimeter) of the phantom, the air kerma at the place of the phantom, the total deposition of $^{137}$Cs and $^{134}$Cs in the nearest area, and the depth distribution of these radionuclides.
Each September of the years 1990–1994, measurements of the external dose to people of various occupational groups were carried out. The difference in effective dose between people living in wooden houses and those living in brick houses was also studied. The measurements were performed with TL dosimeters based on chips of CaF$_2$ and LiF. The LiF dosimeters were covered with 4 mm polymethylmethacrylate, and the CaF$_2$ dosimeters were placed underneath a 2 mm brass filter. The dosimeters were put in a holder that was worn by the participating persons around the neck during one month. Two dosimeters from each participating laboratory were placed in the holders, and the final dose values for the years 1990–1992 were determined as mean values of the independent readings. For the years 1993 and 1994, only Swedish dosimeters were used, and the results presented here are from Swedish data only. The dosimeters were calibrated in terms of air kerma or absorbed dose to water, and the basic measurement result refers to air kerma values on the surface of the body. The calibration procedures for the different dosimeters are described elsewhere [1, 2].

During the transport from Sweden or Norway to the Bryansk region and from this region back to Sweden or Norway, the dosimeters were stored in lead containers. The TL signals accumulated during transport and storage, before and after the dosimeters were worn by the participating persons, were estimated by using control dosimeters stored permanently in the lead containers, and were subtracted from the total dosimeter reading.

Since September 1991, individual urine samples have been collected annually and have been used for estimating the body burden of $^{137}$Cs and the contribution from the internal dose. Measurements of the same individuals have been performed in four different years at most, in some cases only in one year. The total number of persons included was about 150; these persons lived in different villages. The same samples were measured both in Sweden and in Russia. The concentrations of potassium, $^{137}$Cs and $^{134}$Cs were measured in all samples. The mean values of the caesium concentrations in the samples differed by at most 10% between those measured in Sweden and those measured in Russia.

Part of the consumed food was supplied from distant areas. The influence of the level of deposition on the whole body content of $^{137+134}$Cs was small; no significant relation between these parameters has previously been detected in the different villages [3]. The contents of $^{137}$Cs and potassium in urine were used to calculate the whole body content of $^{137}$Cs according to Falk et al. [4]:

$$Cs_{WB} = A \ (Cs_u/K_u) \ K_{WB}$$

where $Cs_{WB}$ is the whole body content of $^{137}$Cs (Bq), $Cs_u$ is the $^{137}$Cs concentration in urine (Bq/g), $K_u$ is the potassium in urine (g/g), $K_{WB}$ is the whole body content of potassium (g), and $A$ is an empirical constant, which is different for children of different ages, for adult females and for adult males (adult males: 3.21; adult females: 2.6; children: 1–6 years: 1.86, 7–12 years: 2.0, and 13–18 years: 2.41).
FIG. 1. Mean effective dose from external irradiation to people in six different Russian villages for each September of the years 1990-1994. The natural background component of 50 µSv was subtracted from the monthly doses. The contamination level in 1992 (in MBq/m²) is given in parentheses for each village. Full decontamination was carried out for Yalovka and Svyatsk, partial decontamination for St. Vishkov and Kusnetz, and none for St. Bobovichi and Veprin.

3. RESULTS AND DISCUSSION

From phantom measurements in three different fields, the ratio of the effective dose to the air kerma at one metre above ground \( (H_E/K_{air}) \) varied between 0.69 and 0.79 Sv/Gy for adults, and between 0.87 and 1.02 Sv/Gy for children. The corresponding alpha values, describing the depth distribution of radioactivity in soil \( (A(x) = A_0 e^{-\alpha x}) \), where \( A_0 \) is the activity at the soil surface), ranged from 0.98 cm\(^{-1}\) to 0. The latter value referred to a uniform distribution (ploughed field). The ratio of the effective dose to the absorbed dose to the body surface \( (H_E/D_{surf}) \) ranged from 0.87 to 0.99 Sv/Gy for adults, and from 0.97 to 1.08 Sv/Gy for children. There were no obvious associations between alpha on the one hand and the \( H_E/K_{air} \) and \( H_E/D_{surf} \) ratios on the other. For two separate indoor measurements, the \( H_E/K_{air} \) ratios were 0.66 and 0.78 Sv/Gy for adults; for the first indoor measurement, these ratios were 0.81 and 0.73 Sv/Gy for children. The corresponding \( H_E/D_{surf} \) ratios were 0.86 and 0.87 Sv/Gy for adults, and 1.01 and 1.02 Sv/Gy for children. The effective dose calculated according to ICRP 60 is normally a few per cent lower than the effective dose equivalent calculated according to ICRP 26.

\(^1\) Gy = 1 J/kg.
FIG. 2. Mean effective dose from external irradiation to people in six different Russian villages for one month in the years 1990–1992, divided by the contamination level of 1992. The levels have been reduced by a background level of 50 μSv. The contamination level in 1992 (in MBq/m²) is given in parentheses for each village. Full decontamination was carried out for Yalovka and Svjatsk, partial decontamination for St. Vishkov and Kusnetz, and none for St. Bobovichi and Veprin.

Figure 1 shows the mean effective dose from external irradiation to people in six different villages for each September of the years 1990–1994. The natural background component of 50 μSv was subtracted from the monthly doses. For two villages (Kusnetz and Veprin) measured annually in four consecutive years, the reduction in the effective dose was between 10 and 30% per year. The low value for the village Yalovka in 1993 is difficult to explain. We suspect that a mistake was made when the dosimeters were distributed.

Figure 2 shows the mean effective dose from external irradiation, divided by the contamination level (μSv/month per MBq/m²) of 1992. Full decontamination was carried out for the most contaminated villages. The villages St. Bobovichi and Veprin were not decontaminated at all; and Kusnetz and St. Vishkov were decontaminated only to a very small extent. These villages also showed higher relative dose values than the other villages.

The results reflect the differences between people with outdoor occupations and people with indoor occupations, as well as between people living in wooden houses and those living in brick houses. Individuals living in wooden houses receive an approximately 50% higher effective dose from external radiation than persons living in brick houses. The effective dose to people spending much of their time outdoors is higher than that to people with indoor occupations.
The whole body content of $^{137}$Cs, calculated from urine measurements, varied from almost zero to 275 kBq. The mean values for the group of persons measured were 21, 7 and 30 kBq for the years 1991, 1992 and 1993, respectively. The corresponding numbers of persons participating each year were 24, 49 and 37; more than 50% of these were children under the age of 18 years. The data for the different age groups are shown in Fig. 3, together with the data for six children who were measured every year from 1991 to 1993. A calculation of the annual effective dose from internal contamination, based on the mean values of internal contamination mentioned above, for a 15 year old child [3, 5] gave 1.1, 0.4 and 1.5 mSv for the years 1991, 1992 and 1993, respectively. The contribution from $^{134}$Cs has been included.

It is evident that the whole body content of $^{137}$Cs differs substantially between different individuals, between groups based on age and sex, and between different years. The economic situation in Russia has deteriorated quickly. Consequently, the villages have received less supplies of food from other, non-contaminated areas. Therefore, people eat more locally produced food, as well as wild growing mushrooms, berries, etc. This is the most probable explanation of the increase in the whole body content of $^{137}$Cs and $^{134}$Cs in 1993—a very good year for mushrooms.

Rough estimates of the external, internal and total effective doses for the villages Kusnetz and St. Bobovichi are presented in Fig. 4. The effective dose from internal contamination was calculated from urine samples, as mentioned above, while the effective dose from external irradiation was calculated by multiplying the value for September by 12. The groups of persons examined for internal and external...
doses in 1992 and 1993 were not exactly the same. The total doses for these years were 2.6 mSv (contribution from internal dose 27%) and 2.9 mSv (48%) for Kusnetz, and 1.5 mSv (19%) and 3.4 mSv (68%) for St. Bobovichi, respectively.

4. CONCLUSIONS

The phantom measurements have shown that the ratio of effective dose to air kerma was between 0.69 and 0.79 Sv/Gy for adults, and between 0.87 and 1.02 Sv/Gy for children, in the case of phantoms placed in a field. For indoor phantoms the ratio was between 0.66 and 0.78 Sv/Gy for adults, and between 0.73 and 0.81 Sv/Gy for children.

Individual external doses during the month of September in the years 1990–1994 have been reduced by 10–30% annually, which is larger than can be expected from physical decay alone. Human activities might have a decontaminating effect, which, together with a possible movement of the radioactivity towards deeper soil layers, may explain the observed reduction. The effect of active decontamination is also clearly demonstrated, since the relative effective doses are lower for decontaminated villages than for villages in which none or very little decontamination was performed.

The whole body content of $^{137}$Cs decreased between 1991 and 1992, but in 1993 it increased, which is probably due to a larger consumption of locally produced food in 1993.
Using data from two villages, the total annual doses from the Chernobyl fallout were estimated to be between 1.5 and 3.4 mSv for the period 1992–1993. The relative contribution of internal contamination to the total effective dose increased from 1992 to 1993.

The measurements performed with Swedish, Norwegian and Russian dosimeters, as well as the measurements of urine samples in Sweden and in Russia showed very good agreement.

REFERENCES


[3] ZVONOVA, I.A., et al., $^{134}$Cs and $^{137}$Cs whole-body measurements and internal dosimetry of the population living in areas radioactively contaminated after the Chernobyl accident (in preparation).


ASSESSMENT OF THYROID DOSES DUE TO $^{131}$I FROM ATMOSPHERIC NUCLEAR WEAPONS TESTS IN NEVADA

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Abstract

ASSESSMENT OF THYROID DOSES DUE TO $^{131}$I FROM ATMOSPHERIC NUCLEAR WEAPONS TESTS IN NEVADA.

About 100 of the atmospheric nuclear weapons tests carried out at the Nevada Test Site (NTS) resulted in off-site detection of radioactive materials. These tests released about 5 EBq of $^{131}$I in the atmosphere, predominantly in 1952, 1953, 1955 and 1957. Radioiodine was deposited across the United States of America, with the highest values immediately downwind of the NTS and the lowest values on the west coast. In the 1980s, three major dose reconstruction studies were undertaken in order to address the assessment of thyroid doses due to $^{131}$I fallout from the NTS: (1) the ORERP study of the US Department of Energy; (2) the Utah thyroid cohort study; and (3) the NCI thyroid study. The first two studies are concerned with doses received by 'local' populations (less than 800 km from the NTS), while the third study deals with the estimation of thyroid doses received by populations across the continental USA. In all three studies, uncertainty estimates were attached to the calculated doses. The second and third studies specifically addressed the assessment of thyroid doses from radioiodines, whereas the first study has a much wider scope, in that it considers both external and internal irradiation of the main organs and tissues of the body from all radionuclides produced by nuclear weapons tests. The thyroid doses from NTS fallout resulted essentially from the ingestion of milk contaminated with $^{131}$I; other, usually less important, pathways of exposure are the consumption of leafy vegetables and eggs. Because children generally drink more milk than adults, and because of the smaller mass of their thyroid gland, children received higher doses than adults for a given deposition of $^{131}$I. The thyroid doses are estimated to have ranged up to a few grays for small children in south-western Utah who drank milk from a family owned goat. A preliminary estimate of the collective thyroid dose to the US population from NTS fallout is $4 \times 10^6$ man-Gy, corresponding to a per capita thyroid dose of about 20 mGy.

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1. INTRODUCTION

Atmospheric testing of nuclear weapons related devices at the Nevada Test Site (NTS) began on 27 January 1951. Atmospheric testing at the NTS continued intermittently until 5 August 1963 when the United States of America signed the Limited Test Ban Treaty (LTBT) prohibiting the testing of nuclear weapons in the atmosphere, under water or in outer space. During the period of atmospheric testing, 105 tests were conducted above the ground surface at the NTS and 14 other tests were conducted below ground at depths where containment was not expected. The total nuclear yield of these explosions was approximately one megatonne of TNT equivalent explosive energy [1]. Most of the atmospheric releases of radioactive materials, including about 5 EBq of $^{131}$I, took place in test series conducted in 1951, 1952, 1953, 1955 and 1957.

In addition, approximately one thousand tests, conducted underground since the 1950s, were designed for containment of radioactive debris; thirty-eight of these had releases of radioactive materials to the atmosphere that were small compared with those of the atmospheric tests, but were of sufficient quantity to be detected by monitoring equipment located off-site [2].

Public concern began to surface in 1953, when several detonations of the Upshot-Knothole test series led to considerable fallout to the north and east of the test site, and numerous sheep died during the spring of 1953 following winter feeding in Nevada north-east of the NTS. An investigation by the US Atomic Energy Commission and the Public Health Service concluded that the sheep deaths were not attributable to radiation [1]. Public concern, however, continued to build up during the late 1950s and early 1960s. Congressional hearings were held in 1957, 1959 and 1963 to evaluate the impact on the public from fallout of nuclear explosions. Although the estimated doses to local populations from external irradiation, reported by Dunning in 1959 [3], were low and of little, if any, local concern, the precise magnitude and uncertainty of those exposures were not well established. Public action groups were formed to seek federal relief for ‘fallout victims’, and congressional representatives from the States of Utah and Nevada began proposing legislative relief for persons suffering from cancers caused by radiation. By the late 1970s, hundreds of damage claims were filed with the US Department of Energy (DOE) alleging that illnesses, primarily cancers, resulted from nuclear testing activities at the NTS. The publication of one epidemiological study [4] implied a causal relationship between radioactive fallout deposition and childhood leukaemia. Public figures and some scientists questioned the reliability of exposure estimates provided by Dunning in 1959, especially since doses from internal irradiation were not considered. These and other related events and concerns prompted the need for a thorough re-evaluation of radiation exposures of the public from fallout produced by nuclear detonations at the NTS (see Refs [1] and [5]). In the 1980s, three major dose reconstruction studies were undertaken in order to address the assessment of thyroid doses due to $^{131}$I fallout from the NTS.
2. DESCRIPTION OF THE THYROID DOSE RECONSTRUCTION STUDIES

The three major thyroid dose reconstruction studies related to NTS fallout are known as: (1) the ORERP study of DOE; (2) the Utah thyroid cohort study; and (3) the NCI thyroid study. The first two studies are concerned with doses received by 'local' populations (less than 800 km from the NTS), while the third study deals with the estimation of thyroid doses received by populations across the continental USA. In all three studies, uncertainty estimates were attached to the calculated doses. The second and third studies specifically addressed the assessment of thyroid doses from radioiodines, whereas the first study has a much wider scope, in that it considers both external and internal irradiation of the main organs and tissues of the body from all radionuclides produced by nuclear weapons tests.

2.1. The ORERP study of DOE

In 1979, DOE established the Off-Site Radiation Exposure Review Project (ORERP) to: (1) collect and organize at one central location all relevant documents and data pertaining to fallout in the off-site area and make these documents available to the public, and (2) produce a dosimetric re-evaluation of the off-site area characterized by region, community/locale and age/occupation [1].

A methodology has been developed by the ORERP to model doses to individuals resulting from specific NTS events. This methodology is based on the establishment of databases containing, for each location of interest, an estimate of the exposure rate at 12 hours after each event (H + 12) together with the estimated time of arrival of the fallout at that location. Measured and calculated relationships involving exposure rate and relative amounts of each fission and activation product for each NTS event as a function of time after the explosion [6, 7] were then used to estimate the deposition density of each radionuclide at each location of interest. These exposure rates and deposition density estimates were used to derive external dose calculations and to estimate doses from internal irradiation by means of environmental transport models.

The first phase of the ORERP, designated as Phase I, was performed at locations within approximately 300 km of the NTS where ground monitoring personnel measured γ exposure rates following nuclear events. These survey meter readings, together with available fallout patterns, were used to compile a Town Data Base with 1950 records involving 74 events at 352 locations [8]. Each record in the Town Data Base contains the values and estimates of uncertainties for the H + 12 exposure rate and for fallout arrival time for an event at a given location.

The second phase of the ORERP, designated as Phase II, extended the geographic areas of interest to include the States of Arizona, New Mexico, Nevada and Utah (excluding areas already included in the Phase I study region), south-eastern California, western Colorado, southern Idaho, south-eastern Oregon and south-western Wyoming. The County Data Base, which is an analogue of the Town Data
Base, was developed for the 142 counties and county segments that are part of the Phase II study region [9]. The County Data Base was constructed from $^{137}\text{Cs}$ deposition density estimates for each of the 142 counties and county segments for each NTS event depositing significant fallout in that county. The $^{137}\text{Cs}$ deposition estimates for each test were derived mainly from the analysis of contemporary soil samples for $^{137}\text{Cs}$ content and for the $^{240}\text{Pu}$ to $^{239}\text{Pu}$ ratio, and from the re-evaluation of the daily deposition of radioactive materials on sticky paper (called gummed film) during the testing period [10, 11].

Doses from internal irradiation were estimated using the PATHWAY model [12]. The modelling approach used state-of-the-art dynamic models and site specific data on agricultural, lifestyle and environmental transport parameters. Where possible, model predictions were tested against real, independent data sets prior to use for this analysis. The computations were carried out for 15,665 location/event combinations, 20 radionuclides, four age classes, 22 organs, and individual and collective dose scenarios. Uncertainties in data and model parameters were propagated to the dose estimates using Monte Carlo simulation techniques. The shapes and magnitudes of uncertainty distributions of data and parameters were estimated from extensive literature searches, elicitation of expert opinion and judgement of the investigators. Induced correlations between certain parameters and calculated quantities were taken into consideration. The concentrations, intakes and doses were typically best described as log-normal distributions. Therefore, medians or geometric means (GM) and geometric standard deviations (GSD) were used as the descriptors for central tendency and dispersion, respectively [13, 2].

The thyroid doses were found to be much greater than the doses to other organs or tissues. Thyroid doses from NTS fallout were due mainly to the consumption of foodstuffs contaminated with $^{131}\text{I}$, with other, relatively minor, contributions resulting essentially from the consumption of foodstuffs contaminated with $^{133}\text{I}$, from external irradiation from fallout activity deposited on the ground, and from inhalation of air contaminated with both $^{131}\text{I}$ and $^{133}\text{I}$. Because children generally drink more milk than adults, and because of the smaller mass of their thyroid gland, children receive higher doses than adults for a given deposition of $^{131}\text{I}$. The consumption of fresh cows' milk usually accounted for most of the dose because milk is consumed regularly and in large amounts and because of its short shelf-life. Other foodstuffs with short shelf-life, such as leafy vegetables, also contributed significantly to the thyroid dose of individuals who consumed little, or no, fresh milk.

2.2. The Utah thyroid cohort study

The Utah thyroid cohort study, which was funded primarily by the National Cancer Institute (with supplemental funding from the Departments of Defence and Energy), was a follow-up to a study conducted in 1965-1970 by the US Public Health Service, in which children living in Washington County, Utah, and Lincoln County,
Nevada, had been examined for the presence of thyroid abnormalities, and children of Graham County, Arizona, had been used as a control group [14]. The Utah thyroid cohort study consisted of locating the same cohort of subjects identified in the 1965–1970 study and of re-examining them for the presence of thyroid neoplasms and other thyroid disease. Altogether, doses were assigned for 3545 subjects, of which 3122 were re-examined [15].

The information necessary to calculate the thyroid doses received from ingestion by all individuals in the cohort included: (1) the deposition densities of $^{131}$I and $^{133}$I; (2) the transfer coefficients from the deposition densities to the radionuclide concentrations in cow’s milk and other important foodstuffs; (3) the dietary and lifestyle habits of all individuals in the cohort; and (4) the thyroid dose coefficients.

The deposition estimates were obtained using the ORERP methodology and the two ORERP deposition databases, supplemented with an additional database, the Other Locations Database, specifically developed for this study. Deposition estimates were ascertained for 5804 locations of subject residences and/or locations of milk producers. The transfer coefficients from the deposition densities to the radionuclide concentrations in cow’s milk, goat’s milk and leafy vegetables were obtained using a suite of models and a survey of dairy management practices [16]. A diet, lifestyle and residence history survey was conducted to obtain specific information on the milk consumption rates, the frequency of consumption of leafy vegetables, the source of milk and vegetables, as well as on residence history. Finally, the age specific thyroid dose coefficients were based on data from the literature.

2.3. The NCI thyroid study

The NCI thyroid study consists, in part, of an assessment of the exposure of the population of the USA to $^{131}$I in fallout originating from the NTS [17]. This study is carried out in response to Public Law 97-414, Section 7(a), which, among other provisions, specifies the following requirements:

"conduct scientific research and prepare analyses necessary to develop valid and credible methods to estimate the thyroid doses of Iodine 131 that are received by individuals from nuclear bomb fallout;"

"conduct scientific research and prepare analyses necessary to develop valid and credible assessments of the exposure to Iodine 131 that the American people received from the Nevada atmospheric nuclear bomb tests."

The estimation of the exposure and thyroid doses received by the population of the USA as a result of $^{131}$I fallout from the NTS has required a significant effort that is nearly complete. Following the recommendations of an advisory committee that was established to assist the NCI in addressing these tasks, thyroid doses are estimated for representative individuals in each of the approximately 3100 counties of the contiguous USA for each event at the NTS that resulted in discernible off-site
fallout. Exposures to $^{131}$I in fallout resulted mainly from the pasture-cow-milk food-chain. Other, less important, exposure routes (inhalation of $^{131}$I contaminated air and ingestion of foodstuffs other than fresh cows’ milk) also are considered, but in a much less detailed manner. In the assessment of the exposures from the pasture-cow-milk food-chain, estimates are made of: (1) the activities of $^{131}$I deposited on soil and vegetation; (2) the amount of $^{131}$I consumed by dairy cows and the resulting $^{131}$I concentrations in cows’ milk; (3) the $^{131}$I ingested by people; and (4) the absorbed doses from $^{131}$I in the thyroids of people [18].

Conceptually, this study is very similar to the ORERP study (as far as the estimation of thyroid doses from $^{131}$I is concerned) and to the dosimetric effort related to the Utah thyroid cohort study. The primary differences among the three studies are that: (1) the NCI fallout study considers the population across the contiguous USA, whereas the other two studies consider only people residing in those States in the vicinity of the NTS; and (2) thyroid doses in the ORERP study and in the NCI fallout study are assessed for representative, unspecified individuals, whereas thyroid doses to identified individuals are estimated in the Utah thyroid cohort study.

Special efforts were made to reconstruct the deposition of $^{131}$I across the USA for each event at the NTS that resulted in discernible off-site fallout. Gummed-film data collected by the Environmental Measurements Laboratory (called at that time the Health and Safety Laboratory) between 1951 and 1958 from the 40-95 monitoring sites located throughout the country at that time were used to calculate estimates of daily depositions of $^{131}$I at those sites [19]. Deposition of $^{131}$I between monitoring sites was estimated by interpolation, using precipitation data and appropriate statistical techniques, especially kriging [20]. For those tests during which the gummed-film network was not in place (i.e. during the Ranger test series in 1951 and for tests carried out after 1958), an atmospheric dispersion and deposition model was developed to provide estimates of $^{131}$I during those tests in the areas of the USA where rainfall coincided with the passage of the fallout cloud [21]. In addition, the ORERP estimates were used for locations in the vicinity of the NTS.

The assessment of the thyroid doses also necessitated information on the pasture practices, milk distribution and consumption in the continental USA in the 1950s. Special surveys, together with extensive literature searches, were carried out for that purpose [22].

For each test and each county, average thyroid doses from $^{131}$I have been estimated for 14 age and sex groups: (1) four foetal ages (0–10 weeks, 11–20 weeks, 21–30 weeks and 31–40 weeks in utero); (2) four infant categories (0–2 months, 3–5 months, 6–8 months and 9–11 months); (3) four child groups (1–4 years, 5–9 years, 10–14 years and 15–19 years); and (4) adult males and adult females. For each of those age and sex groups, four populations are considered: (1) those persons who drank cows’ milk; (2) a specified ‘high exposure’ group that consumed large quantities of cows’ milk with higher than average concentrations of $^{131}$I; (3) a group that drank milk obtained from backyard cows; and (4) a specified ‘low exposure’
group (persons who drank no milk). In addition, the per capita thyroid doses from $^{131}$I are estimated for each test and each county, and the $^{131}$I concentrations in air and in a variety of foodstuffs are provided, so that any individual can estimate his or her own thyroid dose on the basis of residential and dietary habits.

3. THYROID DOSE ESTIMATES

Thyroid doses from NTS fallout received by the populations of Phases I and II of the ORERP have been extensively studied. Anspaugh et al. [23] compiled the estimates of thyroid doses for an infant living in St. George, UT, when the test HARRY occurred on 19 May 1953. St. George was one of the more heavily contaminated communities and the test HARRY accounted for most of the thyroid doses in that area. These historical thyroid dose estimates, which were made by independent researchers in the 1960s, are presented in Table I. It is of interest that these historical values, which were derived from meagre information, are relatively close together, and that they compare well with the modern value of Ng et al. [30], calculated for the ORERP, and with the preliminary estimate of the NCI fallout study, which was added to Table I for comparison purposes.

Table II illustrates both the relative importance of several nuclear weapons tests on the thyroid doses received by individuals living in St. George, UT, and the variation with age of the thyroid doses. It is clear that the test HARRY was responsible for most of the dose in St. George and that infants received thyroid doses that were an order of magnitude greater than those received by adults. The geometric standard deviations attached to the thyroid dose distributions are estimated to range from 1.9 to 2.6 [5].

Estimates of individual thyroid doses for the 3545 subjects considered in the Utah thyroid cohort study are summarized in Table III [15]. The overall mean thyroid dose for the cohort was 98 mGy, with a median dose of 25 mGy. The maximum calculated thyroid dose for any subject was 4600 mGy. For practical purposes, zero doses were assigned to 135 subjects who did not reside within the domain considered between 1951 and 1958.

The importance of the contribution of the consumption of milk to the thyroid dose is illustrated in Table IV. The mean dose among the subjects who did not drink milk was 12 mGy, while the mean dose among the subjects who drank milk was 100 mGy. Of particular importance are the 155 subjects who drank goats' milk at some point in their childhood. The mean dose among this group was 300 mGy, and the highest dose (4600 mGy) was found for an individual in that group. Five subjects received an absorbed thyroid dose greater than 3000 mGy; all of them drank milk from a family owned goat. These data clearly demonstrate the importance of the goat milk pathway [15]. The relative importance of various pathways is presented in Table V for the residents of the three States considered in the Utah thyroid cohort.
TABLE I. HISTORICAL AND CURRENT ESTIMATES OF THYROID DOSE FOR AN INFANT LIVING IN St. GEORGE, UT, AT THE TIME OF THE TEST HARRY (19 MAY 1953) (Based on Ref. [23])

<table>
<thead>
<tr>
<th>Authors</th>
<th>Thyroid dose (mGy)</th>
<th>Central estimate (range of uncertainty)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mays (1963) [24]</td>
<td>680</td>
<td></td>
</tr>
<tr>
<td>Reiss (1963) [25]</td>
<td>1000-7000</td>
<td></td>
</tr>
<tr>
<td>Pendleton et al. (1963)  [26]</td>
<td>840</td>
<td></td>
</tr>
<tr>
<td>Knapp (1963) [27]</td>
<td>1200-4400</td>
<td></td>
</tr>
<tr>
<td>Tamplin and Fisher (1967) [28]</td>
<td>780 (200-1600)</td>
<td></td>
</tr>
<tr>
<td>Perez and Robinson (1967) [29]</td>
<td>680</td>
<td></td>
</tr>
<tr>
<td>Ng et al. (1990) [30]</td>
<td>660 (200-1900)</td>
<td></td>
</tr>
<tr>
<td>NCI (preliminary)</td>
<td>500 (200-1400)</td>
<td></td>
</tr>
</tbody>
</table>

TABLE II. ESTIMATES OF MEDIAN THYROID DOSES FROM INGESTION (mGy) FOR UNSPECIFIED INDIVIDUALS LIVING IN St. GEORGE, UT, WITH A RURAL LIFESTYLE

<table>
<thead>
<tr>
<th>Event</th>
<th>Date</th>
<th>Infant (0-12 months)</th>
<th>Child (1-11 years)</th>
<th>Teenager (12-18 years)</th>
<th>Adult (&gt;18 years)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HARRY</td>
<td>1953-05-19</td>
<td>840</td>
<td>400</td>
<td>170</td>
<td>51</td>
</tr>
<tr>
<td>SMOKY</td>
<td>1957-08-31</td>
<td>240</td>
<td>100</td>
<td>43</td>
<td>13</td>
</tr>
<tr>
<td>ANNIE</td>
<td>1953-03-17</td>
<td>35</td>
<td>20</td>
<td>9.3</td>
<td>2.7</td>
</tr>
<tr>
<td>ZUCCHINI</td>
<td>1955-05-15</td>
<td>32</td>
<td>15</td>
<td>6.3</td>
<td>1.9</td>
</tr>
<tr>
<td>SIMON</td>
<td>1953-04-25</td>
<td>21</td>
<td>9.3</td>
<td>3.9</td>
<td>1.1</td>
</tr>
<tr>
<td>TESLA</td>
<td>1955-03-01</td>
<td>2.4</td>
<td>2.2</td>
<td>2.3</td>
<td>1.4</td>
</tr>
</tbody>
</table>

a The six events selected were those resulting in the greatest thyroid doses. The geometric standard deviations attached to the dose distributions are estimated to range from 1.9 to 2.6 [5].
TABLE III. SUMMARY OF THYROID DOSES (mGy) FROM THE UTAH THYROID COHORT STUDY [15]

<table>
<thead>
<tr>
<th></th>
<th>Washington County, UT</th>
<th>Graham County, AZ</th>
<th>Lincoln County, NV</th>
<th>Overall</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of subjects</td>
<td>1896</td>
<td>1369</td>
<td>280</td>
<td>3545</td>
</tr>
<tr>
<td>Mean dose</td>
<td>170</td>
<td>13</td>
<td>50</td>
<td>98</td>
</tr>
<tr>
<td>Median dose</td>
<td>72</td>
<td>3.6</td>
<td>28</td>
<td>25</td>
</tr>
<tr>
<td>Minimum</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Maximum</td>
<td>4600</td>
<td>450</td>
<td>840</td>
<td>4600</td>
</tr>
<tr>
<td>Mean GSD</td>
<td>2.7</td>
<td>3.0</td>
<td>2.7</td>
<td>2.8</td>
</tr>
</tbody>
</table>

TABLE IV. COMPARISON OF THYROID DOSES (mGy) OF MILK DRINKERS AND NON-MILK DRINKERS [15]

<table>
<thead>
<tr>
<th></th>
<th>Non-milk drinkers</th>
<th>Milk drinkers</th>
<th>Goats' milk drinkers</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of subjects</td>
<td>120</td>
<td>3337</td>
<td>155</td>
</tr>
<tr>
<td>Mean dose</td>
<td>12</td>
<td>100</td>
<td>300</td>
</tr>
<tr>
<td>Median dose</td>
<td>0.5</td>
<td>30</td>
<td>39</td>
</tr>
<tr>
<td>Maximum dose</td>
<td>25</td>
<td>4600</td>
<td>4600</td>
</tr>
</tbody>
</table>

Another important feature of the Utah thyroid cohort study is the evaluation of the radioiodine exposure while in utero. A total of 480 subjects were exposed in utero during the fallout period. The average dose to the thyroid of the foetus was 39 mGy; this accounted, on average, for about 20% of each of these subjects' thyroid dose [15].

The NCI thyroid study is the only one in which thyroid doses have been estimated for each county of the USA and for each important test that was conducted at the NTS. Several aspects of the methodology used to estimate the thyroid doses are available in the literature [18, 19, 21, 22], but the thyroid dose estimates are still considered to be preliminary. Table VI presents a comparison with the ORERP estimates of preliminary NCI calculations of per capita thyroid doses summed across all
TABLE V. RELATIVE IMPORTANCE OF THE THYROID EXPOSURE PATHWAYS IN THE UTAH THYROID COHORT STUDY [15]

<table>
<thead>
<tr>
<th>Pathway</th>
<th>Utah residents</th>
<th>Nevada residents</th>
<th>Arizona residents</th>
</tr>
</thead>
<tbody>
<tr>
<td>Milk</td>
<td>81%</td>
<td>79%</td>
<td>70%</td>
</tr>
<tr>
<td>Leafy vegetables</td>
<td>11%</td>
<td>15%</td>
<td>23%</td>
</tr>
<tr>
<td>Inhalation</td>
<td>2%</td>
<td>1%</td>
<td>2%</td>
</tr>
<tr>
<td>External irradiation</td>
<td>6%</td>
<td>5%</td>
<td>5%</td>
</tr>
</tbody>
</table>

TABLE VI. COMPARISON OF PER CAPITA THYROID DOSES (mGy) SUMMED ACROSS ALL NTS EVENTS

<table>
<thead>
<tr>
<th>Location</th>
<th>ORERP</th>
<th>NCI (preliminary)</th>
</tr>
</thead>
<tbody>
<tr>
<td>St. George, UT</td>
<td>245</td>
<td>113</td>
</tr>
<tr>
<td>Utah County, UT</td>
<td>125</td>
<td>62</td>
</tr>
<tr>
<td>Las Vegas, NV</td>
<td>113</td>
<td>7.8</td>
</tr>
<tr>
<td>Salt Lake County, UT</td>
<td>61</td>
<td>40</td>
</tr>
<tr>
<td>Weber County, UT</td>
<td>42</td>
<td>42</td>
</tr>
<tr>
<td>Bernalillo County, NM</td>
<td>24</td>
<td>23</td>
</tr>
<tr>
<td>Los Angeles County, CA</td>
<td>0.67</td>
<td>0.93</td>
</tr>
</tbody>
</table>

NTS events for localities in Phases I and II. Most of the NCI preliminary results are similar to those of ORERP, given the large uncertainties attached to the estimation of doses received about 40 years ago. The only notable discrepancy concerns dose estimates for the population of Las Vegas, NV; reasons for this discrepancy are yet to be found. An important result of the NCI fallout study is that deposition of $^{131}$I from NTS fallout occurred at one time or another in every county of the contiguous USA and that it is likely that almost all, if not all, of the people who resided in the contiguous USA between 1951 and 1958 received a thyroid dose from NTS fallout. The highest per capita thyroid doses, in the range from 50 to 110 mGy, are estimated not only for populations in counties of States close to the NTS, such as Nevada and Utah, but also for populations in counties of States relatively far away from the NTS, such as Arkansas, Colorado, Idaho, Kansas, Missouri and Montana. Low per capita
thyroid doses, in the range from 0.01 to 1 mGy, are calculated for counties in southern California, while per capita doses in counties of the east coast are typically of the order of 10 mGy. A preliminary estimate of the collective thyroid dose to the US population from NTS fallout is $4 \times 10^6$ man-Gy, corresponding to a per capita thyroid dose of about 20 mGy.

4. CONCLUSIONS

The thyroid doses resulting from nuclear weapons testing at the NTS, principally from 1951 to 1958, have been extensively studied in three dose reconstruction efforts known as: (1) the ORERP study of DOE, (2) the Utah thyroid cohort study, and (3) the NCI thyroid study. The first two studies are concerned with doses received by 'local' populations (less than 800 km from the NTS), while the third study deals with the estimation of thyroid doses received by 'the people of the USA' across the continental USA. In all three studies, uncertainty estimates associated with the calculated doses were included. The first study had a very wide scope, as it considered external and internal irradiation of the main organs and tissues of the body from all radionuclides produced by nuclear weapons tests, whereas the second and third studies addressed specifically the assessment of thyroid doses from radioiodine.

The thyroid doses from NTS fallout resulted essentially from the ingestion of milk contaminated with $^{131}$I; other, usually less important, pathways of exposure are the consumption of leafy vegetables and eggs. Because children generally drink more milk than adults, and because of the smaller mass of their thyroid gland, children received higher doses than adults for a given deposition of $^{131}$I. The thyroid doses are estimated to have ranged up to a few grays for small children in south-western Utah who drank milk from a family owned goat. A preliminary estimate of the collective thyroid dose to the US population from NTS fallout is $4 \times 10^6$ man-Gy, corresponding to a per capita thyroid dose of about 20 mGy.

REFERENCES


[17] WACHHOLZ, B.W., Overview of the National Cancer Institute's activities related to exposure of the public from fallout from the Nevada Test Site, Health Phys. 59 (1990) 511-514.


RADIATION IMPACT OF NUCLEAR WEAPONS TESTS AT THE SEMIPALATINSK TEST SITE ON THE POPULATION OF THE ALTAI REGION

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Abstract

RADIATION IMPACT OF NUCLEAR WEAPONS TESTS AT THE SEMIPALATINSK TEST SITE ON THE POPULATION OF THE ALTAI REGION.

The population of the Altai region was exposed repeatedly to radiation during atmospheric tests of nuclear weapons from 1949 to 1962 and during cratering explosions in 1965. It has now been established that 22 nuclear explosions resulted in transport of radionuclides towards the Altai region. The nuclear explosion of 29 August 1949 alone produced an effective dose exceeding 250 mSv in 44 settlements. In the case of the nuclear explosion of 7 August 1962, the thyroid dose to persons living in a number of settlements was more than a few gray. The impact of the tests on the population of the Altai region has been under study since 1990 (as of 1992 under the "Semipalatinsk test site/Altai" State research programme). This study covers: reconstruction of doses from the tests, risk assessment, epidemiological studies, health and demographic studies, and ecological research (radioactive and chemical contamination). The goals of the research are development and realization of the programme of additional measures for health protection and compensation, and environmental and social protection. The nuclear test of 1949 produced the greatest radiation impact. The total collective dose from this test to the population of the region is estimated at approximately 32 000 man-Sv, with about 250 000 individuals receiving a dose of above 5 cSv. The radiation impact from this test on the cohort and population levels was estimated using up-to-date scientific theories about stochastic health effects of ionizing radiation. The goal of the research is to obtain data on the consequences of the tests in order to plan social protection measures as well as future
research. An analysis of the medico-ecological situation in the affected and control regions and an epidemiological study are being carried out. Some results of the study are presented in the paper. A concept of social protection for the population affected by the tests has been prepared and serves as the basis for taking decisions on practical measures. Its principal points are described. The problems currently faced in evaluating the consequences of the tests are considered.

1. INTRODUCTION

The Semipalatinsk test site (STS) is situated at the boundary of the Pavlodar, Semipalatinsk and Karaganda regions (Kazakhstan) south-west of the Altai region. The minimum distance from the test site to the Altai region is 150 km.

During 1949–1962, tens of above-ground nuclear weapons tests were conducted on the STS [1]. Some of them caused considerable radiation exposure to the population of the adjoining territories. The available data show that part of the population of the Altai region suffered seriously from these tests [2].

A study of the impact of these tests on the population of the Altai region began in 1990 (since 1992 the study has been carried out in the frame of the State research programme “Semipalatinsk test site/Altai” [3]). This programme includes the following sections:

- reconstruction of doses from the tests,
- risk assessment,
- epidemiological studies,
- health and demographic studies,
- ecological research (radioactive and chemical contamination).

The goals of the investigation are the development and realization of a programme of additional measures for social, health and environmental protection for the population of the Altai region who suffered from the STS tests. The results of the studies of the radiation impact of the STS tests on this population group are presented in the paper.

2. DOSE ASSESSMENT

Assessment of the population doses from nuclear weapons tests began in the 1960s, but only for global fallout. For local fallout, such studies became possible at the beginning of the 1990s — 40 years after the first nuclear test at the STS.

2.1. Methodology

As regards local contamination and population doses, mainly relatively short lived radionuclides are created by nuclear weapons explosions. Over a
few years, the environmental radioactive contamination from the STS tests decreased to an insignificant level. More than 50% of the total effective dose was accumulated during the first few days after a test and 97% in the first year after it [4]. After a test, direct measurements of doses from $\gamma$-irradiation and from the environmental radioactive contamination along the traces of the radioactive fallout were carried out by specialists from laboratories at the STS [1, 5]. Unfortunately, the data from these measurements are not sufficient in many cases for population dose assessment in the Altai region. Therefore, a retrospective dose assessment methodology (RDAM) has been developed and used; this is based on mathematical modelling of

- the creation of radioactive aerosols in a nuclear explosion,
- the transport of radionuclides in the atmosphere, natural media and human body organs.

RDAM is based on the modern physics of explosion, concrete information on a sample of a nuclear weapon tested, atmospheric processes, recommendations of the International Commission on Radiological Protection (ICRP), maximum use of data from direct measurements, a probabilistic approach to the description of some intermediate and final parameters, etc. Details of this methodology are described in Ref. [4] (see also Refs [2, 6, 7]).

2.2. Results

General data on the radiation exposure of the population of the Altai region from the STS are given in Table I, in terms of the collective effective dose $S$. For health risk assessment, more detailed data, in terms of equivalent doses to different human body organs, are calculated with RDAM.

Further results are given for the first test (29 August 1949), which had the greatest radiation impact. This test was conducted without waiting for better meteorological conditions because of the political situation at that time. The numbers of people exposed to different doses are given in Tables II and III. The exactness of these numbers reflects the activities performed in the frame of the epidemiological study (preparation of data for the register of exposed

<table>
<thead>
<tr>
<th>Test</th>
<th>$S \times 10^3$ man-Sv</th>
</tr>
</thead>
<tbody>
<tr>
<td>1949-08-29</td>
<td>32</td>
</tr>
<tr>
<td>1962-08-07</td>
<td>3$^a$</td>
</tr>
<tr>
<td>Others</td>
<td>7$^a$</td>
</tr>
</tbody>
</table>

$^a$ First preliminary estimation results.
TABLE II. RADIATION IMPACT OF THE FIRST TEST AT THE STS ON THE POPULATION OF THE ALTAI REGION (DATA ESTIMATED WITH RDAM)

<table>
<thead>
<tr>
<th>$D_E$ (Sv)</th>
<th>Number of people exposed</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Males</td>
<td>Females</td>
</tr>
<tr>
<td>$&gt; 0.5$</td>
<td>4 649</td>
<td>5 695</td>
</tr>
<tr>
<td>0.25–0.5</td>
<td>4 908</td>
<td>6 343</td>
</tr>
<tr>
<td>0.13–0.25</td>
<td>1 452</td>
<td>1 975</td>
</tr>
<tr>
<td>$&gt; 0.13$</td>
<td>11 009</td>
<td>14 013</td>
</tr>
<tr>
<td>$&gt; 0.05$</td>
<td>250 000</td>
<td></td>
</tr>
</tbody>
</table>

TABLE III. DISTRIBUTION OF AGE GROUPS OF THE POPULATION AT EXPOSURE (TIME OF THE FIRST TEST), $D_E \geq 0.13$ Sv

<table>
<thead>
<tr>
<th>Age at exposure (years)</th>
<th>Number of people</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Males</td>
<td>Females</td>
</tr>
<tr>
<td>0–19</td>
<td>5 165</td>
<td>5 198</td>
</tr>
<tr>
<td>20–59</td>
<td>5 119</td>
<td>7 509</td>
</tr>
<tr>
<td>$\geq 60$</td>
<td>725</td>
<td>1 306</td>
</tr>
<tr>
<td>Total</td>
<td>11 009</td>
<td>14 013</td>
</tr>
</tbody>
</table>

people). As a rough estimate, the internal exposure represents approximately 30% of the total effective dose $D_E$. The mean value of the absorbed dose to the thyroid for the most exposed group ($D_E > 0.25$ Sv) was estimated to be 3 Gy. Uncertainties in the estimation of the internal absorbed doses are 100–200%.

3. HEALTH RISK ASSESSMENT

In the framework of the State research programme [3], the research project “Development of the methodology (MAR) and data bank (BARD) on risk analysis” started in 1994.\footnote{This project is actually part of a more general project of MAR and BARD development carried out in the frame of the Russian State research programme (Chernobyl and Altai case studies) and the international (EU–CIS) project JSP2.} The main functions of BARD are:

1 This project is actually part of a more general project of MAR and BARD development carried out in the frame of the Russian State research programme (Chernobyl and Altai case studies) and the international (EU–CIS) project JSP2.
Assessment of the radiological and non-radiological consequences of nuclear tests and accidents, using up-to-date scientific theories about somatic and genetic effects of ionizing radiation;

Assessment of the health of the population in terms of risk indices.

The goal of these assessments is to obtain data on the consequences of the tests in order to plan social protection measures and to perform future research. BARD includes:

- Service and calculation codes for realizing the mentioned methodology;
- Health demographic data (HDD) for radiological and non-radiological risk assessment.

BARD is, to a certain extent, analogous to the computer codes ASQRAD and SPIDER-1 [8, 9]. BARD differs from them by the large intrinsic HDD database, the possibility of calculating non-radiological risks, etc.

The results of the estimation of the radiological consequences of the first nuclear weapons test for the population of the Altai region are presented. They have been obtained with BARD and from the data available at the end of 1994. Only radiogenic lethal cancer cases are considered.

3.1. Methodology

Radiological risk assessment is based on

- a general methodology of risk assessment,
- primary models of radiation risk,
- health demographic data,
- data for radiation doses to the population.

A detailed description of the risk assessment methodology is published in Ref. [10].

A number of primary models have been developed on the basis of epidemiological investigations by international and some national organizations (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), ICRP, National Radiological Protection Board (NRPB), Committee on the Biological Effects of Ionizing Radiation (BEIR), etc.) [11–14]. The latest version of the BEIR model (BEIR V) [13] has been used for the calculations in the present work. The BEIR V model makes more complete allowance for the data obtained by the late 1980s and, in particular, for the dependence of the model parameters on the age of persons, both at the time of radiation exposure and at the time of observation. In BARD, provision is also made for calculations with other models.

Health demographic data for BARD have been prepared partly within the risk assessment project of the State programme [3], using data from the State Statistics Committee of the former Soviet Union for various years. Moreover, this work is also being carried out by V.B. Kolyado and co-workers on the basis of data from local organizations and a more careful analysis of the data of public
health statistics. In the risk assessments made for this paper, use has been made of values of the age specific mortality from spontaneous malignant tumours used in the calculations of the HDD of 1970 and 1989 for the population of the Altai region, and the age structure of the population, considering the data for 1949. Part of the inhabitants of the Uglovskij and Rubtsovskij districts (about 20000 people, mainly the rural population, at the time of the first test) who received relatively high doses (on average, $D_E = 0.8$ Sv) has been considered.

It should be noted that in 1949 the population of the Altai region had a very specific age distribution because of the recent world war: children and juveniles prevailed in the population.

3.2. Results

Some results of the calculations are presented in Figs 1–4 and in Tables IV and V.

The calculated data are presented for two variants: (1) for the total population of 1949 under consideration (Figs 1 and 4 and Table IV), and (2) only for the part of this group that was 0–18 years old at the time of the test (Figs 2 and 3, Table V).

Tables IV and V give the integral (summed over time) calculated data on the excess number of deaths: the total values and the partial sums up to 1994, and the life expectancy after 1994. It should be noted that the figures given in Table V were obtained for exposed persons who were 0–18 years old at the time of the test and who were still alive in 1994.

![Graph](image)

**FIG. 1.** Annual excess mortality $M(t)$ from radiogenic cancers per 100000 persons who suffered from the test, as a function of time $t$ after the test (calculations with BARD).
FIG. 2. Annual excess mortality $M(t)$ from spontaneous cancers per 100,000 persons (age 0-18 years in 1949) who suffered from the test, as a function of time $t$ after the test (calculations with BARD).

FIG. 3. Annual excess mortality $M(t)$ from different radiogenic cancers per 100,000 persons (males, age 0-18 years in 1949) who suffered from the test, as a function of time $t$ after the test (calculations with BARD).
It follows from the results of the calculation that:

- All deaths from leukaemia occurred during about 25 years after the test.
- The largest part of excess deaths from test induced 'solid' cancers for the age group 0–18 years will occur in the future, after 1994, and will amount to about 90% for women and 40% for men relative to the mortality from spontaneous cancers;
- For the population of all ages, about half of the additional number of deaths from radiogenic cancers occurred before 1994 (see Table IV); the other excess deaths from cancer will take place after 1994.

As expected, cancers of different kinds have an essentially different time spectrum of manifestation: the corresponding maxima of the annual mortality $M(t)$ are in a very broad time interval: from 5–10 years after the test for leukaemia to 50–55 years for cancers of the digestive tract and some other cancers.

4. EPIDEMIOLOGICAL STUDY

In planning this study, the results of dose and risk assessment have been used.

In the frame of the study, a register has been compiled which includes a group of people exposed to the first test (29 August 1949) with $D_E \geq 0.13$ Sv.
TABLE IV. FATAL CANCERS IN THE POPULATION EXPOSED TO THE NUCLEAR TEST IN 1949, $D_E = 0.8 \text{ Sv}$

<table>
<thead>
<tr>
<th>Type or localization of malignant neoplasms</th>
<th>Total number of cases</th>
<th>Occurring up to 1994 (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Males</td>
<td>Females</td>
</tr>
<tr>
<td>Leukaemia</td>
<td>150</td>
<td>90</td>
</tr>
<tr>
<td></td>
<td>(42)</td>
<td>(24)</td>
</tr>
<tr>
<td>Respiratory tract</td>
<td>160</td>
<td>50</td>
</tr>
<tr>
<td></td>
<td>(700)</td>
<td>(100)</td>
</tr>
<tr>
<td>Breast</td>
<td>—</td>
<td>33</td>
</tr>
<tr>
<td></td>
<td>(100)</td>
<td></td>
</tr>
<tr>
<td>Digestive tract</td>
<td>280</td>
<td>280</td>
</tr>
<tr>
<td></td>
<td>(600)</td>
<td>(410)</td>
</tr>
<tr>
<td>Others</td>
<td>150</td>
<td>170</td>
</tr>
<tr>
<td></td>
<td>(250)</td>
<td>(340)</td>
</tr>
<tr>
<td>Thyroid$^b$</td>
<td>20</td>
<td>40</td>
</tr>
<tr>
<td>Total</td>
<td>760</td>
<td>623</td>
</tr>
<tr>
<td></td>
<td>(1592)</td>
<td>(974)</td>
</tr>
</tbody>
</table>

$a$ Number of cases per 10,000 persons of all ages; in brackets data on mortality from spontaneous cancers for the same population; calculations with BARD.

$b$ The number of morbidity cases is ten times greater.

Dose and age characteristics of this group are given in Tables II and III. The total number of people in this register is 25,022 (11,099 men and 14,013 women).

Two control groups were chosen for the study. The first group (emigrants) consists of persons born prior to 1951 and relocated to the affected territories during the five years after 29 August 1949. This group included 2,505 men and 2,842 women. The second control group consists of residents of three areas of the Altai region who were not affected by the tests. This group included 1,433 men and 1,715 women.

One of the tasks of the study is to determine the fate of people from the main group. By spring 1995, the necessary data were received for 10,289 individuals (41.1%) out of the total of 25,022. Among them are 4,675 men (42.5%) and 5,614 women (40.1%).

This study is in progress. Only some preliminary results have been received. Figure 5 gives as an example the results for the mortality from cancers of the respiratory tract. They are in reasonable agreement with the risk
TABLE V. PROGNOSIS OF FATAL CANCERS IN THE POPULATION EXPOSED TO THE NUCLEAR TEST IN 1949a

<table>
<thead>
<tr>
<th>Type or localization of malignant neoplasms</th>
<th>From the nuclear test</th>
<th>Spontaneous cases</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Males</td>
<td>Females</td>
</tr>
<tr>
<td>Leukaemia</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Respiratory tract</td>
<td>100</td>
<td>20</td>
</tr>
<tr>
<td>Breast</td>
<td>0</td>
<td>30</td>
</tr>
<tr>
<td>Digestive tract</td>
<td>400</td>
<td>470</td>
</tr>
<tr>
<td>Others</td>
<td>260</td>
<td>300</td>
</tr>
<tr>
<td>Thyroidb</td>
<td>40</td>
<td>80</td>
</tr>
<tr>
<td>Total</td>
<td>800</td>
<td>900</td>
</tr>
</tbody>
</table>

a Number of cases per 10 000 persons being alive in 1994 (age 0–20 years at the test); $D_E = 0.8$ Sv, calculated data; for comparison, data on mortality from spontaneous cancers are given, calculated with BARD.

b The number of morbidity cases is ten times greater.

assessment results. Most of the mortality cases from this radiogenic cancer have already occurred in the past.

In the 1950s to 1960s, no method of diagnosing leukaemia was available. This means that the data for the epidemiological study of this type of malignant neoplasm are lost forever.

5. CONCLUSION

A considerable part of the results obtained in the research programme has an intermediate or preliminary character. This is true for the dose and risk assessment as well as the epidemiological study. However, it is obvious that the population of the Altai region suffered seriously from the tests at the STS, especially from the first one.

In 1993, on the basis of the research results available, a concept for the social protection of the population of the Altai region was elaborated and adopted by the governmental bodies. The significance of the goals and tasks of the concept and its practical elaboration is greater than a simple realization of practical activities in the Altai region. This is the first time that dose levels have been established for decision making in terms of residual doses for the social protection of an accidentally exposed population.
FIG. 5. Annual excess mortality $M(t)$ from respiratory radiogenic cancers per 100000 persons exposed to $D_E > 0.5$ Sv (in relation to the control group); epidemiological study.

In this concept, two effective dose levels have been established for decision making regarding social protection:

$D_E^1 = 0.05$ Sv

$D_E^2 = 0.25$ Sv

The lower level, $D_E^1$, is practically a non-action level: social protection measures are introduced only if the total individual doses, $D_E$, from nuclear weapons tests are higher than $D_E^1$. The population being protected is divided into two categories, depending on the individual doses and on the results of a comparison of these doses with the dose levels.

During 1994, a more general regulation document on radiation and social protection of the population for all Russian territories radioactively contaminated from nuclear accidents and tests was elaborated by the working group of the Russian Council on Radiation Protection. Its section for social protection was further developed. However, after accumulation of practical experience in remediation measures in the Altai and Chernobyl regions and in other areas, and after evaluation of the results of the dose and risk assessment, it has become clear that the current regulation for post-accident radiation and social protection should be improved by establishing more detailed and concrete definitions of the critical organs and groups, taking into account the results of dose and risk assessments.
In the research programme for 1995 and the following years, all sections taking part are trying to obtain up-to-date information. It is necessary to improve the dose assessment with RDAM, especially for internal doses, using additional tests, for those radionuclides which had an essential impact on the population of the Altai region. It will also be tried to obtain additional results by using other methods of dose reconstruction, such as biological dosimetry, electron paramagnetic resonance (EPR) measurements of teeth and measurements of $^{137}$Cs in soil.

Complete sensitivity and uncertainty analyses should be made to obtain final results for dose and risk assessment.

The epidemiological study is principally a long term one for some important solid cancers and will have to be continued for a long period.

REFERENCES


PROBABILISTIC RISK ASSESSMENT
FOR ACCIDENTAL RELEASES FROM
NUCLEAR POWER PLANTS IN EUROPE
*Methods and results*

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and Environmental Protection (RIVM),
Bilthoven, Netherlands

**Abstract**

PROBABILISTIC RISK ASSESSMENT FOR ACCIDENTAL RELEASES FROM
NUCLEAR POWER PLANTS IN EUROPE: METHODS AND RESULTS.

The 1986 accident at the Chernobyl nuclear power plant has shown that severe accidents in nuclear power plants can lead to large scale contamination of Europe. At present, over 200 nuclear power reactors for commercial electricity production are operational in Europe. The paper focuses on an integrated assessment of excess cancer mortality due to possible accidental releases from the European nuclear power plants. For each of these plants the probability of accidental releases per year of operation is combined with the consequences in terms of excess doses received by a person over a lifetime of 70 years. Risk estimates are restricted to cancer mortality and do not include immediate or short term deaths in the direct vicinity (<5-10 km) of plants. Countermeasures to reduce radiation doses are not considered. Location specific risks for Europe are presented on maps. The excess mortality risk due to the combined operation of the European nuclear power plants is estimated to be about $10 \times 10^{-8}$ per year in western Europe. In the eastern direction the risks increase gradually to over $1000 \times 10^{-8}$ per year in regions of the former Soviet Union where reactors of the Chernobyl type are located. Risks from the nuclear power plants in eastern Europe dominate the estimated risk pattern and contribute at least 40-50% to the average risk in western Europe. Improving the reactor safety in eastern Europe could lead to considerable reductions in the estimated excess mortality risk. In western Europe the mortality risk might be reduced by a factor of two and in eastern Europe by a factor of 100 to 1000.

1. **INTRODUCTION**

The safety of nuclear power plants is an important issue in discussions of future scenarios for power generation. The major questions focus on risks connected with the disposal of radioactive waste and risks of severe accidents involving the reactor core. The accident at the Chernobyl nuclear power plant in 1986 has shown that large scale accidents in nuclear power plants can lead to contamination of an entire continent.
This study is aimed at an integrated risk assessment for possible accidental releases from nuclear power plants in Europe. A method has been developed and applied which provides a probabilistic evaluation of the chain of source–dispersion–exposure–risk. Estimation of accidental releases requires an evaluation of accident probabilities and subsequent release scenarios for all nuclear power plants. Detailed safety analyses are not available for many of the European power plants. Therefore, a generalization has been made to estimate accident probabilities and probabilistic releases (see Section 2). Atmospheric dispersion and deposition have been calculated, applying a probabilistic air dispersion model (see Section 3). Radiation exposure of the population can occur through inhalation, external exposure and ingestion of contaminated food products (Section 4). A full description of the model is given in Ref. [1].

Accident probabilities and accidental releases are expressed per year of reactor operation. The exposure is calculated for a lifetime follow-up period of 70 years, and excess risks are expressed in terms of excess cancer mortality due to excess radiation doses received. Short term deaths in the direct vicinity of power plants are not included. Baseline risk estimates are provided for an adult rural population, eating fresh products, with food consumption considered to be at the high end of the consumption range. The group involved is assumed to spend 30% of the time outdoors. Countermeasures are not considered.

The results of the evaluation are presented in risk maps, providing estimated location dependent probabilistic mortality risks due to accidents in nuclear power plants (Section 5; further details are given in Refs [1, 2]). The risk maps do not reflect the situation following a specific accident. They provide a probabilistic view of the risks involved and the major areas at risk.

2. SOURCE TERMS

All 217 European nuclear power reactors with an electric power capacity exceeding 50 MW and operational in July 1992 have been studied [3]. The nuclear power reactors are divided into nine different reactor types (based on Ref. [4]; figures in parentheses indicate operational reactors):

(a) Light water reactors, subdivided into the following types:

- (89) PWR (pressurized water reactor of Western design, with containment);
- (23) BWR (boiling water reactor of Western design, with containment);
- (10) PWR-V230 (the oldest pressurized water reactor of Russian design, without containment or redundancy in safety systems);
- (16) PWR-V213 (improved version of PWR-V230, of Russian design; limited containment and limited redundancy);
— (18) PWR-V1000 (Russian design, similar to some of the western European reactors);
— (19) LWGR (light water graphite moderated reactor of Russian design — the ‘Chernobyl type'; no containment and limited redundancy of safety systems).

(b) Gas cooled reactors:
— (24) GCR (graphite moderated, gas (CO₂) cooled reactor; the reactor core is enclosed by a steel or concrete pressure vessel);
— (14) AGR (advanced gas cooled reactor — an improved version of the GCR).

c) Fast breeder reactors (4).

Reactor design and redundancy of safety features have been considered in estimating the probabilities of severe damage to the reactor core [4]. The nuclear power reactors are classified into four accident probability classes: 10⁻³ per year (29 plants), 10⁻⁴ per year (146 plants), 10⁻⁵ per year (39 plants) and 10⁻⁶ per year (3 plants). The reference probability class is 10⁻⁴ per reactor year. If a safety analysis for a particular plant clearly indicates a substantially lower risk, the plant is placed in a lower risk class. The 29 LWGR and PWR-V230 reactors are placed in a high risk class (10⁻³ per reactor year).

The average probability of reactor core meltdown amounts to 2 x 10⁻⁴ per reactor year, which compares well with the estimate obtained when the history of core damage accidents is considered: 3.3 x 10⁻⁴ per reactor year [1].

Radioactivity release following core meltdown depends upon the quality of the containment. Four accident scenarios for possible releases are considered, on the basis of results from NUREG-1150 [4]. The release fractions for nine radionuclide groups (57 radionuclides) from the reactor core are estimated for these accident scenarios. The probabilistic releases for each reactor are estimated for each of the radionuclides considered, taking into account the meltdown probability, the various accident scenarios, the released fraction of radionuclides for those scenarios and the reactor inventory (in the middle of the fuel cycle).

3. DISPERSION MODELLING

Since an accident can occur at any time, statistically averaged dispersion and deposition need to be estimated for each of the plants considered. Probabilistically time averaged dispersion and deposition calculations have been made on the basis of a previously developed model: the OPS model [5]. The OPS model describes average air concentration and deposition in the Netherlands for continuous releases.
We have considered the applicability of the OPS model for probabilistically averaged accidental releases and an extension of the dispersion calculations to a European scale [1]. Assuming that accident probabilities and accidental releases are independent of the weather conditions, probabilistic releases could be treated as being equivalent to continuous releases. This finding is in line with previous conclusions from BIOMOVS evaluations of the applicability of steady state modelling of contamination of milk as a result of accidents [6]. The approach is valid only in the case of linear release–effect relationships.

Calculations of acid deposition on a European scale and comparative calculations using deposition data from the Chernobyl accident have confirmed that the extrapolation of the dispersion calculations to a European scale is not a source of large errors (less than a factor of three). Large deviations, however, might occur, for example, in mountainous regions with heavy rainfall.

4. EXPOSURE MODELLING

The purpose of the exposure assessment is to estimate the accumulated lifetime dose commitment due to the passage of a radioactive cloud and the resulting deposition of radioactive material on vegetation and soil. The dose received is calculated for a person remaining at the same location for a period of 70 years. The exposure pathways considered are inhalation, ingestion and external exposure.

4.1. Inhalation

Inhalation of radionuclides was modelled according to the method described by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [7], choosing the following major parameters: indoor air concentration equals outdoor air concentration, a breathing rate of 23 m$^3$/d and dose conversion coefficients according to Ref. [8].

4.2. Ingestion

Contamination of vegetation and crops occurs through at least two pathways: direct interception of deposited radionuclides during passage of the cloud and uptake of deposited radionuclides from the soil. After passage of the cloud the soil remains contaminated, although, owing to removal from the upper layers in combination with nuclear decay, the concentration in the upper soil layer will decrease. The intake of radionuclides from food can be either direct, through consumption of contaminated crops, or indirect, through consumption of milk and meat from animals who have eaten contaminated grass and soil. Five major food categories have been distinguished in ingestion modelling: vegetables, cereals, roots/tubers, and milk and meat.
from cows. The modelling expressions are in line with those given in Ref. [9]. Transfer factors for soil–plant and cow–milk/meat transfer have been obtained from Refs [10, 11].

4.3. External exposure

External exposure directly from the radioactive cloud and from deposited radionuclides has been estimated according to a method described by UNSCEAR [7] for the evaluation of the Chernobyl accident. Dose conversion factors for all 57 radionuclides have been obtained from Ref. [12]. Shielding from radiation due to migration of radionuclides into the ground has been considered, according to UNSCEAR, for three different time intervals: in the first month no reduction, from 1 month to 1 year 50% reduction and in the period beyond 1 year 63% reduction. Shielding inside houses is assumed to reduce the doses due to external exposure from the cloud by 30% and the doses due to external exposure from deposited radionuclides by 70%.

4.4. Results of exposure modelling

Ingestion is the major pathway contributing to the dose (±50% of the total dose). External exposure contributes around 33% (primarily due to $^{137}\text{Cs}$), inhalation contributes around 10% and external exposure from the cloud contributes 3% or less. Thus, deposition related contributions amount to around 85% of the total dose. For the adult population, 70% of the 70 year follow-up dose will be received in the first year.

The two major radionuclides contributing to the dose for the various source terms are $^{131}\text{I}$ and $^{137}\text{Cs}$, which contribute 60–75% of the total dose from all 57 radionuclides. In addition, $^{134}\text{Cs}$ contributes approximately 15%; all other radionuclides contribute less than 5% each, and no more than maximally 25% in total.

5. RISK EVALUATION

For 8000 receptor locations in Europe, the excess cancer mortality risks due to possible accidental releases from the 217 operational nuclear power reactors have been calculated, using the methods described above. For each receptor location the probabilistic time integrated air concentration and the total deposition have been calculated for each of the reactors. Next, the 70 year follow-up doses have been calculated and the results for all reactors summed to obtain effective doses. We have applied a dose–mortality risk conversion coefficient of 2.5% per Sievert to obtain mortality risks.
FIG. 1. Estimated mortality risk due to possible accidental releases from nuclear power plants in Europe: the present situation.
The risk estimates are plotted on a map of Europe. Two risk maps have been obtained: one representing the present situation and a second one representing a situation for which it is assumed that eastern European reactor types have a safety level comparable to that of western European reactors.

The estimated excess mortality risk for Europe shows a large variation (see Fig. 1). The risk is less than $10^{-8}$ per year in Iceland and in south-western parts of Spain and Portugal. It increases from west to east: $2 \times 10^{-8}$ per year in Ireland; $3 \times 10^{-8}$ to $10 \times 10^{-8}$ per year in the United Kingdom and in large parts of France, Italy and Norway; and around $10 \times 10^{-8}$ to $30 \times 10^{-8}$ per year in Belgium, Germany and the Netherlands, and in large parts of central Europe. A risk of over $100 \times 10^{-8}$ per year is found for large areas of the former Soviet Union, including the Baltic States, Belarus, Russia and Ukraine. In these countries, the risk of $1000 \times 10^{-8}$ per year is exceeded in small regions around nuclear reactors. Further towards the eastern border of Europe, the risk declines to around $10 \times 10^{-8}$ per year.

Risks from nuclear power reactors in the eastern European countries dominate the estimated risk pattern and contribute at least 40–50% to the average risk in the western European countries. Improving reactor safety in the eastern European countries would lead to considerable reductions in the estimated risk (Fig. 2). These results show that the largest risk reductions can be achieved in eastern Europe (more than a factor of 100), whereas reductions of approximately 50% are possible for western Europe, where the density of nuclear plants is highest.

An indication of the overall uncertainty involved is obtained by assuming that various errors in the chain are independent of each other and are lognormally distributed [1]. The largest uncertainties are due to the estimation of accident probabilities, accident scenarios and radionuclide releases. The overall risk is obtained by a multiplication of the various components in the chain. The overall uncertainty factor estimated (at 95% significance) is 15 in western Europe and 20–25 in eastern Europe (higher as well as lower). These estimates must be seen as first indications of the overall uncertainty.

We have used a dose-mortality risk conversion coefficient of 2.5% per Sievert. Implementing the new ICRP risk coefficient of 5% per Sievert [13] would increase all risk estimates by a factor of two. Implementing the new ICRP dose conversion coefficients [13] primarily influences the $^{131}\text{I}$ contribution, increasing inhalation and ingestion doses from $^{131}\text{I}$ by 70%. The overall doses and risks are increased by 20–30%. Thus, completely accounting for the new ICRP conversion coefficients increases the overall risk by a factor of 2.5.

Another source of variation in the risk assessment may be due to the definition of the risk group under consideration. Small children at the time of an accident are estimated to have threefold to fourfold higher risks than the risk group considered here. A mixed group of adults from rural and urban areas with average food consumption, for which exposure parameters as provided by UNSCEAR [7] are applied, is expected to receive 50% lower doses than estimated in this study.
RISK MAP
"Safe technology"
Risks due to possible accidents with nuclear power plants
(July 1992)
Lethal risk ranges
(per year):

- $< 10^{-8}$
- $10^{-8} - 10^{-7}$
- $10^{-7} - 10^{-6}$
- $10^{-6} - 10^{-5}$
- $> 10^{-5}$

FIG. 2. Estimated mortality risk due to possible accidental releases from nuclear power plants in Europe: assuming that the safety of eastern European reactors becomes comparable to that of western European reactors.
6. DISCUSSION

The aim of environmental policies is the reduction of man-made environmental risks from both regular and accidental releases. This study provides a probabilistic evaluation of individual mortality risks due to possible accidents in nuclear power plants in Europe. The risks evaluated in this study are restricted to radiation induced cancer mortality risks. Thus, victims of acute radiation, which can occur in the close vicinity of a reactor (<5-10 km), are not included. Another aspect not covered in the present evaluation is the fact that the occurrence of a large scale accident could lead to disruption of society.

Although risk evaluation does not cover all aspects of risks involved in accident situations, it serves the purpose of putting risks in perspective and making them comparable with risks from regular emissions. The overall average uncertainty of the evaluation is considerable. The main contributor to the uncertainty is the lack of knowledge of probabilities of accidents and accidental releases, especially for the eastern European reactors. Further systematic safety analyses of reactors are required to reduce uncertainties.

Risks from eastern European reactors currently dominate the calculated risks in Europe; the overall risks could be substantially reduced if the safety measures and procedures applied in western European reactors would also be implemented in eastern European reactors. In that case, reduction of the average risk in western Europe would be nearly twofold, and in eastern Europe the risk reduction might amount to more than a factor of 100. Thus, it is concluded that considerable risk reduction can be achieved by safety design of high quality.

REFERENCES


CONSEQUENCES OF THE RADIOACTIVE CONTAMINATION OF THE SVERDLOVSK OBLAST CAUSED BY THE ACCIDENT IN 1957 AT THE MAYAK INDUSTRIAL COMPLEX

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Abstract

CONSEQUENCES OF THE RADIOACTIVE CONTAMINATION OF THE SVERDLOVSK OBLAST CAUSED BY THE ACCIDENT IN 1957 AT THE MAYAK INDUSTRIAL COMPLEX.

Routine activities at the Mayak Industrial Complex for over 40 years as well as the radiation accidents that occurred have led to an unprecedented radioactive contamination level in a considerable part of the Chelyabinsk, Kurgan and Sverdlovsk regions of Russia. Dumping of radiochemical waste with an activity of 2.8 million curies into the Techa river in 1949–1952 resulted in radioactive contamination of the Techa–Iset–Tobol river system. In September 1957, a storage tank for liquid high activity waste at the Mayak site exploded. Radionuclides with an activity of about twenty million curies were released to the environment, of which around two million curies were discharged outside the territory of the Mayak Industrial Complex, thereby giving rise to the East-Ural radioactive trace. The considerable radioactive contamination of the environment and the radiation exposure of the population, especially in the first decades of operation of the enterprise, are due to routine process discharges of radioactive matter to the atmosphere from the Mayak Industrial Complex. The steps taken before 1991 with a view to alleviating the consequences of the radioactive contamination of this area were not adequate to the extent of the ecological and economic damage and the harm done to the health of the population. In 1991, the Government of the former Soviet Union (FSU) decided to develop a State programme for the period up to the year 1995, focusing on medical and psychological rehabilitation of the radiation affected population and on measures to be taken to render assistance to it. This programme was worked out by a group that was formed in the Ural region, including representatives of the Ministry for Atomic Energy of the FSU, as well as of the Administrations of the Chelyabinsk, Kurgan and Sverdlovsk regions, and the Ural Division of the Russian Academy of Sciences, which was represented by the Institute of Industrial Ecology (IIE). In 1993, the State programme was officially adopted, and the IIE was appointed as scientific co-ordinator and general contractor for assistance of the section dealing with the problems of the territory. Data on the radioecological situation in the Sverdlovsk region have been obtained on the basis of a complex analysis of archival material on the state and dynamics of the radioecological conditions of the territory of the Sverdlovsk Oblast in the period 1957–1995, as well as from a study made by other institutes in 1992–1994. These results are presented in the paper and discussed in detail.
Routine activities at the Mayak Industrial Complex for over 40 years as well as the radiation accidents that occurred have led to an unprecedented radioactive contamination level in a considerable part of the Chelyabinsk, Kurgan and Sverdlovsk regions.

Dumping of radiochemical waste with an activity of 2.8 million Ci\(^1\) into the river Techa in 1949–1952 resulted in radioactive contamination of the Techa–Iset–Tobol river system. Approximately 5500 inhabitants of 22 riverain localities were resettled. So far, the regime of normal vital activities along the river has not been restored, with 8000 ha of farming land abandoned to date.

In September 1957, a storage tank for liquid high activity waste at the Mayak site exploded. Radionuclides with an activity of about twenty million curies were released to the environment, of which around two million curies were discharged outside the territory of the Mayak Industrial Complex, thereby giving rise to the East-Ural radioactive trace (EURT). The northern part of the Chelyabinsk region and the southern part of the Sverdlovsk region suffered radioactive contamination. Twenty-three inhabited localities within the radioactive contamination zone were abandoned and 10 400 dwellers were resettled.

In spring 1967, radioactive substances of about 600 Ci were scattered by gusty winds from the banks of the Karachai lake to adjacent territories.

The considerable radioactive contamination of the environment and the radiation exposure of the population, especially in the first decades of the activities of the enterprise, are due to routine process discharges of radioactive matter to the atmosphere from the Mayak Industrial Complex. The effect of these emissions is traceable over a distance of 20–30 km westwards and 50–70 km eastwards.

The problems associated with the activities of the Mayak Industrial Complex are not restricted to those arising from the previous radioactive contamination of the regional territory. Over the years of operation of the enterprise, radioactive wastes of all categories (about one billion curies) have piled up, including waste deposited in the open air. Such accumulation of very large amounts of radioactive waste in a densely populated region is fraught with the threat of new large scale disasters.

The steps taken before 1991 with a view to alleviating the consequences of the radioactive contamination of the area where the Mayak Industrial Complex is located were not adequate to the extent of the ecological and economic damage and the harm done to the health of the population, nor did those measures resolve the challenges associated with the radioactive contamination of the region and the accumulation of very large amounts of radioactive waste substances. In 1991, the Government therefore decided that a Russian Federation State programme should be developed for the period up to the year 1995, focusing on medical and psychological rehabilitation of the radiation affected population and on measures to be taken to render assistance.

\(^1\) 1 Ci = 3.7 \times 10^{10} \text{ Bq.}
to it. The programme was worked out through a concerted effort of an initiative group that was formed in the Ural region. This group included representatives of the Ministry for Atomic Energy of the former Soviet Union (FSU), the Administrations of the Chelyabinsk, Kurgan and Sverdlovsk regions, and the Ural Division of the Russian Academy of Sciences (UD RAS), which was represented by the Institute of Industrial Ecology (IIE).

The prime objects of the State programme were defined as follows:

— Reduction of the risk of new large scale radiation accidents due to the unprecedented accumulation of radioactive waste on the production site and in the sanitary protection zone of the Mayak Industrial Complex — waste substances capable of leading to additional radiation exposure and re-exposure of the population;
— Medical and psychological rehabilitation of the population exposed to elevated radiation as a result of the activities of the Mayak Industrial Complex;
— Social and economic rehabilitation of the territories of the Ural region that were subjected to radioactive contamination;
— Ecological and radiation sanitation of the territories contaminated by radioactive substances.

Structurally, the State programme consists of five segments, viz. avenues of scientific and practical work:

1. Lowering the degree of risk of radiation accidents and disasters, and taking measures to eliminate potential sources of radioactive contamination (problems relating to the Mayak production site).
2. Monitoring the ecological and radiation situation and sanitizing the territories;
3. Taking health care measures for the radiation affected population;
4. Providing social and economic rehabilitation of the contaminated territories;
5. Taking steps to develop agroindustrial production and forestry.

In 1993, the State programme was officially adopted by the Government and approved by the Supreme Soviet. In accordance with the system of managing and organizing science research work as provided in the State programme, the IIE was appointed as scientific co-ordinator and general contractor for scientific assistance of the State programme section dealing with problems of the territory (four of the five segments).

The IIE concluded contracts for conducting scientific research on specific topics with 42 leading regional and Federal scientific organizations.

Participants in this research were the IIE, the Institute of Plant and Animal Ecology of the UD RAS, the Forestry Institute of the UD RAS, the Geophysics Institute of the UD RAS, the Ural State Technical University, the Economics Institute of the UD RAS, the Ural State Medical Institute (all located in Ekaterinburg), the
Biology Institute of the Komi Science Centre of the UD RAS (Syktyvkar), the Institute of Global Climate and Ecology of the Russian Hydrometeorology Department (Moscow), the Institute of Experimental Meteorology of the Scientific Production Association “Typhoon” (Obninsk), the Institute for Radiation Sanitation (St. Petersburg), the V.G. Khlopin Radium Institute (St. Petersburg), the Radiological Research Station of the Mayak Industrial Complex, the Biophysics Institute (Chelyabinsk-65), the Chelyabinsk Branch of the Russian Institute of Science Research Water Management, the Chelyabinsk Rosgidromet Regional Centre for Hydrometeorology and Monitoring of the Natural Environment, the Ural Science and Practice Centre for Radiation Medicine of the Ministry of Health of the Russian Federation, the Chelyabinsk State Agricultural Engineering University, the Chelyabinsk Centre for Chemization and Agricultural Radiology, and a number of other organizations.

Under the scientific guidance of the IIE, these organizations jointly developed and carried out co-ordinated research work. The results of this work regarding the radioecological situation in the Sverdlovsk region are discussed below. These results have been obtained on the basis of a complex analysis of archival material of various organizations on the state and dynamics of the radioecological conditions of the territory of the Sverdlovsk Oblast in the period from 1957 to 1995, as well as on the basis of results of a study of the environmental conditions of the territory of the Sverdlovsk Oblast from other institutes in 1992–1994. The IIE was not only the co-ordinator but also the executor of this work.

(1) The IIE prepared an official list of previous work on the determination of the radioactive situation on the territory of the EURT, conducted in the initial period, from the first days after the accident in September 1957 to 1961. It was found that during this period, at the request of the Mayak Industrial Complex, the Institute of Applied Geophysics of the Academy of Sciences of the FSU and the Institute of Radiation Hygiene of the Ministry of Health conducted at least ten expeditions in order to investigate the radioactive situation on the territory of the Sverdlovsk Oblast. The initial data and the results of the investigations carried out in 1957–1961 are probably stored in the archives of these organizations, but we have not been able to examine them.

(2) An analysis of the cartographic data from accessible archives on initial and contemporary levels of the radioactive contamination of the territory of the Sverdlovsk Oblast caused by the accident in the Mayak facility in September 1957 (14 maps created by six organizations) shows that there are no reliable maps on the $^{90}$Sr contamination of the territory that could be used as a basis for the realization of practical rehabilitation measures. The overwhelming majority of the available maps is very schematic and does not reflect the real structure of the EURT.

The data on the $^{90}$Sr contamination levels of the southern part of the Sverdlovsk Oblast obtained by different organizations differ from each other by a
factor of two, and the data on the contamination of Kamensk-Uralsky differ by a factor of twenty — from 0.2 to 4.0 Ci/km$^2$.

Some of the maps show distortions: On the official EURT map the city of Kamensk-Uralsky has been erroneously placed several kilometres east of the axis of the radioactive trace.

The situation is complicated by the absence of primary data that would allow the reliability of the maps to be evaluated. Thus, the present levels of the radioactive contamination of the Sverdlovsk Oblast cannot be restored by extrapolation of the initial contamination data.

The only reliable source of information on the radioactive contamination of the territory of the Kamensk, Bogdanovich and Kamyslov districts of the Sverdlovsk Oblast is the results of field investigations performed in 1992–1994.

(3) In the period 1992–1994, more than 3500 soil samples from 370 test sites in the south-eastern part of the Sverdlovsk Oblast were investigated. The investigated area was about 1600 km$^2$. Soil layers of 5–10 cm were cut horizontally, to a depth of 40–75 cm. Radiochemical analyses for $^{90}$Sr and $^{137}$Cs were carried out.

It was discovered that, to this day, there are significant levels of $^{90}$Sr contamination in the Sverdlovsk Oblast, especially in the Kamensk district. Along the axis of the radioactive trace north-east of Lake Tygish there is a large territory where the average $^{90}$Sr contamination is 1–3 Ci/km$^2$, with some spots having activities of more than 5 Ci/km$^2$.

These territories include the settlements Rybnikovskoye, Bogatenkovo, Shcherbakovo, Kluchi, Kluchiki, Kodyinka, Malaya Kodyinka, Cheremkovo, Belovodje and Svoboda, the population of which probably suffers from enhanced intake of $^{90}$Sr with food products, mainly milk and meat from farmers' private production.

From the cross-sections of the radioactive trace in the southern part of the Kamensk district it can be seen that for an activity of up to 1.0 Ci $^{90}$Sr/km$^2$ the width of the trace is 12–15 km, and for an activity of up to 0.1 Ci $^{90}$Sr/km$^2$ the width is about 30 km. The radioactive trace is at least two or three times wider than stated in the official documents of 1958 and 1959.

The present $^{90}$Sr contamination of Kamensk-Uralsky varies from 0.06 to 6.9 Ci/km$^2$. The average contamination levels vary from 0.3 Ci/km$^2$ in the eastern part of the town to 0.7 Ci/km$^2$ in the western part. The average contamination of the Bogdanovich and Kamyslov districts is 0.2–0.4 Ci/km, with a maximum value of up to 1.2 Ci/km$^2$.

The $^{137}$Cs contamination levels in the investigated part of the Sverdlovsk Oblast are in the range of 0.02–3.1 Ci/km$^2$, with an average value of 0.3 Ci/km$^2$. This value is three times higher than the level of global fallout.

The plutonium content in samples from three soil sections on the EURT axis in the southern part of the Kamensk district corresponds to the level of surface contamination (0.005–0.03 Ci/km$^2$).
(4) The vertical depth profile of the $^{90}$Sr distribution pattern is virtually unchanged for all soils typical of the region under review. In the overwhelming majority of cases, 70–95% of the total amount of radionuclides resides in the upper 15 cm layer. A forecast of the vertical distribution of $^{90}$Sr for the next 10 or 15 years shows that the distribution pattern will not change fundamentally; 55–65% of the entire reserve of $^{90}$Sr will be in the 20 cm deep soil layer containing the roots and will remain accessible to plants.

(5) The concentration of $^{90}$Sr in the main wood species ranges at present from 1 to 100 Bq per kg air-dry wood for pine trees, from 2 to 230 Bq per kg air-dry wood for birch trees and from 3 to 470 Bq per kg air-dry wood for aspen trees. The contamination of wood by $^{137}$Cs is up to 7 Bq per kg air-dry wood for pine trees, up to 35 Bq per kg air-dry wood for birch trees and up to 50 Bq per kg air-dry wood for aspen trees.

The levels of contamination of mushrooms in the territory of the Kamensk and Bogdanovich districts (6–53 Bq per kg air-dry mushrooms for $^{90}$Sr and 29–143 Bq per kg air-dry mushrooms for $^{137}$Cs) are much higher than the background levels but still lower than the derived intervention levels; therefore, these products may be used safely.

(6) From 1960 onwards, the level of $^{90}$Sr concentration in the water of Lakes Tygish, Sungul' and Chervyanoe was at least two times lower than the permissible level. At present, the $^{90}$Sr concentration in Lakes Tygish, Sungul' and Chervyanoe is about $(7-20) \times 10^{-12}$ Ci/L, i.e. one to two times lower than the permissible level. Over the whole period after the accident the concentration of $^{90}$Sr in the water of all three lakes decreased by a factor of two in 15 years, which is somewhat shorter than the half-life of $^{90}$Sr.

Because of the levels of $^{90}$Sr contamination of sapropel from Lake Tygish (mean concentration up to $(5-20) \times 10^{-9}$ Ci per kg of dry substance; maximum up to $56 \times 10^{-9}$ Ci per kg of dry substance) the use of sapropel as fertilizer is limited.

Even in 1992, 35 years after the accident, the radioactive contamination of fish in these lakes was very high. Over the past ten years the $^{90}$Sr concentration in fish from Lake Tygish was $(5.0-12.3) \times 10^{-9}$ Ci/kg, from Lake Sungul' it was $(1.7-3.7) \times 10^{-9}$ Ci/kg and from Lake Chervyanoe it was $(1.2-3.8) \times 10^{-9}$ Ci/kg.

Up to 1993, the maximum permissible concentration of $^{90}$Sr in fish was $1.0 \times 10^{-9}$ Ci/kg. This permissible concentration value was changed in 1993 and is now $2.7 \times 10^{-9}$ Ci/kg. But, even though the permissible concentration level was lowered, the contamination of fish by $^{90}$Sr in 1993 was still higher than the permissible level (crustaceans from Lake Tygish: $5.0 \times 10^{-9}$ Ci/kg, and crustaceans from Lake Chervyanoe: $3.8 \times 10^{-9}$ Ci/kg).

In view of the fact that uncontrolled fish consumption and the use of sapropel from Lake Tygish are still hazardous, it is advisable to keep up the existing order,
according to which the use of Lake Tygish for rural economy (fish production, use of water for vegetables, use of silt as fertilizer) must be agreed to by State sanitary officials.

(7) The \(^{90}\text{Sr}\) concentration in the water of the Kamenka River in the summer of 1993 was \((1-2) \times 10^{-12}\) Ci/L; in the water of its small tributaries it was \((1-15) \times 10^{-12}\) Ci/L. This is much higher (100-1000 fold) than the background levels, but it is still lower than the limit (by a factor of 10-100), so that this water can be used without limitations.

The concentration of \(^{137}\text{Cs}\) in the water of the Kamenka River and its small tributaries in 1993 was \((0.3-1.7) \times 10^{-12}\) Ci/L, which is 10 000 times lower than the derived intervention level.

The contamination of the bottom sediments of the Kamenka River by \(^{90}\text{Sr}\) in 1993 was \((0.6-2.0) \times 10^{-9}\) Ci per kg dry weight and the contamination of aquatic plants was \((0.1-1.9) \times 10^{-9}\) Ci per kg dry weight.

The average annual \(^{90}\text{Sr}\) concentration in the water of the Sinara River in 1992 was 0.7-0.8 Bq/L and in 1993 it was 1.5 Bq/L. The average annual \(^{137}\text{Cs}\) concentration in the water of the Sinara River in 1993 was 0.2 Bq/L.

(8) At present, the \(^{90}\text{Sr}\) contamination of peat-bogs of the Kamensk district (11 peat bogs) is 40-1600 Bq per kg air-dry peat and the \(^{137}\text{Cs}\) contamination of peat-bogs is 100-1700 Bq per kg air-dry peat.

(9) Because of the long time period that has passed since the deposition of the radioactive fallout, the radiation situation is quite stable. The role of geochemical migration, wind transfer and washout by surface run-off in the spatial redistribution of radionuclides and in the change of the boundaries of radiation contaminated territories is currently negligibly small. A major factor conducive to lowering the radioactive contamination of the territory is the physical decay of \(^{90}\text{Sr}\).

(10) The main sources of human irradiation in the contaminated territory were internal irradiation from incorporated \(^{90}\text{Sr}\), irradiation of the gastrointestinal tract from nuclear fission products (mainly \(^{95}\text{Zr} + ^{95}\text{Nb}\) and \(^{144}\text{Ce} + ^{144}\text{Pr}\)) and external irradiation.

Estimates of annual intakes of \(^{90}\text{Sr}\) into the human body were obtained from an analysis of the material in the archives of the State sanitary supervision service of the Sverdlovsk Oblast, which contain information on the contamination of foodstuffs (meat, milk, grain, potatoes). The accumulated doses (1993) due to internal irradiation from incorporated \(^{90}\text{Sr}\) for the most contaminated sites, Rybnikovo and Shcherbakovo, were estimated as follows: bones — from 11.2 to 19.6 cSv, depending on age; red bone marrow — from 5.1 to 8.8 cSv; effective equivalent dose — from 0.4 to 0.7 cSv.
The average irradiation doses to the lower part of the large intestine were from 1.5 to 11.0 cSv, depending on age; and the effective equivalent dose due to irradiation of the gastrointestinal tract was from 0.4 to 0.7 cSv. The accumulated doses (1993) due to external irradiation for the most contaminated settlements of the Sverdlovsk Oblast were from 0.3 to 1.7 cSv.

It was estimated that the critical group includes children born between 1955 and 1957, and, to a lesser degree, those born between 1940 and 1945. For this critical group the integral effective equivalent dose due to all sources of irradiation is about 4.8–5.2 cSv.

According to official data of the State sanitary supervision service, the doses due to internal irradiation from incorporated $^{90}$Sr in the city Kamensk-Uralsky do not differ greatly from the average doses in the FSU. It is, however, necessary to take into account that these official data include only the centralized supply of food from non-contaminated areas. When estimating the doses for Kamensk-Uralsky we did not consider the $^{90}$Sr intakes for the urban population consuming milk and vegetables from private farms and kitchen-gardens on the western boundaries of Kamensk-Uralsky. The accumulated doses (1993) due to external irradiation for Kamensk-Uralsky are 0.1–0.7 cSv.

An epidemic analysis of the indexes specifying the health of the population dwelling in the territory of the EURT testifies that the primary morbidity of the adult population, as well as the rates at which chronic and protracted diseases increase in number for both adults and children exceed significantly the mean regional sickness rates and the morbidity of the population in other rural areas.

An examination of the inhabitants of the city Kamensk-Uralsky revealed substantial changes for the worse in the immuno-haematological status of those who were children in 1957–1962 in the areas with the highest level of radioactive contamination. Also, noticeable but less pronounced changes in health were found in children living in the contaminated areas and who are second and third generation descendants of persons who lived in the territory of the EURT in the early post-accident period. Thus, the data obtained testify to the negative effect of the EURT on the health of the population in the affected territories of the Sverdlovsk Oblast.
MEDICAL CONSEQUENCES OF RADIATION ACCIDENTS

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Abstract

MEDICAL CONSEQUENCES OF RADIATION ACCIDENTS.

Since 1945, more than $1.8 \times 10^{21}$ Bq of artificial radionuclides have been released into the atmosphere. Approximately $2.04 \times 10^{18}$ Bq, i.e. $\approx 0.11\%$, are the result of accidents at nuclear industrial facilities. This percentage is causing increased interest among researchers. This is due to the fact that in the wake of accidental releases radionuclides become distributed unevenly across the Earth's surface, and the associated exposures, fluctuating from background level to several grays, can induce both stochastic and deterministic effects in the irradiated population. A comparative analysis of the medical consequences of the twentieth century's most serious nuclear events, namely the authorized dumping of high level radioactive waste into the river Techa in 1950, the explosion of a storage tank containing long lived radioactive waste in the Southern Urals in 1957, the fire at Sellafield in 1957 and the accident at the Chernobyl nuclear power plant in 1986, has shown that when timely countermeasures are taken, the worst immediate and delayed medical consequences of an accident can be avoided. The consequences that have since been ascertained are a brief rise in the mortality rate during the first five years, with a dose in excess of 500 mSv; an increase in the incidence of leukaemia, with an absolute risk of up to $1.1 \times 10^{-4}$ man-years/Gy; and increased mortality among children with external radiation doses of up to 1000 mSv, and internal doses of 99-190 mSv on the bone surfaces of neonates or 170-600 mSv on the bone surfaces of the mother. There is reliable evidence that, with external gamma radiation doses in excess of 520 mSv, the mortality rate for all malignant tumours increases by 45-58% compared with the control level. There is also a significant increase in thyroid cancer frequency four to ten years after the incorporation of iodine isotopes by children aged up to 7 years, including an accumulation period in the womb.

In the second half of the twentieth century, a mixture of radionuclides amounting to $1.8 \times 10^{21}$ Bq has entered the atmosphere, 99.89% of it being due to atmospheric nuclear weapons testing and only 0.11% being the result of nuclear accidents (Table I). Paradoxically, precisely that 0.11% is attracting considerable attention from researchers worldwide. This is because the radionuclide dispersion area after accidents is comparatively small and fallout densities can be very high. As a result, there are small areas where the radiation doses are capable of producing deterministic effects.
TABLE I. MAIN Instances OF Radionuclides entering the Atmosphere BETWEEN 1946 AND 1986

<table>
<thead>
<tr>
<th>Source</th>
<th>Total activity ($10^{16}$ Bq)</th>
<th>$^{137}$Cs</th>
<th>$^{90}$Sr</th>
<th>$^{131}$I</th>
<th>Dispersion ($\text{km}^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atmospheric nuclear weapons testing</td>
<td>181 060</td>
<td>96</td>
<td>66</td>
<td>10 000</td>
<td>$510 \times 10^6$</td>
</tr>
<tr>
<td>Accident at Unit 4 of the Chernobyl nuclear power plant (1986)</td>
<td>185</td>
<td>7</td>
<td>0.44</td>
<td>63</td>
<td>$250 \times 10^6$</td>
</tr>
<tr>
<td>Accident near Kyshtym (1957)</td>
<td>7.4</td>
<td>0.02</td>
<td>0.2</td>
<td>0</td>
<td>$23 \times 10^3$</td>
</tr>
<tr>
<td>Wind borne radionuclides from the Lake Karachaj flood plain (1967)</td>
<td>0.003</td>
<td>0.00004</td>
<td>0.00004</td>
<td>0</td>
<td>$2 \times 10^2$</td>
</tr>
<tr>
<td>Authorized dumping of radioactive waste into the river Techa (1950)</td>
<td>10.2</td>
<td>1.24</td>
<td>1.18</td>
<td>0</td>
<td>$2 \times 10^2$</td>
</tr>
<tr>
<td>Fire at the Windscale (Sellafield) facility (1957)</td>
<td>1.1</td>
<td>0.0046</td>
<td>0.00002</td>
<td>0.12</td>
<td>$3 \times 10^2$</td>
</tr>
</tbody>
</table>

Thus, in 1950, authorized dumping of nuclear waste into flowing water, the river Techa, took place. The total volume discharged was 76 million m$^3$, with an activity of $10.1 \times 10^{16}$ Bq, the average daily release being up to $1.6 \times 10^{14}$ Bq. The discharges were made up of $\approx 12.2\%$ $^{137}$Cs, $8.8\%$ $^{89}$Sr, $11.6\%$ $^{90}$Sr, $26.8\%$ of a mixture of rare earth elements, $12.6\%$ $^{95}$Zr and $^{95}$Nb, and $25.9\%$ $^{106}$Ru-$^{106}$Rh [1–4].

There were high radionuclide concentrations in all parts of the river system, creating an intense gamma field. A fairly serious radiation situation developed over a distance of 240 km along the river. The beta emitter activity of the water initially reached 851 Bq, and the exposure rate was 5500 mR/h,$^1$ while 30 years later the exposure rate was still 9 mR/h, i.e. 1000 times higher than the natural background level [1, 4, 5].

The activity levels decreased in proportion to the distance from the point where the radionuclides were dumped, as did exposure rates. Among the population on the upper reaches of the river the effective dose equivalent amounted to 1.7 Sv over

\[ 1 \text{ R} = 258 \mu \text{C/kg} = 10 \text{ mSv}. \]
one year, while on the middle stretch it was 0.28 Sv over 38 years and on the lower reaches it was 0.07 Sv over 38 years. The radionuclide concentration, which in the first year was 23–851 kBq/m$^3$, fell after 38 years to 1.0 kBq/m$^3$.

The overall effective dose equivalent over 30 years among the inhabitants of the population centres is estimated to be 36 mSv for those living on the lower reaches of the river, the value for red bone marrow being 10 mSv; for those living on the middle and upper reaches of the river the values are 1400 and 1220 mSv, respectively. The health status of people living along the banks of the river Techa is presented in Table II. It can be seen that among the inhabitants in group II no cases of chronic radiation sickness were recorded and that the occurrence of radiation reactions was between two and five times less frequent than in group I.

The micro-organic injuries to the nervous system at 1220 mSv are a notable feature. The fertility rate of women living on the middle and lower stretches of the river hardly changed at all, while that of women living on the upper stretch even increased (doses to gonads up to 930 mSv, effective dose equivalent 1400 mSv). This may be a consequence of the attitude to child bearing among the Turkic population there as compared to the Russian population living on the middle stretch of the river.

**TABLE II. CHRONIC RADIATION SICKNESS AND RADIATION REACTIONS AMONG INHABITANTS OF AREAS ALONG THE RIVER TECHA**

<table>
<thead>
<tr>
<th>Type of radiation injury</th>
<th>Frequency of appearance of injury symptoms (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Group I (1000–1700 mSv)</td>
</tr>
<tr>
<td>Chronic radiation sickness</td>
<td>935 cases</td>
</tr>
<tr>
<td>Suppression of medullary haematopoiesis</td>
<td>7</td>
</tr>
<tr>
<td>Hyperplasia of haematopoietic tissue within 15–20 years</td>
<td>Isolated cases</td>
</tr>
<tr>
<td>Leukopenia</td>
<td>24</td>
</tr>
<tr>
<td>Neutropenia</td>
<td>48</td>
</tr>
<tr>
<td>Thrombopenia</td>
<td>45</td>
</tr>
<tr>
<td>Bone pain syndrome</td>
<td>11</td>
</tr>
<tr>
<td>Functional neurological disorders</td>
<td>25</td>
</tr>
<tr>
<td>Micro-organic neurological symptoms</td>
<td>6 (1220 mSv)</td>
</tr>
<tr>
<td>Immunosuppression</td>
<td>12</td>
</tr>
</tbody>
</table>
In the first six years after the population was subjected to irradiation the stillbirth rate rose from $5.8 \pm 1.3$ to $13.9 \pm 7.0$ per 1000 births, and the frequency of stillbirths rose by a factor of 1.5 compared to the control group. In this period the doses were $\approx 1$ Sv. For the period 7–13 years after the event the stillbirth rate was $(5.6 \pm 1.2) \times 10^{-3}$, as against $(6.2 \pm 0.9) \times 10^{-3}$ in the control group.

The information in Table III shows that the surge in early child deaths in the first two years gradually abated, in parallel with the decrease in the dose to children's bone surfaces, despite a continuing dose build-up among the adults. This is evidence of the direct effect of radiation on children.

High child mortality and increased mortality in persons of mature age have affected the average life expectancy in both groups. Thus, in the control period the average life expectancy in groups I and II was $49.9 \pm 2.8$ and $49 \pm 1.3$ years, respectively. In the first six years the average life expectancy decreased to $48.8 \pm 2.4$ years in group I, and it increased to $58.1 \pm 1.1$ years in group II, as against 60 years in the control group, i.e. the actual reductions in average life expectancy for the two groups were 11 and 2 years, respectively. After 7–13 years the average life expectancy in groups I and II was $62.0 \pm 4.2$ and $68.1 \pm 1.6$ years, respectively, as against 68.1 in the control group. Only in the 14–19 year period did the differences between groups I and II level out.

The maximum reductions in average life expectancy at effective dose equivalents of 1400 mSv and 500 mSv were 12.9% and 3.6% of the normal life expectancy or 23 days and 18 days per 10 mSv of effective dose equivalent. In the model adopted, the reduction of the average life expectancy is 7–8 days per 10 mSv.

### TABLE III. CHILD MORTALITY UP TO THE AGE OF 1 YEAR

<table>
<thead>
<tr>
<th>Years of radiation effect</th>
<th>Number of deaths per 10^3 births</th>
<th>Excess mortality compared with the expected level (10^3)</th>
<th>Average dose on bone surfaces (mSv)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>I</td>
<td>II</td>
<td>Groups I and II combined</td>
</tr>
<tr>
<td>Control</td>
<td>$92.9 \pm 10.2$</td>
<td>$103.6 \pm 8.0$</td>
<td>0</td>
</tr>
<tr>
<td>1–2</td>
<td>$150.9 \pm 17.4$</td>
<td>$126.0 \pm 12.2$</td>
<td>$57.3 \pm 5.8$</td>
</tr>
<tr>
<td>3–6</td>
<td>$127.5 \pm 10.4$</td>
<td>$97.1 \pm 7.7$</td>
<td>$22.9 \pm 1.8$</td>
</tr>
<tr>
<td>7–10</td>
<td>$100.2 \pm 11.2$</td>
<td>$60.0 \pm 6.1$</td>
<td>$15.5 \pm 1.7$</td>
</tr>
<tr>
<td>11–13</td>
<td>$95.6 \pm 15.2$</td>
<td>$38.2 \pm 7.8$</td>
<td>$0.4 \pm 0.06$</td>
</tr>
</tbody>
</table>
TABLE IV. MORTALITY COEFFICIENT FOR ALL MALIGNANT TUMOURS PER $10^5$ PERSONS

<table>
<thead>
<tr>
<th>Race</th>
<th>Dose (mSv)</th>
<th>Average mortality coefficient</th>
<th>Percentage increase compared with spontaneous level</th>
</tr>
</thead>
<tbody>
<tr>
<td>Russian</td>
<td>1400</td>
<td>267.9 (186.5-372)(^a)</td>
<td>58</td>
</tr>
<tr>
<td>Tatar</td>
<td>520</td>
<td>165 (132.2-205)(^a)</td>
<td>45</td>
</tr>
<tr>
<td>Tatar</td>
<td>240</td>
<td>127.7 (101-158.9)</td>
<td>12</td>
</tr>
<tr>
<td>Russian</td>
<td>111</td>
<td>215.6 (169-271)</td>
<td>27</td>
</tr>
<tr>
<td>Russian</td>
<td>75</td>
<td>191.4 (159-237)</td>
<td>13</td>
</tr>
<tr>
<td>Russian</td>
<td>0</td>
<td>168.7 (152-186.5)(^b)</td>
<td>0</td>
</tr>
<tr>
<td>Tatar</td>
<td>0</td>
<td>114.1 (103.4-125.3)(^b)</td>
<td>0</td>
</tr>
</tbody>
</table>

\(^a\) Statistically significantly higher than control.
\(^b\) Control.

of effective dose equivalent. Evidently, the actual doses received by some of those who died may have been substantially higher than the average values.

The information in Table IV shows that only at doses of 1400 mSv among the Russian population and 520 mSv among the Tatar population was there found to be a clear increase in the mortality factors for all malignant neoplasms as compared with those of the control group.

For 1 Sv of effective dose equivalent the increase in tumour occurrence is 45-58\%, compared with the spontaneous level: Calculation shows that the risk of death per $10^4$ man·Sv of dose equivalent from all malignant neoplasms is 8.34-14.85, of which deaths from leukaemia account for 1.48-1.92, cancer of the oesophagus for 1.89-1.83, stomach cancer for 1.7, cervical cancer for 2.1-8.46 and skin cancer for 0.9.

In the course of 35 years' observation the excess number of leukaemia cases was 14, with an average individual dose of 540 mSv and a collective dose of 6772 Sv. The absolute risk was therefore $2 \times 10^{-3}$ Sv$^{-1}$, which is in line with ICRP Publication 26. The combined risk for all malignant tumours is 1.25-5.0, or 175-500 cases per $10^4$ man·Sv [1, 5].

On 29 September 1957 the rupture of a storage tank in a repository for highly radioactive waste at the Mayak radiochemical complex led to the release of $7.4 \times 10^{16}$ Bq of long lived waste into the atmosphere [5-10]. The radioactive trail, up to 300 km long and up to 50 km wide, produced various fallout levels. The maximum level for $^{90}$Sr alone amounted to $1.5 \times 10^8$ Bq/km$^2$. 
On the basis of hundreds of measurements it has been established that the activity of the local diet is 140 pCi with 1 Ci per km$^2$ [5, 10]. Therefore, with 4 Ci per km$^2$ (148 kBq/m$^2$) the diet absorbs 560 pCi per day or 0.2 µCi per year (7800 Bq). But, according to the ICRP and the National Radiation Safety Standards, the chronic $^{90}\text{Sr}$ intake should not exceed 0.32 µCi per year (11 840 Bq). The margin thus existing in respect of intake and hence dose made it possible to plan appropriate emergency measures, including resettlement.

The dynamics of the radiation situation have changed constantly. After 25 years, the total radioactivity had decreased by a factor of 33, the $^{90}\text{Sr}$ activity by a factor of 2, and the exposure rate by a factor of 2600. After 75 years, the dose rate will have decreased by a factor of 9000, while within 15 years of the accident the radiation background was already no different from the natural gamma background level [7, 9]. However, in the early stages, exposure levels were significant and, at various times after the accident, planned evacuations were carried out in order to prevent overexposure. Delay in evacuation from places where people in group A lived may have led to overexposure and the occurrence of deterministic effects.

The examination of 2055 people (1032 children) revealed, in a number of cases, the presence of leukocytosis, leukopenia and thrombopenia. Fifteen months after the accident, no deviations from the control group were found among those resettled from group A (dose 520 mSv) (152 adults and 93 children) or those from points D (56 mSv) [1, 5, 10].

The bulk of the irradiated population was examined in the first weeks and months after the accident, and the 153 people who had been working in close proximity to the site of the explosion were hospitalized. They had received an instantaneous dose of approximately 1 Sv. A moderate decrease was noted in their leukocyte and lymphocyte counts. Stab neutrophils had developed in 50% of them, 20% being in juvenile Dorlands form. The lymphocyte count in 71% of those examined was no more than 1800 per mm$^3$, and in 14% of them it was less than 1300 per mm$^3$. Radiation sickness was not diagnosed.

After two years, more cases of leukocytosis were observed in those who had been irradiated (32.5% versus 7.5%), with >9000 per mm$^3$, as well as more cases of thrombocytosis (13.8% versus 2.5%), with 350,000 per mm$^3$, although all the average indices agreed with those of the control group.

The picture with regard to general somatic diseases among the resettled inhabitants is typical of a rural location. Among the adults there are abnormalities in 52% of the cases, involving diseases of the cardiovascular system, respiratory organs and digestive system.

Thirty years after the accident, a proportion of the people examined were listed as healthy, and 47% of the remainder as having a chronic, but not acute, infection.

$2 \ 1 \ \text{Ci} = 3.7 \times 10^{10} \ \text{Bq}.$
TABLE V. DATA ON EARLY CHILD MORTALITY FROM 1957 TO 1961 AND MORTALITY DUE TO CONGENITAL ANOMALIES PER 1000 NEONATES

<table>
<thead>
<tr>
<th>Cause of death</th>
<th>Area of the radioactive trail</th>
<th>Control areas</th>
</tr>
</thead>
<tbody>
<tr>
<td>Have congenital anomalies</td>
<td></td>
<td>Edge of the radioactive trail (0.1 Ci/km²)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Far from the radioactive trail</td>
</tr>
<tr>
<td></td>
<td></td>
<td>All of the Chelyabinsk region</td>
</tr>
<tr>
<td>All causes except congenital anomalies</td>
<td>27.7</td>
<td>31.4</td>
</tr>
<tr>
<td></td>
<td>38.6</td>
<td></td>
</tr>
<tr>
<td>Alimentary disorders</td>
<td>15.2 ± 2.8</td>
<td>12.3 ± 3.0</td>
</tr>
<tr>
<td></td>
<td>5.0 ± 1.0</td>
<td></td>
</tr>
<tr>
<td>Pneumonia</td>
<td>1.7 ± 1.0</td>
<td>3.1 ± 1.5</td>
</tr>
<tr>
<td></td>
<td>16.1 ± 1.8</td>
<td></td>
</tr>
<tr>
<td>Infections</td>
<td>1.6 ± 0.9</td>
<td>2.3 ± 1.3</td>
</tr>
<tr>
<td></td>
<td>3.0 ± 0.8</td>
<td></td>
</tr>
<tr>
<td>Illnesses among neonates</td>
<td>8.7 ± 2.2</td>
<td>13.8 ± 3.2</td>
</tr>
<tr>
<td></td>
<td>14.5 ± 1.7</td>
<td></td>
</tr>
<tr>
<td>Congenital anomalies</td>
<td>1.92</td>
<td>1.93</td>
</tr>
<tr>
<td></td>
<td>2.66</td>
<td></td>
</tr>
</tbody>
</table>

Three people developed epilepsy owing to trauma. Nothing unusual was observed in the pattern of the somatic illnesses among the irradiated population.

Among the radiation effects that have been determined, early child mortality and congenital anomalies provide a sensitive indicator. It can be seen from Table V that in the period 1957–1961, early child mortality was high, both inside and outside the contaminated area. No differences were found, either in overall child mortality or in respect of individual causes of death.

Among the adult population there have been variations in the mortality rate pattern. Among those from group A resettled within a year, the mortality rate was $13.3 \times 10^{-3}$ — distinctly higher than among those who received a lesser dose and were resettled within 250–650 days. Over the next 30 years, the standardized mortality rate was $(8.3–10.4) \times 10^{-3}$ per year, as in the control group [2, 3].

Among the groups examined that had sustained various doses, delayed consequences in the form of malignant tumours did not occur with excessive frequency. The lower frequency of neoplasms among the inhabitants who had sustained the highest dose — 79 cases against 129–157 cases in other groups, per $10^5$ per year — is probably explained by the comparatively small size of the group, which comprised 1054 people.

On 10 October 1957, an accident occurred in a nuclear processing plant at Sellafield which led to the release of 1000 TBq of radioactive materials [11, 12], including $(0.6–1.1) \times 10^{15}$ Bq of $^{131}$I, $8.5 \times 10^{12}$ Bq of $^{210}$Po, $4.6 \times 10^{12}$ Bq of
The ratio of the radionuclides in the release differed from that in the Southern Urals accident, but the ratio of $^{131}$I to $^{137}$Cs and $^{90}$Sr was consistent with the releases from Chernobyl [11].

At a distance of 3–6 km from Sellafield, the contamination levels found in grass were 370 000 Bq/m$^2$ for $^{131}$I; 9000 Bq/m$^2$ for $^{137}$Cs; 7000 Bq/m$^2$ for $^{103-106}$Ru and 11 000 Bq/m$^2$ for $^{95}$Zr–$^{95}$Nb; 60 Bq/m$^2$ for $^{90}$Sr and 270 Bq/m$^2$ for all alpha emitters. Increased $^{131}$I contents in air, grass and cows’ milk were detected at a distance of 400 km from the source.

As a result of the measures taken, thyroid doses in adults and children were successfully reduced. The theoretical and measured doses do not always coincide, particularly for the children. Naturally, some of the calculations were not perfect, and the health countermeasures were not always observed. The thyroid doses of children were 2–8 times higher than those of adults. It should be noted that after the Chernobyl accident the thyroid doses of children were 6.5 times higher than those of adults [11].

The collective effective dose equivalent commitment in man·Sv has been calculated by British scientists. The contribution by external gamma radiation was around 10%, the main contribution being due to inhalation. In Cumbria, even though a ban was imposed on milk distribution, the collective dose contribution from milk was 59%.

The health status of the population and of those who witnessed the Windscale accident has been studied in detail for over 35 years. The health of the 500 employees who dealt with the accident has been monitored, together with that of 2500 employees who worked there since 1957 and of 14 000 employees who worked there before the accident. Epidemiological studies have also been conducted among the half million inhabitants of the county of Cumbria, where the thyroid doses of adults are within 20 mSv and those of children are in the range of 40–160 mSv. The analysis showed no abnormalities in health indicators. On the contrary, the actual morbidity is less than that expected and less than that in the control group. On the basis of a linear non-threshold model of the relationship between tumour occurrence and dose it has been shown that, of the 100 potential cancers in 36 years of surveillance, 66 were caused by natural radiation, 14 by emissions from the plant, nine by the fallout from weapons testing, nine by medical irradiations and two by the fire at Windscale. These calculations disregard the attenuating effect of dose protraction on tumour induction. No increase in thyroid tumours was recorded in the 36 years of the study.

The destruction of Unit 4 of the Chernobyl nuclear power plant in 1986 led to the release into the atmosphere of $10^{18}$ Bq of radionuclides and to the overexposure of large numbers of people. The dose levels and the consequences thereof have now been studied in sufficient detail. For example, it has been found that diseases of the cardiovascular system, respiratory organs and nervous system among
TABLE VI. MORTALITY RATES OVER 30 YEARS, $10^3$ PER YEAR

<table>
<thead>
<tr>
<th>Dose at time of accident (mSv)</th>
<th>720 to &gt; 500</th>
<th>496</th>
<th>240</th>
<th>150 (50-300)</th>
<th>120</th>
<th>90</th>
<th>40</th>
<th>$^{239}$Pu operators</th>
<th>Control</th>
</tr>
</thead>
<tbody>
<tr>
<td>Area</td>
<td>Techa, Sellafield</td>
<td>Southern Urals</td>
<td>Techa</td>
<td>Chernobyl</td>
<td>Southern Urals</td>
<td>Techa</td>
<td>Southern Urals</td>
<td>Sellafield</td>
<td>All areas</td>
</tr>
<tr>
<td>Mortality rate</td>
<td>6.6-8.7</td>
<td>8.6</td>
<td>4.9</td>
<td>2.6-8.8</td>
<td>8.9</td>
<td>4.7</td>
<td>9.4</td>
<td>8.3</td>
<td>5.3-9.0</td>
</tr>
<tr>
<td>Main characteristics</td>
<td>River Techa (data by Kosenko [1])</td>
<td>Atomic bomb survivors (data by Shimizu)</td>
<td>Radiation treatment of spondylitis (data by Darby)</td>
<td>Radiation treatment of cervical cancer</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>---------------------------------------</td>
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<td>---------------------------------------</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Numbers observed</td>
<td>28 000</td>
<td>42 000</td>
<td>14 000</td>
<td>83 000</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sex ratio, percentage of women</td>
<td>56%</td>
<td>59%</td>
<td>17%</td>
<td>100%</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Age at irradiation (years)</td>
<td>0–90</td>
<td>0–90</td>
<td>&gt;15</td>
<td>&lt;30 to &gt;70</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Type of control</td>
<td>Regional</td>
<td>Internal</td>
<td>National</td>
<td>National</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Type of irradiation</td>
<td>γ, 89Sr, 90Sr, 137Cs</td>
<td>Intense gamma irradiation</td>
<td>Fractionated, local</td>
<td>Chronic, fractionated</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Type of dosimetry</td>
<td>Personal, internal and calculated external</td>
<td>DS-86</td>
<td>Personal</td>
<td>Average doses from sample</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average doses (Gy)</td>
<td>0.4</td>
<td>0.24</td>
<td>1.9</td>
<td>Maximum unknown</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dose range (Gy)</td>
<td>0–3.0</td>
<td>0.01–6.0</td>
<td>0–8.6</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Man-years of persons at risk</td>
<td>422 000</td>
<td>1 134 000</td>
<td>184 000</td>
<td>623 800</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Absolute risks per 10^4 man-years (Gy)</td>
<td>0.48–1.10</td>
<td>2.94</td>
<td>2.02</td>
<td>0.61</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
the 'liquidators' of 1986 are clearly more frequent than among the 'liquidators' of 1988. The doses recorded were, respectively, 190 ± 50 mSv and 20 ± 7 mSv for the period worked.

Over the period since the accident, an increase in the incidence of thyroid cancer has been found in children among the irradiated population of Belarus, Russia and Ukraine [11]. The frequency of other malignant tumours, including leukaemias, has not increased.

A comparative analysis of the medical consequences of the four most serious accidents is presented in Tables VI and VII. It can be seen that local accidents, even those accompanied by the accumulation of large absorbed doses, do not substantially affect the mortality rate for the irradiated population if the dose range is within 720 mSv at the time of the accident.

All types of radiation involve a risk of leukaemia occurring, the risk being twice as high in the case of acute and protracted irradiation.

The frequency of occurrence of thyroid tumours is undoubtedly the consequence of irradiation of that organ and is a function of the dose and of the age of the irradiated person. There are grounds for supposing that the role of the collective dose in the development of this pathology is of minimal significance.

REFERENCES


ENVIRONMENTAL REMEDIATION

(Session 8)

Chairman

M. BALONOV

Russian Federation
RADIOLOGICAL CRITERIA FOR THE PROTECTION OF THE PUBLIC FROM RADIONUCLIDES IN THE ENVIRONMENT

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Abstract

RADIOLOGICAL CRITERIA FOR THE PROTECTION OF THE PUBLIC FROM RADIONUCLIDES IN THE ENVIRONMENT.

The advice on public protection given by the National Radiological Protection Board for application in the United Kingdom is broadly in line with recent international recommendations. For practices, the system of protection is based on the principles of justification, optimization and limitation of dose or risk. However, application of the principles of justification and constrained optimization in practical circumstances has provoked some debate. The paper discusses some of the issues raised. There may be situations where land is contaminated with radioactive material from previous practices but, whilst access is prohibited, there is no exposure of the public. However, if a change of use of the land is proposed, such as de-licensing a nuclear site or sale of premises contaminated with radionuclides, this may lead to a situation where members of the public may be exposed. It seems appropriate to judge this situation according to the same criteria as those applied for practices. Development of advice on appropriate criteria is described. If an accidental release of radionuclides occurs, intervention may be required in order to avoid or reduce doses. At the time of the accident, or in its immediate aftermath, it may be necessary to implement urgent countermeasures to avoid short term relatively high doses. However, in the event of a very serious accident, there may be a need to consider long term countermeasures. Drastic countermeasures, such as relocation, would be considered only for the purpose of avoiding substantial doses and only if other methods of reducing doses were not effective. For small dose savings, other procedures such as decontamination would be considered so as to reduce doses, avoid restriction on individuals' lifestyles and so promote a return to normality. Similar considerations apply to situations where contamination from past practices is discovered on land to which the public has access.

1. INTRODUCTION

Within the United Kingdom the National Radiological Protection Board is directed by health ministers to advise the appropriate government departments and statutory bodies on the acceptability of the recommendations or proposals to, and their application in, the UK whenever the International Commission on Radiological Protection (ICRP) and other bodies publish recommendations or proposals. The
ICRP has published new recommendations in Publication 60 [1] which take account of new biological information on the effects of radiation and the trends in setting safety standards. The Board has considered these recommendations, formulated draft advice, which was widely disseminated for consultation, and subsequently published its advice to the government [2, 3]. Meanwhile, the International Atomic Energy Agency (IAEA) has been revising its Basic Safety Standards [4]. All these documents will be considered in advice to government on the revision of the Euratom Basic Safety Standards Directive.

The Board's advice on public protection [3] focused on practices and touched on potential exposures. Whilst every effort is made to formulate clear advice, it seems inevitable that some questions will arise concerning the interpretation and application of that advice. Some of the issues that have arisen are discussed in this paper.

There is currently no international guidance on radiological criteria for restoration of contaminated sites. The Board is currently developing advice on such criteria for release or change of use of contaminated land, i.e. situations which can be dealt with using the same criteria as those for practices.

The Board also has the responsibility for specifying emergency reference levels (ERLs) to guide the implementation of countermeasures to protect the public in the event of an accidental release of radionuclides to the environment. The Board had issued advice on urgent countermeasures in 1990 [5] and this advice was consistent with that given by the ICRP [6]. However, both the ICRP [6] and the IAEA [4] have formulated criteria for long term countermeasures after accidental releases and the Board is currently developing guidance in this area for application in the UK. The question of the criteria to be applied when contamination is discovered in the public environment is also being addressed.

This paper brings together these various strands of Board advice and discusses some of the issues that have arisen.

2. PRACTICES AND INTERVENTION —
THE PRINCIPLES TO BE APPLIED

International guidance now distinguishes between 'practices' and 'intervention' and, in general, this distinction has been found to be helpful. Some activities increase exposure to radiation by introducing new sources, pathways and individuals or by modifying existing exposure pathways from existing sources to humans. These activities, which add exposures, are known as practices and the system of protection recommended by the ICRP for practices is based on the following general principles:
(a) No practice involving exposure to radiation should be adopted unless it produces sufficient benefit to the exposed individuals or to society to offset the radiation detriment caused — justification of a practice.

(b) In relation to any particular source within a practice, the magnitude of individual doses, the number of people exposed and the likelihood of incurring exposures when they are not certain to be received should be kept as low as reasonably achievable, economic and social factors being taken into account (ALARA principle). This procedure should be constrained by restrictions on the doses to individuals (dose constraints) or the risk to individuals (risk constraints) so as to limit the inequity likely to result from the inherent economic and social judgements — optimization of protection.

(c) The exposure of individuals resulting from the combination of all relevant practices should be subject to dose limits or to some control of risk in the case of potential exposures. These are aimed at ensuring that no individual is exposed to radiation risks from these practices that are judged to be unacceptable in normal circumstances. Not all sources are amenable to control and it is necessary to specify which sources are to be included as relevant — individual dose and risk limits.

There are some situations, such as accidental release of radionuclides to the environment or the normal presence of naturally occurring radionuclides in the environment, where doses will be received unless measures are taken to prevent or reduce them. The reduction of dose is achieved by a process of intervention. Implementation of a countermeasure will lead to some harm (e.g. social disruption and monetary cost) and this must be balanced against the benefit (primarily the dose reduction). The system of radiological protection for intervention is based upon the following principles:

(a) The proposed intervention should do more good than harm, i.e. the reduction in detriment resulting from the reduction in dose should be sufficient to justify the harm and the costs, including social costs, of the intervention.

(b) The form, scale and duration of the intervention should be optimized so that the net benefit of the reduction in dose, i.e. the benefit of the reduction in radiation detriment less the detriment associated with the intervention, is maximized.

Furthermore, serious deterministic effects should be avoided by introducing countermeasures to keep doses to individuals to levels below the thresholds for these effects. This principle overrides the other two. However, it should rarely be important in practice since, for most situations, the introduction of countermeasures will be justified and protection optimized at levels of individual dose well below those at which serious deterministic effects occur. Dose limits applied to practices are not relevant in decision making or intervention.
3. PROTECTION IN PRACTICES

3.1. Justification

Justification of a practice goes far beyond the scope of radiological protection alone. Other factors to be considered include the benefits supplied by the practice, social factors and non-radiological forms of detriment. Decisions on such matters are made by society through elected members and public debate.

To take nuclear power as an example, the Board's view is that justification should apply to the entire practice, including waste disposal, not to each individual nuclear power reactor, nor to waste disposal in isolation. Other issues that should be considered include the energy supply strategy and the environmental impact of alternative means of generating power. In this context, the principle of justification requires consideration of a long term strategy. It would not be sensible to review these elements of a justification decision on a frequent basis, since many of the factors considered relate to long time periods. Justification of a practice should be reviewed only if circumstances change significantly. However, even in the event of a significant change which might lead to an existing practice being considered no longer justified if it were to be considered anew, it may equally be unjustified to withdraw it. The consequences of replacing that practice with another would need to be considered.

However, a judgement in the English courts ruled that the process of justification should be applied to a particular activity, at a particular site and at a particular time. As a consequence, the UK authorizing departments are now requiring a statement on justification each time an authorization for disposal is renewed.

3.2. Optimization

Optimization of protection for practices is the balancing of the benefit of radiation dose reduction against the resources expended to achieve that reduction. The Board has recommended that the optimization process should be constrained by restrictions on the doses or risks to individuals. A dose constraint is an upper bound on the annual dose to members of the critical group resulting from the planned operation of a controlled source. The Board has recommended a maximum dose constraint of 0.3 mSv/a for controlled sources and has noted that lower values could be set where such doses are readily achievable. The dose constraint is a prospective concept and applies to the current and future operation of a planned source. Furthermore, the Board stated that it expected existing sources to be operated within the dose constraint. However, it recognized that some existing sources were designed to meet less stringent standards and so the Board recommended that, if an existing source could not comply with the appropriate dose constraint, then the regulatory bodies should ensure that the doses were as low as reasonably achievable and within the dose limit.
An important aspect of the dose constraint is that it applies to the annual dose to the critical group, summed over all exposure pathways, arising from the current and future operations of a controlled source. That raises the question of what is meant by ‘source’. Nuclear sites in the UK may have a number of different buildings from which discharges of radioactive materials into the environment occur. There may also be more than one operator on a site. In considering what is a ‘single controlled source’, it is necessary to ensure that the exposure of members of the public is kept within acceptable levels. Any radioactive discharges from a single site, even if they are from different locations on the site, are likely to lead to exposure of the same group of people. It is important, therefore, to consider all sources, new and old, on a site in assessing doses to the critical group. This may also apply to sites which are divided between a number of operators, but who have a common purpose.

Radiation doses for comparison with the dose constraint should be calculated for all routine discharges of radionuclides to the atmosphere and to water bodies from the whole site together with discharges due to direct radiation from the site itself. The doses should be calculated for all exposure pathways and summed in a realistic manner. Account should be taken of the discharges continuing into the future, but not of historical discharges. The resulting estimate of dose should then be compared with the appropriate dose constraint. If the estimated dose exceeds the dose constraint because of doses from new plant or new operations on the site, then action would be required to reduce the doses to below the relevant constraint. If, however, the estimated dose is largely due to operation of old plant on the site, then the situation would need to be reviewed. Provided that the dose limit was adhered to and the doses were considered to be as low as reasonably achievable, the regulatory authorities may decide that the situation is acceptable.

3.3. Limitation

The Board has recommended that the annual dose limit for members of the public should be 1 mSv/a. This encompasses all controlled sources, excluding occupational and medical sources, and does not include exposure to natural radiation. Furthermore, the Board recommends that exposure from past controlled releases should be included in any comparison with the dose limits.

4. CRITERIA FOR INTERVENTION

4.1. Emergency reference levels

In the event of an accidental release of radionuclides to the environment, dose limits do not apply. Instead, it is necessary to have criteria which represent a
TABLE 1. EMERGENCY REFERENCE LEVELS FOR EARLY COUNTER-MEASURES

<table>
<thead>
<tr>
<th>Countermeasure</th>
<th>Body organ</th>
<th>Dose level (mSv)</th>
<th>Lower</th>
<th>Upper</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sheltering</td>
<td>Whole body</td>
<td>3</td>
<td>30</td>
<td>300</td>
</tr>
<tr>
<td></td>
<td>Thyroid, lung, skin</td>
<td>30</td>
<td>300</td>
<td>3000</td>
</tr>
<tr>
<td>Evacuation</td>
<td>Whole body</td>
<td>30</td>
<td>300</td>
<td>3000</td>
</tr>
<tr>
<td></td>
<td>Thyroid, lung, skin</td>
<td>300</td>
<td>3000</td>
<td></td>
</tr>
<tr>
<td>Stable iodine</td>
<td>Thyroid</td>
<td>30</td>
<td>300</td>
<td></td>
</tr>
</tbody>
</table>

judgement on the balance between the benefits and harms of intervention. The major benefit of intervention is, of course, the reduction in individual and collective dose, but reassurance may also be counted as a benefit. The disadvantages of intervention include: individual and collective physical risks, monetary costs, social and individual disruption and anxiety. The Board has given advice on the ranges of emergency reference levels for urgent countermeasures (Table I). The Board's advice [5] was published before the most recent international recommendations on this matter [1, 4, 6], but is compatible with that guidance.

The Council of the European Communities has issued Regulations [7-9] which specify intervention levels for radioactive contamination in marketed foods and animal feeds. These would be legally binding in the UK following a future accidental release of radioactive material to the environment. The Board has considered the radiological implications of those levels: if restrictions were imposed on foods at the levels specified, then the doses received by most individuals in the UK would be very small. Control of foods at lower levels would not be warranted.

4.2. Long term intervention

International guidance [1, 4, 6] now includes criteria for long term intervention after a release of radioactive material to the environment.

Following a very severe accident, it is possible that the levels of contamination in the environment may result in continuing potential exposures that are considered unacceptable. In this case it would be necessary to advise people to leave the contaminated area or, if they had been evacuated, to advise against their return until
such time as the potential exposures had reduced to more acceptable levels, whether naturally, through radioactive decay and weathering, or as a result of decontamination measures. This removal of people from an area for weeks, months or years is termed relocation. Whilst the Board has not yet given any formal advice on this matter for the UK, there has been considerable discussion on this topic. The ICRP states that relocation would be almost always justified if it could save a dose of 1000 mSv and if the range of optimized values for relocation is 5-15 mSv per month. The inter-agency Basic Safety Standards [4] give generic optimized intervention levels for initiating and terminating temporary relocation of 30 mSv per month and 10 mSv per month, respectively. If the dose accumulated in a month is not expected to fall below this level within a year or two, permanent resettlement with no expectation of return should be considered. Permanent resettlement should also be considered if the lifetime dose is projected to exceed 1000 mSv.

Relocation is a countermeasure very different from evacuation. The purpose of relocation is to protect individuals in the long term from chronic, continuing exposure from radionuclides deposited in the environment after an accident. Evacuation is necessarily a short duration countermeasure which is expected to continue for no more than a day or so, after the source of the accidental release had been made safe. Its purpose is to avoid a ‘one-off’ exposure. Relocation, by contrast, is a long term countermeasure to provide protection against a level of exposure that is considered unacceptable because of its long duration. It is the week on week, month on month, or year on year exposure that is of concern.

Any decision on relocation would be a complex matter, involving social and monetary considerations, as well as the potential dose saved. Relocation may be regarded as a countermeasure of last resort: other actions would be considered to reduce doses before relocation was judged necessary. To guide decisions on long term intervention after an accident, it may be helpful to specify an ‘action level’. If doses in the long term were to exceed the action level, some action should be taken to reduce doses. This could be decontamination or temporary relocation whilst decontamination measures are carried out. If doses were below the action level, this does not mean that no action would be taken to reduce doses. Indeed, if simple measures could reduce doses, they would be taken.

These matters are still under discussion within the UK and no numerical criteria have been formally adopted. However, some perspective can be obtained by considering advice on action levels for remedial measures against radon in homes. The ICRP suggests that some remedial measures are almost always justified when the continued annual effective dose is above 10 mSv [10]. For simple remedial measures, a somewhat lower figure could be considered. Thus, the ICRP recommends that the action level for radon in homes should be within the range of 3-10 mSv/a.

One way forward might be to consider a graded response to long term countermeasures following a severe accident. If high dose rates were likely to persist for
a short while (months), then decisions on relocation or decontamination would be guided by ICRP's criterion of 5-15 mSv/month. However, if doses were likely to persist at unacceptably high levels for a period of years, then some action to reduce doses would be required if they were to exceed 10 mSv/a. Below this level, measures may be taken to reduce doses, and decisions on this would be guided by consideration of the costs and benefits. However, it would not be sensible to expend effort to reduce doses below the annual dose limit.

4.3. Contaminated land

There is as yet no international advice on the restoration of sites contaminated with radioactive material, other than that noted above for severe accidents. Land may become contaminated as a result of the normal operation of practices, including minor incidents whose consequences are or were confined to the premises of the practice. The distinction between practices and intervention is generally helpful, but in the situation of land contamination it may not always be clear. For example, where contaminated land is not currently accessible to the public, remedial action taken solely to reduce actual or potential exposures would constitute intervention. However, if the land was subsequently reused in such a way as to allow public access, then this would introduce new exposures from the activity remaining after remedial action; such exposures might reasonably be expected to be subject to criteria similar to those arising from practices.

Nevertheless, two main categories of situation can be identified, as follows:

(a) A change of use is proposed such that an area of land, known to be contaminated, to which members of the public do not currently have access will become accessible, e.g. redevelopment of an industrial site for housing, or 'release' of land from a nuclear site after decommissioning. In this case (assuming that not all of the contamination is removed), new groups of people will become exposed to an existing (and currently controlled) source, indicating that the system of control for practices should be used. The practice itself may no longer exist, but the exposures would continue to be regarded as a subset of those arising from the practice.

(b) Contamination is discovered on land already in use in the public domain. In this case, exposures will already be occurring — the source, exposure pathways and exposed people are already in place — and therefore this situation should be considered as a candidate for intervention.

The extent of any remediation of contamination should be such that the future exposures of occupants of the site, as well as other exposures such as those of workers performing remedial work, are as low as reasonably achievable, economic and social factors being taken into account.
In order to ensure that risks to the public will not be unacceptable, it is considered that the excess risks to the critical group, attributable to the residual contamination resulting from the future use of the site, should not exceed $10^{-5}$ a$^{-1}$. This is numerically equal to the risk constraint recommended by the Board for future exposures arising from the land based disposal of solid radioactive wastes [11]. For extended exposures, this also corresponds to the risk associated with an annual effective dose of about 0.3 mSv/a, the maximum dose constraint recommended by the Board for the operation of new controlled practices.

Optimization will, in general, require consideration of the full range of options, from the 'null option' (no remedial action) to complete decontamination. Other options may include actions to immobilize or isolate the radioactive material without removing it from the site. In evaluating these options, the expected future use of the site will be important, since this will determine the likely future exposures as well as the costs and doses associated with the remediation work (if any).

A number of quantitative techniques exist that may be used to assist in performing optimization studies. However, it is unlikely that simple techniques such as cost/benefit analysis will be adequate for contaminated land situations, where factors such as public perception of the site are often important — multiattribute decision aiding methods are likely to be more appropriate.

In these situations, it is useful to provide guidance on a level of future exposure below which the optimization requirement can be relaxed, i.e. in this case a level below which it is not necessary to consider the application of significant resources to achieve further reductions.

In its advice on land based disposal of solid radioactive waste, the Board recommended a 'design target' of $10^{-6}$ a$^{-1}$, representing a level of risk to the most exposed group considered to be broadly acceptable. For contaminated land situations, it is unlikely that significant expenditure to reduce the excess risk to an average member of the critical group of future site occupants to below about $10^{-6}$ a$^{-1}$ would be warranted. However, there may be simple and inexpensive measures that could further reduce the risk; clearly, such measures should be taken if they are apparent.

In cases where there is existing exposure of occupants on sites contaminated with the residues of past practices, the principles of protection in intervention are applicable. In particular, the dose limit for members of the public does not apply in these situations.

In these cases, it is necessary to justify any remedial actions and to optimize the level of remediation on a case by case basis. The optimization should take account of all the relevant factors, including health effects, monetary costs and social factors, such as disruption to the lives of the population, and the origin of the contamination.

A very wide range of contaminated land situations can be envisaged that would fall into the 'intervention' category, involving widely differing levels (and half-lives) of contamination, affected areas and possible remedial actions. If low levels of
residual dose can be achieved relatively easily, without incurring large costs or causing significant disruption, then the optimum level of intervention would be expected to result in a level of protection approaching that recommended for practices. If it is anticipated that the remediation will involve severe disruption (e.g. requiring people to leave their homes for weeks or months), major expense or serious non-radiological health effects, then much higher levels of residual dose would be tolerable.

5. SUMMARY

The Board has a duty to give advice to UK government departments on the radiological protection criteria to be applied to the protection of the public from radiation hazards, specifically from radionuclides present in the environment. The Board has given formal advice on control of doses from practices and on criteria for early intervention measures. This advice has been summarized in this paper. The Board is continuing to develop advice for application in other circumstances of public exposure, in particular exposure that may continue for extended periods of time. Two such circumstances have been considered in this paper: long term exposure, following a severe accident that released radionuclides to the environment; and release of a contaminated site to the public domain. Although the origins are different, both may lead to situations where members of the public may be exposed for long periods of time. It is suggested that the release of contaminated land should be dealt with according to the principles applied to a practice: the public dose limit applies and a constraint can be set. For intervention in the long term after an accident, the cost, both monetary and social, must be balanced against the benefit of dose reduction. Costly and disruptive measures are likely to be justified only for a substantial dose saving, 10 mSv/a or more, but, if simple measures can reduce doses, then they should be taken. International advice on reduction of radon levels in homes provides some perspective to guide decisions.

REFERENCES


RADIOLOGICAL IMPACT ON THE ENVIRONMENT DUE TO MINING AND MILLING OF URANIUM BEARING HARD COAL IN CENTRAL GERMANY (FREITAL REGION, SAXONY)

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Abstract

RADIOLOGICAL IMPACT ON THE ENVIRONMENT DUE TO MINING AND MILLING OF URANIUM BEARING HARD COAL IN CENTRAL GERMANY (FREITAL REGION, SAXONY).

The Freital region is a densely populated and economically intensively used area near the capital of Saxony. The infrastructure of this region is characterized by mining and processing of hard coal, which started in the 16th century and was abandoned in 1968. After World War II, the Soviet-German company Wismut started mining and milling of uranium bearing coal and black shale as well as reprocessing of mining wastes. The production of uranium was abandoned, for commercial and other reasons, and the last pit was closed in 1990. Problems of radiological protection may be associated with the relics and properties from mining, milling, processing and burning of the coal. In particular, utilization of wastes and of former mining and milling sites may lead to radiation exposure of the public which is higher than that due to natural (terrestrial) radiation. As in other eastern German mining districts, the Bundesamt für Strahlenschutz (BfS) has investigated the situation in the framework of a Federal project. On the basis of recommendations by the German Commission on Radiological Protection, the BfS developed a multistage procedure in order to register and assess the relics, with the aim of defining those objects that have to be considered as relevant sources of environmental contamination and radiation exposure of the public. The following types of objects have to be considered as sources of radiation exposure of the public: Sites of abandoned uranium mills that had not been decontaminated but had been used for other industrial purposes; areas open to the public where mining residues and mill tailings have been dumped carelessly; areas, paths or roads covered by coal ash or slags; and waste rock piles, especially deposited slags and other burning residues. The total area that has to be classified as being relevant is small compared with the area investigated. The 'radiologically relevant' objects, sites and properties have been identified and should be subjected to remedial actions, taking into account aspects of radiological protection and other health and environmental protection, as well as the interest of communities, costs, social aspects, and technical and financial resources.
1. INTRODUCTION

Since the Middle Ages, industrial development has caused many relics that are still possible sources of emissions into the atmosphere, geosphere and biosphere. The exposure to substances discharged via various pathways and over different time-scales may have damaging consequences to the public.

It is necessary to consider the kind and extent of remedial actions that have to be carried out, taking into account the level of financial and economic resources of the society as well as the awareness of the public. Well founded criteria regarding acceptable levels of relevant harmful substances in the environment and an acceptable level of exposure of man are required for deciding whether remedial actions are necessary or not.

2. MINING AND MILLING IN THE FREITAL REGION

Mining and milling of various raw materials has a centuries old tradition in certain parts of eastern Germany. The Freital region near the capital of Saxony is a densely populated and intensively used area (by industries and other activities). In this region, a large hard coal deposit was mined from 1542 to 1968. In an area of about 23 km$^2$ the coal seams were mined because they provided an energy source for the quickly established glass and porcelain works and the steel industry, and also a source of domestic fuel. The ashes and slags were used by the residents as additives for building materials.

After World War II, the Soviet–German joint stock company Wismut used the uranium bearing hard coal and black shale for uranium production. Until 1962, these materials, as well as uranium ores from other Saxonian and Thuringian mines, were milled to produce yellow cake. The haulage was abandoned in 1990.

At the start of uranium mining and milling, regulations for radiological protection either did not exist or were not at an advanced standard or were not observed. Thus, mining residues and mill tailings were deposited on waste rock piles and in tailings ponds, dumped carelessly on the ground or used for land fill and other purposes. Slags and other residues were used to cover ground, paths or roads. Buildings and areas that were no longer needed for coal and uranium production were passed on to other enterprises or communities without careful decontamination or restoration. These relics of mining and milling (objects, sites and properties) may cause levels of radiation exposure of the public that are higher than those of natural radiation. In addition, the situation is much more complex because in some areas the natural conditions (e.g. coal seams near the surface) may also influence the average level of radiation exposure.

Careful investigations and a proper radiological assessment of the mining relics had to be carried out in the Freital region. Criteria for radiological assessment of
such situations and rules for investigations have been developed by the Bundesamt für Strahlenschutz (BfS) in the framework of a Federal project [1].

3. CRITERIA FOR RADIOLOGICAL ASSESSMENT

The German Commission on Radiological Protection arrived at the conclusion that the radiation exposure of the public due to former mining and milling is a situation of the ‘intervention’ or ‘pre-existing’ type. Such situations cannot be assessed and handled with the legal tools (e.g. dose limits or dose constraints) used for radiological protection in the case of ‘planned practices’ and, therefore, reference levels had to be established as a prerequisite for decisions concerning the necessity of remedial actions.

The Commission recommended that the radiation dose to man due to the residues of former mining activities and, in addition, due to the natural radiation level should not exceed the reference level of 1 mSv/a (effective dose). This primary reference level has to be interpreted as follows: If the radiation dose to man due to an existing radiation source (relic of mining or milling) does not exceed the primary level, no remedial action is required. In relation to radiation exposure, this relic can be classified as ‘not relevant’ regarding aspects of radiation protection. If the radiation dose to man due to a relic exceeds the reference level, this relic is to be classified as ‘relevant’ and remedial actions should be considered.

In addition to the primary dose level, the Commission recommended measurable quantities (specific radioactivity in soil, local gamma dose rate, volume of disposed materials and dimensions of areas covered by waste materials) to be used as criteria for the release of areas affected by uranium mining to unrestricted or restricted use, and reference levels for concentrations of natural radionuclides in drinking water affected by uranium mining. The exposure to radon in houses is regulated by a special non-intervention level, which is equal to the ‘upper end’ of the normal concentration range (250 Bq/m$^3$). If the measured values are below this level, no measures are necessary.

As a result of long term measurements made by the BfS, the normal geogenic level of outdoor radon concentrations was found to be up to 80 Bq/m$^3$. This value, which represents the ‘upper end’ of the normal outdoor range caused by geological conditions, can be applied as non-intervention level like the non-intervention level for indoor radon. If the radon concentration measured in a residential estate very close to a waste rock pile or other mining relic is below 80 Bq/m$^3$, no remedial actions regarding the inhalation pathway are required. If the radon concentration is above this value, the source related contribution to the outdoor radon concentration should be calculated, taking into account realistic but sufficiently conservative assumptions for exhalation of radon from the relic and for its transport by the
FIG. 1. Schematic diagram of the generic classification procedure.
TABLE I. CRITERIA AND REFERENCE LEVELS RECOMMENDED BY THE GERMAN COMMISSION ON RADIOLOGICAL PROTECTION

Radiation exposure of man due to residues of former mining and milling activities and in addition to the natural radiation level

Reference level for the effective dose: 1 mSv/a

Use of land

Measurable quantities (e.g. specific activity $C_A$ of the soil; dose rate), including the natural radiation level:

$C_A < 0.2$ Bq $^{226}$Ra/g — unrestricted

$0.2$ Bq/g $< C_A < 1$ Bq/g — restricted

$C_A > 1$ Bq $^{226}$Ra/g — site specific investigation

Water affected by uranium ore mining

Reference levels for radionuclide concentrations ($C_A$) in water used for drinking purposes:

<table>
<thead>
<tr>
<th>$U_{nat}$</th>
<th>$^{226}$Ra</th>
<th>$^{210}$Pb</th>
<th>$^{210}$Po</th>
<th>$^{228}$Ra</th>
<th>$^{224}$Ra</th>
</tr>
</thead>
<tbody>
<tr>
<td>$&lt; 7$ Bq/L</td>
<td>$&lt; 0.7$ Bq/L</td>
<td>$&lt; 0.4$ Bq/L</td>
<td>$&lt; 0.6$ Bq/L</td>
<td>$&lt; 0.7$ Bq/L</td>
<td>$&lt; 2$ Bq/L</td>
</tr>
</tbody>
</table>

Radiation exposure to indoor radon

Non-intervention level for indoor radon concentration: 250 Bq/m$^3$

Radiation exposure to outdoor radon

Non-intervention level for outdoor radon concentration in the surroundings of a relic: 80 Bq/m$^3$

atmospheric pathway. If the source related contribution to the outdoor radon level does not exceed 50 Bq/m$^3$, no remedial actions need to be considered (Table I) [2].

The Gesellschaft für Reaktor- und Anlagensicherheit (GRS), in co-operation with BfS, has developed schemes [3] for standardizing the procedure of generic radiological assessment. Figure 1 shows a simplified schematic diagram of such an assessment.

4. PROCEDURE OF INVESTIGATION AND ASSESSMENT

On the basis of the recommendations mentioned above, a multistage procedure, with increasing profundity of investigations, has been developed in order to
register and assess the mining relics with the aim of defining the objects that have to be considered as relevant sources of environmental contamination and radiation exposure of the public. This procedure allows clarification of the situation in a mining region in general and has been specially adapted to the situation of the Freital region.

As a first step, the aerial measurements of the gamma radiation of the ground made in 1982 were evaluated. The available information and the data for the hard coal mining and uranium industry were analysed to identify areas where radiation exposure above the natural level can be expected ('areas of suspicion'). In the Freital region, 72 km² were identified as being suspicious.

All the available reports, data and other information on mining and other relevant activities and sites were evaluated to identify the objects that might be possible sources of public radiation exposure or radioactive contamination of the environment. Because the data and information were old and insufficient for a proper radiological assessment, additional investigations were required.

The co-ordinates, sizes and volumes of objects as well as other relevant information were checked and verified by field inspections. Screening measurements of the local gamma dose rate were included in the verification procedure. The local gamma dose rate was estimated for all objects, as well as for the surrounding grounds not impacted by waste materials. Other information, such as demographic data and projections, land use forecasts and agricultural and industrial data, as well as information about goods and other items of interest that should be protected were also collected [4].

As a result of the verification, 160 mining related objects were identified in the Freital region and were subjected to a first radiological evaluation, applying the criteria already mentioned (Fig. 1); 93 objects were classified as 'not relevant'. For these objects it is not necessary to perform more detailed investigations. Only the remaining objects (67), classified as 'possibly relevant', were subjected to the measuring programme in the next stage. The intention of this measuring programme is to determine:

- The gamma dose rate on and adjacent to the objects in a narrow grid;
- The dimensions of the impacted areas and the depth of radioactive contamination;
- The concentrations of radionuclides of the uranium decay chain in the dumped materials;
- The emissions of radioactivity from objects into the environment, as well as the order of magnitude of such emissions and the resulting radiation concentrations in the environment.

The investigation areas, which amounted to one third (30 km²) of the total area of suspicion, were determined by taking into account the results of the verification and the interests of the affected communities. The investigations were carried
### TABLE II. SCOPE OF INVESTIGATIONS IN THE FREITAL REGION

<table>
<thead>
<tr>
<th>Area of suspicion</th>
<th>72 km²</th>
</tr>
</thead>
<tbody>
<tr>
<td>Investigated area</td>
<td>30 km²</td>
</tr>
<tr>
<td>Number of objects</td>
<td>160</td>
</tr>
<tr>
<td>Number of gamma dose rate measurements</td>
<td>45 500</td>
</tr>
<tr>
<td>Number of ram drills</td>
<td>840</td>
</tr>
<tr>
<td>Number of samples from ram drills</td>
<td>1 200</td>
</tr>
<tr>
<td>Measurement of the soil air radon concentration</td>
<td>180</td>
</tr>
<tr>
<td>Sampling and radionuclide measurements of</td>
<td></td>
</tr>
<tr>
<td>— soil</td>
<td>340</td>
</tr>
<tr>
<td>— water</td>
<td>80</td>
</tr>
<tr>
<td>— sediments</td>
<td>30</td>
</tr>
<tr>
<td>— plants</td>
<td>90</td>
</tr>
<tr>
<td>— building materials</td>
<td>50</td>
</tr>
<tr>
<td>Samples from the sewerage system</td>
<td>10</td>
</tr>
<tr>
<td>Assessment of the groundwater hydraulics</td>
<td>Yes</td>
</tr>
<tr>
<td>Determination of the outdoor radon concentration; number of measuring points</td>
<td>40</td>
</tr>
</tbody>
</table>

out by the BfS or on its behalf by other enterprises. The kind and scope of the investigations were established, taking into account the specific conditions in the investigation areas (Table II). The sites still in possession of the uranium industry (Wismut Company) will be investigated by the Wismut Company itself, taking into account the methods established, by the BfS [5]. On the basis of the results, a final assessment of the mining relics was carried out.

### 5. RESULTS

The investigations have shown that only a small part of the investigated area should be considered as being seriously contaminated. External exposure of people staying on radioactively contaminated ground was found to be the major pathway in the Freital region. All other exposure pathways, including exhalation of radon from the mining relics and inhalation of radon and radon daughter products, can be neglected.

The following types of objects have to be considered as sources of radiation exposure of the public:

— Sites of abandoned uranium mills that had not been decontaminated but had been used for other industrial purposes;
— Areas open to the public where mining residues and mill tailings have been dumped carelessly;
— Areas, paths or roads covered by coal ashes or slags;
— Waste rock piles, especially deposited slags and other burning residues.

In the present situation, uranium mill tailings ponds do not contribute to the radiation exposure of the public. These sites have been fenced, stabilized and covered, for reasons of mining safety. However, further investigations are needed to check whether the existing covers consisting of water or soil are stable enough to prevent long term leaching and spread of radioactivity.

In the investigations, contamination was found on numerous private properties where ashes from domestic burning had been dumped in the past. Erection of buildings on these contaminated properties or use of such ashes and slags may lead to enhanced indoor concentrations of radon compared with the normal range. Also, in houses on building sites where the coal seams are near the surface, the indoor radon concentration may be enhanced. Therefore, the responsible authority has recommended careful investigation of sites before commencement of building, as well as consideration of preventive measures if required.

6. SUMMARY

The mining relics in the Freital region have been registered, investigated and assessed, using a multistage procedure and standardized criteria for radiological assessment.

The 'radiologically relevant' objects, sites and properties have been identified and should be subjected to remedial actions, taking into account aspects of radiological protection and other health and environmental protection, as well as the interest of communities, costs, social aspects, and technical and financial resources.

The results of the investigations have been reported to the competent authorities of Saxony because of their responsibility regarding remedial actions.

REFERENCES


USE OF CLINOPTILOLITE AMENDMENT AS A COUNTERMEASURE FOR CONTAMINATED SOILS

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Abstract

USE OF CLINOPTILOLITE AMENDMENT AS A COUNTERMEASURE FOR CONTAMINATED SOILS.

Widespread contamination with radiocaesium and radiostrontium of large areas in Ukraine, Belarus and Russia occurred after the Chernobyl accident. The intervention potential of man to reduce food contamination and consequently to decrease the radiation dose to the population basically consists of trying to reduce the radionuclide levels in the soil solution by specific soil treatments or amendments. Laboratory tests, using freshwater sediments (for which the sorption mechanisms are similar to those of soils) and a zeolite material (clinoptilolite) as adsorbent, have been carried out in order to provide reliable tools for predicting the amendment effectiveness and the required dose for a given scenario. Results for radiostrontium and radiocaesium partitioning between sediments and clinoptilolite, and values of the distribution coefficient $K_d$ for adsorption and desorption for sediments and clinoptilolite have been obtained, using two protocols. These protocols differ merely in the sequence of the addition of reagents, the entire procedure being a common final state experiment. The results show that for radiostrontium the desorption yields from sediments are consistent with complete reversibility, the cation exchange capacity values and the Sr/Ca selectivity behaviour in both systems. On the other hand, for radiocaesium, significant fractions are irreversibly retained in sediments. It is concluded that in the area of terrestrial radioecology the efficiency of the use of zeolites in soil amendments can be predicted on the basis of measurements of radionuclide $K_d$ values for soil and adsorbent in a solution of a composition identical with that of the ‘in situ’ soil solution.

1. INTRODUCTION

After the Chernobyl accident and the widespread contamination of large areas in Ukraine, Belarus and Russia, a variety of countermeasures have been taken to reduce food contamination, thus lowering the radiation dose to the population.
Among the various measures taken, applications of lime and mineral fertilizers (mostly potassium) have been most frequent [1–5]. For assessing the efficiency of such measures the effect on the transfer factor (ratio of the radionuclide levels in crop and soil) has been used. In the early stages, it was found that these countermeasures lead to a reduction in the transfer factors for radioceasium by a factor of about two to four [1], but now the efficiency of these measures has decreased considerably. These procedures, the effect of which has been known since the 1960s [6], appear to operate essentially at the plant physiological level through some ion competitive interaction process.

In the recent past, emphasis in countermeasure strategy has shifted and has been directed at the chemical level of the soil. In particular, attempts are currently made to reduce the level of radionuclides ($^{137}$Cs, $^{90}$Sr) in the soil solution by applying adsorbents characterized by high radionuclide sorption properties, thus lowering the levels of radioactivity in the roots and resulting in reduced uptake [4]. The procedure currently applied for assessing the efficiency of such measures is again based on their (dose) effect, quantified in terms of transfer factors for the crops. These measures are, however, rarely backed up by quantitative chemical tests of the soil in the laboratory, and the criteria used for choosing adsorbents are based on very general properties, such as cation exchange capacity (CEC), organic matter content and specific sorption properties.

Zeolites are crystalline hydrated aluminosilicates of alkali and alkaline earth cations with infinite three-dimensional crystal structures whose cation exchange property is utilized in a variety of processes, such as decontamination of radioactive wastes, water purification and detergent formation [7]. These materials also have a considerable potential as selective caesium binding agents when they are used in the initial stage and in limited soil layers after caesium fallout [8]. Clinoptilolite is one of the most frequently used zeolite materials because of its efficiency in reducing $^{90}$Sr and $^{137}$Cs uptake by plants [9], with its effect depending on the soil type [10]. The wide application of clinoptilolite is also related to its occurrence in large quantities; it can be excavated at low cost in quarries, all over the world [11].

The objective of this study is to develop chemical tests of soils that will provide reliable tools for predicting the effectiveness of clinoptilolite in soil amendment activities and the required dose for a given scenario. Laboratory tests have been performed with freshwater sediments and clinoptilolite, which is used as adsorbent material because of its high selectivity for caesium and strontium ions. Such tests should evidently be based on relevant characterization studies of the sediments and the adsorbent (clinoptilolite) in terms of competitive sorption potentials for radioceasium and radiostrontium [12]. Since the mechanisms of radiostrontium and radioceasium sorption in freshwater sediments are identical with those in soils, the conclusions and experimental protocols of this study are also applicable in the area of terrestrial radioecology.
2. PARTITIONING OF $^{85}$Sr BETWEEN SEDIMENTS AND CLINOPTILOLITE

2.1. Materials and methods

Partitioning of $^{85}$Sr between clinoptilolite (CP) (reference material from Hector, California, United States of America) and the sediments T2 (Tejo river, Portugal), A (Tejo estuary, Portugal) and KR-4 (Kiev reservoir, Ukraine) has been studied according to the following procedure:

Sediment samples (1 g, enclosed in a dialyse membrane with 5 mL 0.005N CaCl$_2$) and clinoptilolite (1 g, also in a membrane with the same solution) were equilibrated with 200 mL of 0.005N CaCl$_2$ solution for 4 days (one-time equilibration). This procedure leaves both systems essentially in the homoionic bivalent ion form because of the ionic composition of the sediments (Table I) and the 2.4-fold calcium excess with respect to the capacity of clinoptilolite (essentially in the sodium form). Consequently, the level of calcium in the equilibrium solution should be about 0.003N (Ca-CEC for clinoptilolite is 46.7 meq/100 g).

After equilibration, two protocols were followed.

2.1.1. Protocol (a)

The clinoptilolite sample was set aside (to be used in a desorption test) and the sediment was contaminated with $^{85}$Sr by equilibration with 50 mL 0.005N CaCl$_2$ solution labelled with $^{85}$Sr (2 days, end-over-end shaking). The values of the distribution coefficient $K_d$ were obtained by monitoring the activities of the equilibrium solution and the sediment. Clinoptilolite (in the membrane) was then reintroduced and the systems were equilibrated (end-over-end shaking). Countings of solution, sediment and clinoptilolite were made after an equilibration time of

<table>
<thead>
<tr>
<th>Sediments</th>
<th>CEC</th>
<th>Na</th>
<th>K</th>
<th>Ca</th>
<th>Mg</th>
</tr>
</thead>
<tbody>
<tr>
<td>T2</td>
<td>7.5</td>
<td>0.1</td>
<td>0.3</td>
<td>4.4</td>
<td>1.9</td>
</tr>
<tr>
<td>A</td>
<td>27.9</td>
<td>0.3</td>
<td>1.7</td>
<td>56.9</td>
<td>9.1</td>
</tr>
<tr>
<td>KR-4</td>
<td>45.0</td>
<td>0.3</td>
<td>0.9</td>
<td>31.2</td>
<td>6.7</td>
</tr>
</tbody>
</table>
10 days. With these measurements it is possible to obtain the partitioning of $^{85}$Sr between sediment and clinoptilolite, the $K_d$ (adsorption) values for the sediment and clinoptilolite (in the desorption process from the sediment) and the $K_d$ (desorption) value for the sediment.

2.1.2. Protocol (b)

This protocol is exactly identical with protocol (a), except that the sediment sample was first set aside and $^{85}$Sr was first introduced into clinoptilolite. This procedure generates $K_d$ (ads.) values for clinoptilolite and sediment (in the desorption process from clinoptilolite), and $K_d$ (des.) values for clinoptilolite. Protocols (a) and (b) differ merely in the sequence of addition of reagents, and the entire procedure is a common final state experiment.

2.2. Results and discussion

Table II shows the results obtained for the two protocols, with an equilibration time of 10 days, in terms of the ratio of $^{85}$Sr activities in clinoptilolite and sediments, and the various $K_d$ values as defined above.

**TABLE II. RADIOSTRONTIUM PARTITIONING BETWEEN CLINOPTILOLITE AND SEDIMENTS (10 DAYS EQUILIBRATION TIME)**

<table>
<thead>
<tr>
<th>Sediments</th>
<th>$^{85}$Sr in CP/$^{85}$Sr in sed.</th>
<th>$K_d$ sed. (ads.)</th>
<th>$K_d$ CP (ads.)</th>
<th>$K_d$ sed. (des.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>T2</td>
<td>87.7 ± 4.5</td>
<td>51 ± 2</td>
<td>6020 ± 338</td>
<td>69 ± 1</td>
</tr>
<tr>
<td>A</td>
<td>40.4 ± 0.2</td>
<td>101 ± 2</td>
<td>4676 ± 592</td>
<td>116 ± 14</td>
</tr>
<tr>
<td>KR-4</td>
<td>30.4 ± 1.1</td>
<td>137 ± 5</td>
<td>4207 ± 442</td>
<td>138 ± 10</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sediments</th>
<th>$^{85}$Sr in CP/$^{85}$Sr in sed.</th>
<th>$K_d$ CP (ads.)</th>
<th>$K_d$ sed. (ads.)</th>
<th>$K_d$ CP (des.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>T2</td>
<td>133.3</td>
<td>3423 ± 81</td>
<td>37</td>
<td>4860</td>
</tr>
<tr>
<td>A</td>
<td>46.5 ± 0.2</td>
<td>2527 ± 38</td>
<td>101 ± 7</td>
<td>4715 ± 340</td>
</tr>
<tr>
<td>KR-4</td>
<td>35.1 ± 1.7</td>
<td>2464 ± 153</td>
<td>126 ± 10</td>
<td>4406 ± 133</td>
</tr>
</tbody>
</table>

* The $K_d$ values (average and standard deviation) are given in mL/g.
It is seen that for both protocols the $K_d$ (ads.) and $K_d$ (des.) values vary in a rather narrow range, i.e. the system appears to be reasonably reversible (direct evidence of this is provided by the values of the ‘partition coefficient’ of $^{85}$Sr between clinoptilolite and sediment in both protocols). Consequently, the systems appear to be at equilibrium. Most important, however, is the partitioning of radiostrontium between sediment and clinoptilolite. It is clearly seen that the efficiency of clinoptilolite in removing radiostrontium from the sediments improves at lower sediment CEC values (Table I), as had been expected. In fact, the ratios of the radiostrontium levels in both systems should be identical with the ratios of the sorption $K_d$ values. This appears to be the case. In order to rationalize the partitioning behaviour of radiostrontium in terms of the CEC values of both components, a higher (factor of about five to ten) Sr/Ca selectivity coefficient for clinoptilolite than for sediments [12] will be taken into account. In these terms, the ratio of radiostrontium in zeolite and sediment can be expected to be given by the ratio of the CEC values times the ratio of the Sr/Ca selectivity coefficients in the two systems. From such an analysis, it appears that the radiostrontium partitioning data are consistent with a Sr/Ca selectivity coefficient for zeolite exceeding the value for sediments by a factor of 15 to 30.

It is also of interest to rationalize the radiostrontium $K_d$ (ads.) values of sediments on the basis of CEC and the calcium concentration in the equilibrium solution. Using the ratio of CEC (Table I) and the calcium concentration in the equilibrium solution (0.003N), i.e. $K_d$ Ca, the values of 25 (T2), 93 (A) and 150 (KR-4) mL/g were obtained. This means that radiostrontium sorption is consistent (on the basis of $K_d$ Sr values) (Table II) with a $K_d$ Sr/Ca value of about one to two in sediments.

It is useful to provide a predictive tool for estimating the potential beneficial effects of zeolite amendments as a countermeasure for $^{90}$Sr contaminated soils. On the basis of the evidence presented, the partitioning of radiostrontium between the exchange complex of zeolite and that of soil can be written as:

$$\frac{N_{Sr}(z)}{N_{Sr}(s)} = \frac{(CEC)_z}{(CEC)_s} \frac{m_z}{m_s} \frac{p}{1}$$

where $N_{Sr}$ refers to the radiostrontium fractions in zeolite and soil, $m$ refers to the masses of zeolite and soil, and $p$ represents the ratio of the Sr/Ca selectivity coefficients in zeolite and soil for the prevailing ionic scenario. Since $N_{Sr}(z) = 1 - N_{Sr}(s)$, Eq. (1) can be put in the following form:

$$\frac{1}{N_{Sr}(s)} = 1 + \frac{(CEC)_z m_z}{(CEC)_s m_s} \frac{p}{1}$$
The values of CEC and p can be measured and the term \((CEC)_z p/(CEC)_s\) represents the ratio of the \(K_d\) values for radiostrontium in zeolite and soil. Evidently, when applying a given dose of zeolite to a soil, the exchange complex of zeolite will adjust to an ionic configuration in equilibrium with that of the soil. Consequently, for making efficiency predictions it is necessary to measure \(K_d\) values of radiostrontium for soil and zeolite in a solution with a composition identical with that of the soil solution.

In conclusion, it is of interest to make some predictions of efficiency on the basis of Eq. (2). For a soil of low CEC (say, 5 meq/100 g) and a zeolite of an (effective) CEC of 50 meq/100 g, a dose of 1% zeolite (or 40 t/ha) should lead to a decrease of the radiostrontium loading in the soil (and therefore of the radiostrontium level in the soil solution) by a factor of two (assuming a ten times higher value for the Sr/Ca selectivity coefficient in the soil). If, however, the same dose were applied to a peat soil (CEC = 100 meq/100 g), then the decrease of the radiostrontium level in the soil would be 5%.

3. PARTITIONING OF \(^{137}\text{Cs}\) BETWEEN SEDIMENTS AND CLINOPTILOLITE

3.1. Material and methods

Exactly the same common final state procedure was used as that for \(^{85}\text{Sr}\) described in Section 2.1, except that the composition of the liquid phase used for pre-equilibration of sediment and clinoptilolite was 5 meq/L \(\text{CaCl}_2\) and 0.1 meq/L \(\text{KCl}\).

3.2. Results and discussion

Table III shows the results obtained for protocol (a) (sediment first labelled) and protocol (b) (clinoptilolite first labelled) and for equilibration times of 3 and 9 days. It is apparent that the values of the partition coefficient of radiocaesium, expressed in terms of the ratios of \(^{137}\text{Cs}\) in clinoptilolite and sediment, are very different, depending on the direction in which the equilibrium is being approached. For 3 days equilibration, the ratios differ by two orders of magnitude; after 9 days, the difference reduces to a factor of about 10 to 20. Evidently, this phenomenon is due to the fact that considerable fractions of radiocaesium are irreversibly retained and are therefore not involved in the repartition process between the two systems. It is of interest to attempt to quantify this fixation effect on the desorption of radiocaesium from sediments.

If the processes are reversible for both sediment and clinoptilolite, then the ratios of the \(^{137}\text{Cs}\) contents should be identical with the \(^{137}\text{Cs}\) distribution coefficients obtained in the adsorption steps for clinoptilolite (protocol (b)) and sediment.
TABLE III. RADIOCAESIUM PARTITIONING BETWEEN SEDIMENTS AND CLINOPTILOLITE FOR EQUILIBRATION TIMES OF 3 AND 9 DAYS

<table>
<thead>
<tr>
<th>Sediments</th>
<th>Protocol (a)</th>
<th>Protocol (b)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{137}\text{Cs in CP}$</td>
<td>$^{137}\text{Cs in CP}$</td>
</tr>
<tr>
<td></td>
<td>$^{137}\text{Cs in sed.}$</td>
<td>$^{137}\text{Cs in sed.}$</td>
</tr>
<tr>
<td>$K_d$ sed. (ads.)</td>
<td>$K_d$ CP (ads.)</td>
<td>$K_d$ CP (ads.)</td>
</tr>
<tr>
<td><strong>Three days</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>T2</td>
<td>0.31 ± 0.01</td>
<td>5962 ± 529</td>
</tr>
<tr>
<td>A</td>
<td>0.10 ± 0.01</td>
<td>17491 ± 659</td>
</tr>
<tr>
<td>KR-4</td>
<td>0.11 ± 0.01</td>
<td>13556 ± 944</td>
</tr>
<tr>
<td><strong>Nine days</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>T2</td>
<td>0.84</td>
<td>5962 ± 529</td>
</tr>
<tr>
<td>A</td>
<td>0.28 ± 0.01</td>
<td>17491 ± 659</td>
</tr>
<tr>
<td>KR-4</td>
<td>0.32 ± 0.02</td>
<td>13556 ± 944</td>
</tr>
</tbody>
</table>

* The $K_d$ values (average and standard deviation) are given in mL/g.

(protocal (a)) (results presented in Table III). It is seen that, for the three systems studied, the $K_d$ CP values exceed the sediment values and the ratios are:

$$
\frac{K_d \text{ CP(ads.)}}{K_d \text{ sed. (ads.)}} = 3.4 \text{ (T2), 1.6 (A), 1.8 (KR-4)}
$$

These values are three to five times higher than those obtained in protocol (a) and three to five times lower than those for protocol (b) (9 days equilibration), i.e. sorption irreversibility occurs from both sides. The experimental $^{137}\text{Cs CP}$/$^{137}\text{Cs sed.}$ ratio can be represented by the symbol $p$, and the ratio of $K_d$ CP/$K_d$ sed. by the symbol $q$. The total $^{137}\text{Cs}$ activity in the system (after equilibration) can be represented by the sum: $a_{\text{CP}} + (a_{\text{CP}}/p)$. The quantity of mobile (reversible) radiocaesium in the system is given by the term $a_{\text{CP}} + (a_{\text{CP}}/q)$, where $(a_{\text{CP}}/q)$ refers to the residual quantity of mobile radiocaesium in sediment (participating in partitioning). The quantity of fixed radiocaesium is given by the difference between these two terms. The fraction of radiocaesium fixed is, of course, given by the ratio of this difference to the total amount present in the system (at the start of the desorption
experiment, in the sediment). Consequently, the fraction of radiocaesium fixed can be represented by:

\[
\frac{1 - \frac{1}{p}}{1 + \frac{1}{p}} = \frac{q - p}{q(p + 1)}
\]

Applying such calculation to the data shown in Table III, fixation levels for sediments of 41% (T2), 64% (A) and 62% (KR-4) were found. These numbers are in good agreement with the results obtained for short ageing times by the infinite bath technique: 50-55% (K and NH₄) for T2, 40-35% (K and NH₄) for A and 70% (NH₄) for KR-4 sediments [12].

Consequently, the desorption of radiocaesium from sediments is consistent with the ratios of the \(K_d\) (ads.) values for the two systems, but the process is restricted to the 'mobile' fraction. The efficiency of radiocaesium desorption in the systems studied is, of course, rather low and the use of zeolites as a countermeasure against \(^{137}\text{Cs}\) contamination is most likely limited to soils with a very low specific radiocaesium interception potential (podzols, peat soils).

The laboratory tests necessary for making predictions of the efficiency of a particular zeolite (or other adsorbent) are the same as those described for radiostontium: measurement of \(K_d\) values for soil and adsorbent in a 'representative' soil solution. If this ratio were of the order of \(10^2\), then a dose of 40 t zeolite/ha would reduce the level of (still available) radiocaesium in the soil by a factor of two.

4. CONCLUSIONS

The efficiency of the use of zeolite materials in soil amendments for curbing the chemical availability of radiocaesium and radiostontium in the soil can be predicted on the basis of a simple test: a measurement of the \(K_d\) values for both soil and zeolite in a solution of a composition identical with that of the 'in situ' soil solution. In the case of radiostontium, the desorption yields from sediments are consistent with complete reversibility, the CEC values and the Sr/Ca selectivity behaviour in both systems. In the case of radiocaesium, significant fractions are irreversibly retained in sediments and, accordingly, the desorption yields are lower. However, the fractions being desorbed (from the available soil pool) are consistent with the fixation levels ascertained on the basis of the infinite bath method. For radiostontium, 'practical' results can be expected only in cases of soils of very low CEC and by using zeolite materials of high capacity and high radiostontium selectivity. For radiocaesium, successful application of such amendments will be limited to soils of very low radiocaesium interception potential.
REFERENCES


EFFECTS OF AMELIORATIVE TREATMENTS APPLIED TO RADIOCAESIUM CONTAMINATED UPLAND VEGETATION IN THE UNITED KINGDOM

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Abstract

EFFECTS OF AMELIORATIVE TREATMENTS APPLIED TO RADIOCAESIUM CONTAMINATED UPLAND VEGETATION IN THE UNITED KINGDOM.

Relatively high levels of $^{137}$Cs have persisted in some upland areas of the United Kingdom following the Chernobyl accident. The $^{137}$Cs deposition in the British Isles was related to rainfall patterns, and deposition was highest in areas of heavy rainfall, including Cumbria and north Wales. Because of the peat soils, low nutrient status and semi-natural vegetation of these upland areas, the radiocaesium deposited has remained available for plant uptake and transfer to livestock. The radiocaesium concentration in lambs grazing this contaminated vegetation continues to exceed the statutory level of 1000 Bq/kg live weight, and restrictions on the movement, sale and slaughter of livestock remain in force. The ability of soil treatments to reduce the radiocaesium concentration in vegetation via immobilization in soil and/or competition of caesium and potassium for plant uptake was investigated. Field plots in north Wales and Cumbria were treated with potassium, ammonium ferric hexacyanoferrate (AFCF), bentonite and clinoptilolite in 1990, and the radiocaesium concentrations of vegetation and soil were monitored for three years following application. The $^{137}$Cs concentration of vegetation on both treated plots and control plots exhibited a strong seasonal variation, with the peak concentration being in early summer. The variation in $^{137}$Cs concentration between different plant species was large, and the response to treatments also varied between plant species. The potassium treatment produced reductions in the radiocaesium concentration of the natural vegetation, especially in the initial years after treatment. Clinoptilolite and AFCF treatments also produced reductions in radiocaesium concentration, with some plant species showing greater responses to the various treatments. Potassium, clinoptilolite and AFCF have shown some potential for use as soil ameliorative treatments for $^{137}$Cs reduction on upland peat soils with grazed semi-natural vegetation.
1. INTRODUCTION

In peat soils characteristic of large upland areas of the United Kingdom, $^{137}$Cs deposited following the Chernobyl accident has remained relatively available to plants, giving rise to contamination of grazing livestock. The use of soil amendments to immobilize $^{137}$Cs in soil and/or reduce plant uptake was investigated. Field plots in contaminated upland areas of the British Isles were treated with potassium, ammonium ferric hexacyanoferrate (AFCF), bentonite and clinoptilolite. Caesium is known to compete with potassium for plant uptake in these potassium deficient soils [1]. AFCF, bentonite and clinoptilolite sorb caesium to various degrees and may reduce the available caesium in soil solution [2]. It was thought that the effect of these chemicals on the organic upland soils of the UK could sufficiently reduce the radiocaesium uptake of the semi-natural vegetation to prevent livestock from accumulating radiocaesium concentrations above the restriction level of 1000 Bq/kg. This field trial was designed to assess the potential of various treatment chemicals in reducing radiocaesium uptake of native vegetation following simple addition to the surface.

2. EXPERIMENTAL DESIGN

Two field sites were chosen in areas where movement of sheep with a radiocaesium concentration of $>1000$ Bq/kg live weight remains under restriction. The first site was at Llyn Eigiau in north Wales, comprising blanket bog with Erica tetralix L. and Scirpus caespitosus L. co-dominant on raised peat hags, and other species present in the surrounding peat bogs. The second site was at Corney Fell, Cumbria, with blanket bog dominated by Eriophorum vaginatum L. on the raised hags. The plot design was based on zones of dominant vegetation on the raised peat hags because of the heterogeneous nature of these sites.

The treatments investigated were: control, potassium chloride (16.7 g/m$^2$), AFCF (0.3, 1.0 and 2.0 g/m$^2$), bentonite (500 and 1000 g/m$^2$) and clinoptilolite (500 and 1000 g/m$^2$). Each chemical treatment was applied singly to three replicate plots, each 6 m $\times$ 3 m, positioned at least 1 m apart (27 plots at each site). The chemicals were applied to the plots directly by hand and watered in, with AFCF being watered in as a colloidal suspension in 12 L of nearby stream water. The treatment rates were chosen to be equivalent to normal agricultural application rates or, in the case of AFCF, rates which were estimated from the literature to be effective in administration to livestock.

The treatments were applied to the plots in October 1990. The plots were sampled prior to treatment and then on twelve occasions over the next three years, collecting bulk vegetation and individual plant species on each occasion and soil samples each October. A different quadratic plot (1 m$^2$) was sampled for bulk
vegetation from each plot on each occasion, thus providing a sample of the standing vegetation at a given time. Sampled areas were not resampled for bulk vegetation during the project. Individual species were collected from across the treated plots and bulked to form a single sample on each occasion. The collected samples were dried, ground and analysed for $^{137}$Cs using a Ge–Li gamma spectrometer. Total potassium analysis was also conducted on many samples.

3. RESULTS

The average soil $^{137}$Cs concentrations for all 27 plots at each depth sampled at both sites for four consecutive October samples is shown in Fig. 1. (In 1993, the 0–2.5 cm and 2.5–5 cm layers were combined to provide an average value for a 0–5 cm layer.) There is a greater $^{137}$Cs inventory at Corney Fell than at Llyn Eigiau, but the distribution down the soil profile is similar at both sites. At Corney Fell the variation between years was not significant and is thought to be due to the spatial variability of the deposit. These results confirm that the majority of the $^{137}$Cs inventory is present in the root mat and upper soil layers, indicating minimal migration of $^{137}$Cs down the soil profile since deposition.

Soil samples from control plots and potassium treated plots were analysed for potassium. The potassium treatment increased the soil potassium concentrations slightly in the lower soil layers for two years after treatment, confirming that added potassium is not washed out of the system within 1 year.

The $^{137}$Cs concentration in the vegetation shows strong seasonal fluctuations; the peak concentrations in early summer coincide with the start of the grazing period on these uplands. Concentrations in the bulk vegetation during summer reach around 1000 Bq/kg and are rather higher in Cumbria than in north Wales owing to the greater soil inventory, but show the same pattern (Fig. 2). All treatments have reduced the $^{137}$Cs concentrations in bulk vegetation to some extent, with bentonite the least effective. The results are averages of triplicates, and statistical analysis (not provided here) has shown that a $^{137}$Cs concentration of $-60\%$ of the control value or lower represents a significant reduction (95% confidence limit).

Figures 3–6 give the $^{137}$Cs transfer coefficients (Bq/kg plant dry weight per Bq/m$^2$ soil and root mat dry weight to 10 cm depth) relative to control for the June and October vegetation samples. This enables the reduction with treatment at each sampling compared with the control to be seen clearly on a deposit basis. Values of around 0.6 thus represent a 60% reduction on control and are statistically significant in the majority of cases.

At Llyn Eigiau the effectiveness of potassium treatment in reducing $^{137}$Cs transfers to bulk vegetation is clear (Fig. 3) and has persisted until the third year after treatment. Treatments with AFCF, bentonite and clinoptilolite have also produced
reductions, many of which are significant. The effectiveness of clinoptilolite treatment appears to be continuing to improve, especially in the October samples. At Corney Fell the effectiveness of treatment is slightly lower (~50% reductions); the effectiveness of potassium treatment is still clear, though diminishing in the third year. AFCF treatment has consistently produced slight transfer reductions; bentonite has not been effective and clinoptilolite again appears to be improving its effectiveness through three years (Fig 4). A further sampling of the plots is planned for 1995, to investigate the longer term effectiveness of the treatments.
FIG. 2. Caesium-137 concentration in bulk vegetation samples from Llyn Eigiau.
Botanical composition is an important factor influencing $^{137}$Cs uptake by vegetation. The response to treatment was greater at Llyn Eigiau than at Corney Fell because of differences in plant species response to treatment and different botanical composition. At Llyn Eigiau, Erica species co-dominate with a tussocky grass, Scirpus caespitosus. Erica showed reductions of up to 30% of the control values with potassium treatment, initial reductions with AFCF and reductions of up to $\sim 50\%$ with bentonite and clinoptilolite (Fig. 5). Scirpus caespitosus showed a lower response to treatments, but potassium and clinoptilolite especially produced more than 50% reduction on control (Fig. 6). The dominant vegetation at Corney Fell, Eriophorum vaginatum, was more resistant to treatment generally and, although some of the minor species responded strongly to the treatments, the response of E. vaginatum dominated the response of the bulk vegetation (Fig. 4).

Potassium analysis of vegetation samples indicated that none of the treatments increased the potassium content of the vegetation. A slight increase in dry matter production (bulk vegetation) with potassium treatments was noted at Llyn Eigiau after 1 year. This did not continue in subsequent years and was not evident at Corney Fell.
In deciduous graminaceous species (S. caespitosus, E. vaginatum), which typically grow in blanket bogs, nutrients are remobilized from leaves prior to senescence in autumn and are translocated to overwintering basal tissues. The stored nutrients are translocated back to shoots when new growth resumes in spring [3, 4]. It is possible that $^{137}$Cs may be translocated and cycled similarly. In contrast, shrub-like species such as Erica tetralix depend on nutrient uptake from the soil rather than stored reserves [5] and this may explain the greater response to soil treatments, particularly potassium, noted in Erica species.

The response to AFCF treatment was variable and the effect appears to have declined after the first year. Information on interactions between the colloidal AFCF and soil–plant systems is lacking, and the potential mobility of the Cs–FCF complex is not fully understood, though the complex is known to be reasonably stable.

Bentonite addition produced inconsistent reductions in $^{137}$Cs transfer to vegetation, probably because of the low specificity of bentonite for caesium sorption. Other researchers have been relatively unsuccessful in using clay minerals to reduce soil–plant transfer of $^{137}$Cs [2, 6, 7].
Although initially the effect of clinoptilolite was not as marked as that of potassium, its effectiveness has surpassed that of potassium after three years in some cases. Because of its specificity, immobilization of $^{137}$Cs by clinoptilolite is relatively irreversible and long term. The difficulty in incorporation of clinoptilolite into the dense root mat of this vegetation would explain the delay in the realization of its effect. Previous work [8] found that mixing clinoptilolite with soil, rather than surface spreading, reduced the uptake of $^{137}$Cs by crops.

4. CONCLUSIONS

Significant reductions in $^{137}$Cs concentrations in upland vegetation have been achieved with application of treatment chemicals, notably potassium, in the early years after treatment, and clinoptilolite increasingly as it becomes incorporated into the soil. The variability in response to treatments is primarily due to differences in the response of various plant species and the heterogeneous botanical composition at the sites.
Chemical applications have potential for reducing $^{137}$Cs concentration in vegetation on upland peat soils of low nutrient status, dependent on the economics and applicability of field treatment. The reductions in general $^{137}$Cs concentrations achieved in this trial could be sufficient to reduce or remove the problems of livestock contamination on these open grazing lands.

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REFERENCES


ALTERNATIVE AGRICULTURE AS A SUBSTITUTE FOR ENVIRONMENTAL REMEDIATION

Production of poultry in radiologically contaminated areas

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Aiken, South Carolina,
United States of America

Abstract

Exploiting the physiological and ecological characteristics of domesticated species has seldom been considered as a means of returning radiologically contaminated areas to safe agricultural production. However, the proper choice of cultivated and domesticated species, together with appropriate husbandry practices, may allow safe production of foods, fibre and energy. As an example, factors that could permit safe production of food products for human consumption from poultry raised in contaminated areas are considered. These factors include radionuclide transfer from the environment into poultry and methods for reducing radionuclide uptake and/or decontaminating chickens to yield acceptable food products. Studies of growth and feed intake rates of chickens under intensive management and free-ranging husbandries, 

$^{137}$Cs uptake by chickens exposed to contaminated sediments, potential effects of husbandry on $^{137}$Cs concentrations and $^{137}$Cs elimination by chickens after removal of contaminated feed are described. Data from these and other studies are combined in simulation models of the $^{137}$Cs kinetics of chickens. Chicken product $^{137}$Cs concentrations ($[^{137}Cs]_s$) decrease with increases in body mass, apparently as a result of decreasing mass specific intake rates. Husbandries that increase contaminant intake (e.g. free-ranging rearing conditions versus brooder house production) or access to contaminated soils (e.g. scattering feed directly onto contaminated soils versus the use of feeders) increase total body $[^{137}Cs]_s$. However, model simulations indicate that it is possible to produce safe poultry products (especially eggs) at all but unrealistically high feed contamination levels. Even when dietary $^{137}$Cs levels produce $[^{137}Cs]_s$ in poultry products that exceed acceptable limits, the high metabolism of poultry leads to a rapid decrease in $[^{137}Cs]_s$ once the chickens are placed on uncontaminated rations. This permits the use of contaminated feed or forage for most of the growing period of chickens.
1. INTRODUCTION

The Chernobyl nuclear accident demonstrated the potential for damage from large scale radionuclide releases [1-4]. This accident alone resulted in the exclusion of over 3000 km$^2$ of land from public access, and tens of thousands of square kilometres remain severely contaminated [4]. Subsistence agriculture in such areas leads to significant radionuclide accumulation by free-ranging livestock — an important pathway for human exposure [5-8].

The cost of restoration of contaminated areas worldwide is expected to exceed US $10^{10}$-$10^{12}$ and remediation is unlikely to be completely successful. Decontamination technologies may also adversely affect soil integrity and compromise the sustainable productivity of existing agroecosystems. We consider here the alternative of exploiting the physiological and ecological characteristics of appropriate domesticated species to allow production of safe products for consumers, promote removal of radionuclides from the soil and concentrate radionuclides at central processing facilities. This could allow agricultural production to continue while radionuclides are removed or isolated by physical and ecological processes.

Although the forage-cow-milk pathway has been the most studied (e.g. [9-11]) and transfer coefficients/factors of radionuclides from forage to poultry products have been estimated (e.g. [11-17]), no studies have yet considered keeping poultry as an alternative to keeping other livestock in contaminated areas, or how radionuclide concentrations of such products might be manipulated by husbandry practices and post-processing. Poultry have greater feed conversion efficiencies (typically 20% [16]) than cattle and exhibit less efficient contaminant transfer to non-meat food products [11]. Their high metabolic rates also permit radionuclides to be eliminated during relatively short ‘decontamination periods’ while they are fed uncontaminated rations prior to human consumption, thus permitting use of contaminated feed for most of the growing period.

We summarize here factors that determine whether poultry products can be safely used for human consumption and suggest means of minimizing human exposure. These include: (1) factors determining the characteristics of radionuclide transfer from the environment into poultry, (2) intrinsic and extrinsic variables influencing radionuclide concentrations, and (3) means by which radionuclide uptake can be minimized and/or chickens can be decontaminated to yield acceptable food concentrations. We describe studies of: (1) growth and feed intake rates of chickens under intensive management and free-ranging husbandries, (2) uptake of $^{137}$Cs by chickens free-ranging on contaminated sediments, (3) potential effects of husbandry practices on $^{137}$Cs concentrations, and (4) elimination of $^{137}$Cs by chickens after removal of contaminated feed. We combine these and other data into conceptual and simulation models of the $^{137}$Cs kinetics of chickens that permit evaluation of the possibility of producing safe food products.
2. METHODS

The parameters used in modelling the $^{137}$Cs kinetics of chickens are presented in Fig. 1 and Table I. Additional information on these parameters is provided below. All masses are given in either wet or live values unless otherwise indicated.

**FIG. 1.** Conceptual models of the $^{137}$Cs kinetics of chickens. The $^{137}$Cs is of breast meat, leg meat and eggs ($C_{\text{breast}}$, $C_{\text{leg}}$ and $C_{\text{egg}}$, Bq) are increased by ingestion of feed ($R_f$, kg feed·chicken$^{-1}$·d$^{-1}$), with different simulated $^{137}$Cs is ($C_I$, Bq/kg). Assimilation of dietary $^{137}$Cs into breast meat, leg meat and eggs is considered independently ($A_{\text{breast}}$, $A_{\text{leg}}$ and $A_{\text{egg}}$, respectively). The concentrations of $^{137}$Cs decrease by physical decay of the radionuclide ($k_p$) and by biological elimination ($k_{\text{breast}}$, $k_{\text{leg}}$, $k_{\text{egg}}$, d$^{-1}$), all of which are modelled as first-order processes.
## TABLE I. PARAMETERS USED IN MODELLING $^{137}$Cs KINETICS AND DECONTAMINATION TIMES OF CHICKENS

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Description</th>
<th>$\bar{x}$ (SD)</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 $M_0$</td>
<td>Initial mass of chicken (g)</td>
<td>41.8 (1.60)</td>
<td>This study</td>
</tr>
<tr>
<td>7 $M_\infty$</td>
<td>Asymptotic mass of chicken (g)</td>
<td>3498 (49.4)</td>
<td>This study</td>
</tr>
<tr>
<td>3 $T$</td>
<td>Time to reach 90% of $M_\infty$ (d)</td>
<td>239 (3.61)</td>
<td>This study</td>
</tr>
<tr>
<td>4 $m$</td>
<td>Richards shape parameter (dimensionless)</td>
<td>0.535 (0.023)</td>
<td>This study</td>
</tr>
<tr>
<td>5 $a$</td>
<td>Intercept of feed intake equation ((kg \text{ feed/chicken}) \text{ per (kg chicken/d)})</td>
<td>0.072 (0.001)</td>
<td>This study</td>
</tr>
<tr>
<td>6 $b$</td>
<td>Slope of feed intake equation (dimensionless)</td>
<td>0.583 (0.010)</td>
<td>This study</td>
</tr>
<tr>
<td>8 $A_{\text{total}}$</td>
<td>Assimilation fraction to total body (dimensionless)</td>
<td>0.25 (0.05)</td>
<td>(\bar{x}) (SD) of Refs [12, 16, 17]</td>
</tr>
<tr>
<td>9 $A_{\text{breast}}$</td>
<td>Assimilation fraction to breast meat (dimensionless)</td>
<td>0.137 (0.004)</td>
<td>(\bar{x}) (SD) estimated from TCs in Refs [15, 17]</td>
</tr>
<tr>
<td>10 $A_{\text{leg}}$</td>
<td>Assimilation fraction to leg meat (dimensionless)</td>
<td>0.049 (0.007)</td>
<td>(x) (SD) estimated from TCs in Refs [15, 17]</td>
</tr>
<tr>
<td>11 $A_{\text{egg}}$</td>
<td>Assimilation fraction to whole egg (dimensionless)</td>
<td>0.012 (0.005)</td>
<td>(x) (SD) estimated from TCs in Refs [15, 17]</td>
</tr>
<tr>
<td>12 $k_p$</td>
<td>Physical decay constant for $^{137}$Cs (d(^{-1}))</td>
<td>(6.3 \times 10^{-5})</td>
<td>Ref. [20]</td>
</tr>
<tr>
<td>13 $k_{\text{total}}$</td>
<td>Total body biological elimination rate constant (d(^{-1}))</td>
<td>0.027 (0.002)</td>
<td>(\bar{x}) (SD) of Ref. [12] and estimate from Ref. [23]</td>
</tr>
<tr>
<td>14 $k_{\text{breast}}$</td>
<td>Breast meat elimination rate constant (d(^{-1}))</td>
<td>0.038 (0.009)</td>
<td>(\bar{x}) (SD) of Refs [15-17]</td>
</tr>
<tr>
<td>15 $k_{\text{leg}}$</td>
<td>Leg meat elimination rate constant (d(^{-1}))</td>
<td>0.075 (0.017)</td>
<td>(\bar{x}) (SD) of Refs [15, 17]</td>
</tr>
<tr>
<td>16 $k_{\text{egg}}$</td>
<td>Rate constant for decrease in egg $^{137}$Cs(i)s in successive layings (d(^{-1}))</td>
<td>0.223 (0.009)</td>
<td>(\bar{x}) (SD) of Refs [15, 17]</td>
</tr>
</tbody>
</table>

* Means and standard deviations for the parameters used to simulate growth (parameters 1 through 4, Eq. (1)) were estimated from non-linear least squares fits to growth data from a random bred line of domesticated chickens. Parameters for the feed intake rate equation (parameters 5 and 6, Eq. (2)) were estimated by regression analyses for these chickens. Assimilation fractions for meat and eggs were estimated from indicated literature values for transfer coefficients (parameters 7 to 10). Elimination rate coefficients were also obtained or estimated from literature values (parameters 11 to 16). Circled parameter numbers correspond to the conceptual model (Fig. 1).
2.1. Model parameters

2.1.1. Growth model

Growth rates were measured for a breed of random bred chickens [18] raised on commercial feed in heated battery brooders and for feral bantam chickens [19] under battery brooding and free-ranging conditions (Table II). We fitted data to the Richards sigmoidal model [21, 22]:

\[ M_t = \left\{ M_\infty (1-m) \left[ 1 - e^{\frac{-2t(m+1)}{T}} \right] + M_0 (1-m) e^{\frac{-2t(m+1)}{T}} \right\} \frac{1}{1-m} \]  

(1)

where: \( M_t \) is the mass of the chicken at time \( t \), \( M_\infty \) is the asymptotic body mass, \( M_0 \) is the body mass at hatching, \( T \) is the time required to achieve approximately 90% of asymptotic body mass, and \( m \) is a shape parameter that produces a variety of sigmoidal curves with different inflection points [21, 22].

TABLE II. EFFECTS OF SIZE, SEX AND HUSBANDRY CONDITIONS ON GROWTH AND SIMULATED [\(^{137}\)Cs]S OF BANTAM AND RANDOM BREED CHICKENS

<table>
<thead>
<tr>
<th></th>
<th>( M_\infty \bar{x} ) (SD)</th>
<th>([^{137}\text{Cs}] ) (Bq/kg)</th>
<th>Total body</th>
<th>Breast meat</th>
<th>Leg meat</th>
<th>Eggs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bantams</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \varphi ) ( \varphi )</td>
<td></td>
<td>709.2 (12.65)</td>
<td>762</td>
<td>1292</td>
<td>382</td>
<td>141</td>
</tr>
<tr>
<td>Indoor</td>
<td></td>
<td>715.1 (26.65)</td>
<td>759</td>
<td>1287</td>
<td>381</td>
<td>142</td>
</tr>
<tr>
<td>( \sigma ) ( \sigma )</td>
<td></td>
<td>1352 (79.34)</td>
<td>585</td>
<td>382</td>
<td>296</td>
<td>-</td>
</tr>
<tr>
<td>Outdoor</td>
<td></td>
<td>1117 (19.13)</td>
<td>630</td>
<td>381</td>
<td>316</td>
<td>-</td>
</tr>
<tr>
<td>Random breeds</td>
<td></td>
<td>3498 (49.35)</td>
<td>392</td>
<td>666</td>
<td>198</td>
<td>335</td>
</tr>
</tbody>
</table>

A Bantams were reared under brooder house conditions (indoor) or were parent reared under free-ranging conditions (outdoor). The growth of both sexes of bantams under each rearing condition was estimated with Richards sigmoidal models (see Section 2.1.1). The resulting growth parameters (1) to (9), Table I) were then used to describe growth and ingestion rates in simulation models of poultry product \(^{137}\text{Cs} \) kinetics. The predicted equilibrium \([^{137}\text{Cs}] \)s from these simulations are based on an assumed dietary \([^{137}\text{Cs}] \) of 1 kBq/kg feed.
2.1.2. Ingestion model

We combined growth and feed consumption data to estimate size dependent feeding rates \( R_f \), kg feed \( \cdot \) chicken \(^{-1} \cdot \text{d}^{-1} \) as:

\[
R_f = a M^b \text{ (kg of chicken)} \tag{2}
\]

Ingestion rates were then estimated for random breds under penned rearing conditions, substituting \( M_t \) from Eq. (1) for \( M \).

The estimates of growth and ingestion rates were combined with literature values for \(^{137}\text{Cs} \) assimilation fractions and elimination rate constants.

2.1.3. Assimilation fractions

The total body assimilation fraction \( A_{\text{total}} \) was estimated from studies of chronic ingestion of \(^{137}\text{Cs} \) contaminated feed \([16, 17]\) and from two-component elimination of \(^{134}\text{Cs} \) following an acute oral \(^{134}\text{Cs} \) dose \([12]\). In these studies, about 20-30% of ingested radiocaesium was assimilated into the body.

Because of difficulties in measuring assimilation, radionuclide transfer to meat and eggs is generally described using transfer coefficients calculated at radionuclide equilibrium. We indirectly estimated the assimilation fraction to meat \( A_{\text{meat}} \) from meat transfer coefficients \( \text{TC}_{\text{meat}} \) as:

\[
A_{\text{meat}} = \text{TC}_{\text{meat}} \text{ (d/kg)} \times k_{\text{meat}} \text{ (d}^{-1}) \times \text{mass of meat (kg)} \tag{3}
\]

where \( k_{\text{meat}} \) is either the breast meat or the leg meat elimination rate constant (see Section 2.1.4) \([14]\). Published values of \( \text{TC}_{\text{meat}} \) fall into two ranges: about 1.2–1.6 d/kg (e.g. \([15, 17]\)) and 3.0–4.5 d/kg (e.g. \([11, 16]\)), probably reflecting both chemical availability \([17]\) and metabolic activity of the muscle. Because the TCs of breast meat are larger than those of leg meat \([15, 17]\), we simulated uptake and elimination from breast meat and leg meat separately, using the higher TC range for breast meat in the model. We assumed that the mass of meat was 0.41 of the whole body mass \([14, 16]\) and that breast meat and leg meat were 0.56 and 0.34 of the edible meat mass, respectively \([14]\).

We also indirectly estimated the assimilation fraction to eggs \( A_{\text{egg}} \) from egg transfer coefficients \( \text{TC}_{\text{egg}} \) as:

\[
A_{\text{egg}} = \text{TC}_{\text{egg}} \text{ (d/kg)} \times P_{\text{egg}} \text{ (kg/d)} \tag{4}
\]

where \( P_{\text{egg}} \) is the production rate of eggs (0.0226 kg/d) \([14]\). Published values of \( \text{TC}_{\text{egg}} \) range from about 0.1 to 0.8 \([11-13, 15-17]\). Using only whole-egg values from chickens with chronic intake \([11, 16, 17]\), we found that the mean and the standard deviation of these values (Table I) were similar to this range.
2.1.4. Elimination rate coefficients

The total body biological elimination rate constant ($k_{\text{total}}$) of $^{137}\text{Cs}$ in chickens has been estimated as 0.026 d$^{-1}$ ($T_{\text{total}} = 27$ d) [23]. We also estimated $k_{\text{total}}$ from the long component of a two-component elimination of $^{134}\text{Cs}$ following an acute oral dose [12]. Estimates for the breast meat elimination rate constant of chickens under chronic intake ($k_{\text{breast}}$) are similar to those for the total body [15, 17]. Elimination from leg meat seemed to be substantially more rapid than that from breast meat [15, 17]. We assigned the longer retention time to breast meat, producing conservative estimates for meat as a whole.

The rate constant for the decrease in $^{137}\text{Cs}$ concentrations of whole eggs with successive layings ($k_{\text{egg}}$) was estimated from a weighted average (in proportion to mass) of rate constants for yolk and albumen [15] and for whole egg [17].

2.2. Model construction

Simulations were constructed using STELLA II simulation modelling software (High Performance Systems, Inc., Hanover, NH). The elimination time until safe $^{137}\text{Cs}$ concentrations ($[^{137}\text{Cs}]_s$, Bq/kg) were attained by random bred ($600$ Bq/kg for cooked meat, $300$ Bq/kg for eggs [24]) was estimated for different dietary $[^{137}\text{Cs}]_s$ and different times of providing contaminated feed. We did not estimate $^{137}\text{Cs}$ uptake from routes other than ingestion.

2.3. Field studies of the $^{137}\text{Cs}$ kinetics of bantam chickens

We conducted field studies on the $^{137}\text{Cs}$ kinetics of chickens at the US Department of Energy's Savannah River Site (SRS), a nuclear weapons facility located near Aiken, SC [25, 26]. Extensive areas of the SRS have been contaminated with low level releases of radionuclides, and we have studied the effects of body size (including relationships between sex and body size) and husbandry conditions on the $^{137}\text{Cs}$ kinetics of bantam chickens maintained in these contaminated habitats (Table III).

3. RESULTS

3.1. Growth and ingestion rate estimates

3.1.1. Growth and ingestion rates of random bred and bantam chickens

Estimates for the growth parameters $M_0$, $M_\infty$, $T$ and $m$, and the intercept and slope of Eq. (2) for the ingestion data are presented in Table I (parameters (1)-(4) and (5)-(6), respectively).
TABLE III. SOIL AND TOTAL BODY $[^{137}\text{Cs}]$ OF BANTAM CHICKENS ALLOWED ACCESS TO HABITATS CONTAMINATED BY NUCLEAR REACTOR DISCHARGES ON THE SAVANNAH RIVER SITE

<table>
<thead>
<tr>
<th>Location</th>
<th>$[^{137}\text{Cs}]$ in sediments (Bq/kg, dry)</th>
<th>$[^{137}\text{Cs}]$ in chickens (Bq/kg) $\bar{x}$ (95% confidence interval)</th>
<th>Soil:chicken concentration ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>H-area floodplain $^a$</td>
<td>7800 ($\bar{x}$ of 99 samples, this study)</td>
<td>$\varphi \varphi$ 5.74 (2.89–8.60)</td>
<td>1357</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\sigma \sigma$ 1.43 (−1.10–3.96)</td>
<td>5452</td>
</tr>
<tr>
<td>Group I $^b$</td>
<td></td>
<td>$\varphi \varphi$ 44.8 (35.5–54.2)</td>
<td>174</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\sigma \sigma$ 22.5 (17.3–27.6)</td>
<td>347</td>
</tr>
<tr>
<td>Group II $^c$</td>
<td></td>
<td>$\varphi \varphi$ 330 (x of 27 samples, this study)</td>
<td>58</td>
</tr>
<tr>
<td>Par Pond cooling reservoir $^d$</td>
<td>330 (x of 27 samples, this study)</td>
<td>$\varphi \varphi$ 5.63 (2.92–8.34)</td>
<td>138</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\sigma \sigma$ 2.39 (0.45–4.33)</td>
<td></td>
</tr>
<tr>
<td>Day 138</td>
<td></td>
<td>$\varphi \varphi$ 3.90 (2.33–5.47)</td>
<td>84</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\sigma \sigma$ 3.45 (1.20–5.70)</td>
<td>95</td>
</tr>
<tr>
<td>Day 321</td>
<td></td>
<td>$\sigma \sigma$ 127 (79–175)</td>
<td></td>
</tr>
<tr>
<td>Steel Creek floodplain $^e$</td>
<td>6600 [26]</td>
<td>$\sigma \sigma$ 87 (10–164)</td>
<td></td>
</tr>
</tbody>
</table>

$^a$ Equilibrium $[^{137}\text{Cs}]$ of bantam chickens released into an enclosure on the $^{137}\text{Cs}$ contaminated floodplain of a former reactor effluent stream.

$^b$ Uncontaminated feed provided from hanging feeder. The enclosure was denuded of plants by the chickens within 1 week after their release into the enclosure, and no native plants were present thereafter.

$^c$ Uncontaminated feed scattered directly onto contaminated soil. No native plants were available.

$^d$ $[^{137}\text{Cs}]$ of bantam chickens released into an enclosure located on the exposed lake bed of a drained nuclear reactor cooling reservoir. Feed was provided from a hanging feeder from days 0 to 160 and scattered directly onto the contaminated soil thereafter. No native plants were available.

$^e$ Uptake by free-ranging bantams 69 d after release into the $^{137}\text{Cs}$ contaminated floodplain of a former reactor effluent stream. The chickens had free access to uncontaminated feed and also to contaminated food items in the surrounding habitat; their gradually increasing use of these food items prevented their achieving equilibrium caesium concentrations.

Random breeds were weighed in mixed-sex groups, precluding tests of the effect of sex on growth, but there was a significant sex effect on the growth of bantams, with $\sigma \sigma$ achieving about 70% larger body sizes than $\varphi \varphi$ under both indoor and outdoor rearing conditions (Table II). Within each sex group, however, rearing conditions did not significantly affect the estimated values for $M_{\infty}$, $T$ and $m$. 
3.2. Simulation model predictions of $^{137}\text{Cs}$s in random bred chickens

3.2.1. Simulated $^{137}\text{Cs}$s of chicken meat and eggs

Simulation results indicated that larger chickens accumulated lower $^{137}\text{Cs}$s than smaller chickens (Table II). When the growth rate parameters for each sex/rearing condition were used in the simulation model, the resulting estimated equilibrium $^{137}\text{Cs}$s were about 5.1 times larger in breast meat and 1.2 times larger in leg meat of $\varnothing \varnothing$ compared with those of $\sigma \sigma$ (Table II). The concentration factors of breast meat, leg meat and eggs of random breds were about 0.67, 0.20 and 0.34 of feed, respectively, but were larger in the smaller bantams (Table II).

3.2.2. Predicted decontamination times for chicken meat and eggs

Simulation results showed that $^{137}\text{Cs}$ elimination from breast meat, leg meat and eggs was rapid once the chickens were removed from a contaminated diet (Fig. 2(a)), regardless of their size. We simulated the raising of random breds for 60–300 d on feed containing 1 kBq to 1 MBq of $^{137}\text{Cs}$/kg (Fig. 3). We determined the $^{137}\text{Cs}$s of meat and eggs under each combination of conditions and the number of days of providing uncontaminated feed that should be necessary for each food product $^{137}\text{Cs}$ to decline to values within the limits set by the European Economic Community (EEC) [24]. Our results indicated that the decontamination time was most influenced by the $^{137}\text{Cs}$ of feed and was little influenced by the amount of time the chickens were fed contaminated feed.

3.3. $^{137}\text{Cs}$s of bantam chickens in $^{137}\text{Cs}$ contaminated areas

3.3.1. Effects of body size and sex

In all four studies of bantams (Table III, Fig. 2(b)), $\varnothing \varnothing$ attained significantly higher $^{137}\text{Cs}$s than $\sigma \sigma$. However, the elimination rates of the sexes (Fig. 2(b)) were not significantly different ($F_{1,168} = 1.44; P = 0.23$). These findings, together with those from the simulation model, suggest that sex differences in $^{137}\text{Cs}$s can be explained by body size alone and that they result from differential $^{137}\text{Cs}$ intake, with the larger males having a lower mass specific ingestion rate.

3.3.2. Effects of husbandry

Bantams raised in enclosures on bare soil showed much lower $^{137}\text{Cs}$s than free-ranging chickens permitted to forage on contaminated biota, even when uncontaminated feed was provided ad libitum to both groups (Table III). From the differences in the amount of feed consumed, it appeared that the free-ranging
FIG. 2. (a) Simulated growth and $^{137}$Cs uptake of random bred chickens raised on feed contaminated with 1 kBq $^{137}$Cs/kg for the first 180 days after hatching. The graph illustrates the rapid decline in $[^{137}\text{Cs}]$ of the total body, breast meat, leg meat and eggs that results from high elimination rates of $^{137}$Cs in chickens and food products once the chickens are switched to an uncontaminated diet. (b) $^{137}$Cs kinetics of free-ranging bantam chickens foraging on the Steel Creek floodplain, which was contaminated by nuclear reactor effluents from production reactors on the Savannah River Site. Symbols and error bars indicate the mean and the 95% confidence intervals for the mean $^{137}$Cs of 7 ♀♀ and 7 ♂♂. After 69 d, the chickens were removed from the contaminated area and fed only uncontaminated feed. The half-times of the resulting elimination curves ($T_{\text{total}}$) closely match the simulation model predictions for total body elimination.
FIG. 3. Model estimates of production times required to raise chickens and then to 'decontaminate' the meat and eggs of chickens raised on $^{137}$Cs contaminated feed. Values on the x-axis indicate the $^{137}$Cs contamination level in the feed (Bq/kg). Values on the y-axis indicate the number of days that chickens are fed on $^{137}$Cs contaminated feed. The contour lines indicate the estimated number of days on uncontaminated feed that would be required for each food product from chickens raised under those conditions to sufficiently decrease in $^{137}$Cs to meet EEC limits (600 Bq/kg for cooked meat, 300 Bq/kg for eggs [24]).
bantams were obtaining a significant amount of their energy intake from contaminated forage (including detritus, plants and arthropods). Bantams showed increases in $^{137}\text{Cs}$s when feed was scattered directly onto contaminated soils (Table III). In one enclosure study (Par Pond), the body $^{137}\text{Cs}$ did not significantly increase above detectable levels as long as the feed was not scattered onto the contaminated sediments.

4. DISCUSSION

Decisions on acceptable limits of radionuclide concentrations in poultry products can also be made on a dose equivalence basis to accommodate differences in human diets and other variables. However, our results suggest that under indoor husbandry, the probability of producing unsafe poultry products (especially eggs) is likely to be low at realistic feed contamination levels. Even when the dietary $^{137}\text{Cs}$s are sufficiently high to produce poultry $^{137}\text{Cs}$s above the proscribed limits, those levels may be reduced within a reasonable time span by providing uncontaminated feed. Our predictions of short decontamination periods for $^{137}\text{Cs}$ contaminated meat and eggs are further validated by other studies of chickens consuming contaminated feed [16, 17].

On the basis of our model results and field studies, we can offer recommendations on ways to reduce $^{137}\text{Cs}$ uptake by chickens. First, if meat production is to be emphasized, larger breeds (preferably $\sigma\sigma$) are preferable because of their lower mass specific feeding rates and the concomitantly decreased $^{137}\text{Cs}$s in meat. However, it appears likely that laying hens can also be converted into meat products at the end of their productive egg laying period if they are fed for a short time uncontaminated feed prior to slaughter. Second, managing the $^{137}\text{Cs}$ of feed to produce acceptable $^{137}\text{Cs}$s in breast muscle should also result in acceptable leg meat and egg concentrations. Third, indoor rearing conditions appear to reduce potential equilibrium $^{137}\text{Cs}$s significantly, primarily by preventing access to contaminated plants. If chickens are to be reared in outdoor pens, we recommend clearing those pens of all plants prior to use and (if possible) covering the exposed earth with an uncontaminated substrate.

Additional studies are necessary to fully evaluate the practicality of poultry production in contaminated areas, including production costs, recycling of byproducts (including possible conversion of inedible portions of carcasses into protein meal, or the production of heat and electricity from methane generated from manure), and problems associated with concentration and disposal of waste products. Future studies should also consider the kinetics of other long lived radionuclides and their possible contrast to the conclusions for $^{137}\text{Cs}$ in this study. However, the long lived radionuclides with the greatest potential for affecting the safety of poultry meat and eggs ($^{90}\text{Sr}$ and Pu) are even less readily incorporated into edible portions of
poultry products than is $^{137}$Cs [11, 14]. Other pre- and post-processing techniques can further reduce radionuclide levels. For example, additions of chelating agents to the feed can retard radionuclide assimilation [16]. Commercial packaging can reduce public exposure to bone seeking radionuclides by washing [27] or by removing the more highly contaminated egg shells and bones prior to delivery to commercial markets. Additional removal of radionuclides from food products may be accomplished through cooking of the meat [28].

We believe that alternative agricultural production practices have the potential to return many contaminated areas to productive use. Although we have emphasized poultry production here, other agricultural products might also be produced safely in contaminated areas. For example, production of fibre crops (e.g. hemp) or energy crops (e.g. alcohol or methane production from plant biomass) offer commercially practicable alternatives to food production. Obviously, the potential risks from such practices should be considered, but they should be weighed against the financial and ecological costs of remediation, and the financial and sociological costs to rural populations denied access to their traditional way of life. These risks should also be evaluated in the light of the fact that thousands of persons currently inhabit contaminated areas and are already practicing subsistence agriculture without the benefit of safety evaluations.

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REFERENCES


USE OF CAESIUM BINDERS TO REDUCE RADIOCAESIUM CONTAMINATION OF MILK AND MEAT IN BELARUS, RUSSIA AND UKRAINE

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Abstract

USE OF CAESIUM BINDERS TO REDUCE RADIOCAESIUM CONTAMINATION OF MILK AND MEAT IN BELARUS, RUSSIA AND UKRAINE

Caesium binders of the hexacyanoferrate type (Prussian blue, PB) have been tested under field conditions in territories of Belarus, Russia and Ukraine that were relatively heavily contaminated by fallout from the Chernobyl accident. Typically, the $^{137}$Cs concentrations in milk and meat of cattle consuming contaminated feeds were reduced by factors of two to six when PB was provided by means of sustained release boli. Similar reductions were also observed in cattle receiving PB through salt licks or in the form of powder. By administering PB boli twice during the grazing season it was possible to produce milk that did not exceed the temporary permissible caesium levels of 370 Bq/L on pastures which otherwise led to $^{137}$Cs concentrations of 800–1300 Bq/L milk. Uptake of $^{137}$Cs by legumes and grasses was
reduced by 37–51% when the soil was fertilized with manure from cows receiving PB supplements. It is concluded that PB treatment is a simple and effective way of controlling the transfer of $^{137}\text{Cs}$ through the food-chain from soil to domestic animals and man. The techniques tested in the present experiments are being used routinely for 40,000 to 50,000 cattle in Belarus, Russia and Ukraine.

1. INTRODUCTION

In order to reduce the transfer of radionuclides from the Chernobyl accident through the food-chain to man, a range of countermeasures, including deep ploughing and fertilization of soils, have been implemented in Belarus, Russia and Ukraine. Despite such measures, some areas have continued to experience difficulties in achieving radiocaesium concentrations in milk and meat from domestic animals that are below national or regional temporary permissible limits (TPLs, Table I). These TPLs are lower than the FAO/WHO Codex Alimentarius Guideline levels for food moving in international trade. In particular, relatively high $^{137}\text{Cs}$ concentrations persist in milk from privately owned cattle. These animals have limited access to deep ploughed fields and graze natural vegetation to a large extent. Even in 1994, the products from a considerable number of farm animals still had $^{137}\text{Cs}$ levels that exceeded the existing TPLs (Table I).

In several countries, different caesium binding agents have been used to control the radiocaesium concentrations in animal products [1]. Iron-hexacyanoferrate compounds with ligands of potassium, ammonium or iron, here collectively called Prussian blue (PB), have been most useful in treatments of grazing ruminants [2–4], particularly because of their effectiveness in small quantities. Typically, reductions in radiocaesium transfer of 50-90% can be obtained by daily doses of PB in the range of 0.5–10 mg/kg body weight [4]. Different strategies for administration of PB have been developed and tested earlier [5]. The present experiments have been performed to assess PB treatments under field conditions in private and collective farms in the three Republics of the commonwealth of independent States (CIS). Since private farms rely heavily on cow manure as a fertilizer, the effect of manure containing PB on the transfer of $^{137}\text{Cs}$ to cultivated plants was also tested. Recent literature surveys describe PB compounds as toxicologically safe, both for the animals themselves and for humans consuming their products [6]. Similar results have been obtained in official toxicological tests of meat and milk collected from cattle treated with PB boli in experiments carried out as part of the process required to receive the necessary permits for use in each of the three CIS Republics.

The experiments reported focus on the use of sustained release boli which dissolve in the rumen of cattle over time and release the PB to the rumen liquid. Experience gained in experiments not reported here has also prompted the use of other methods for administration of PB caesium binders.
TABLE I. ESTIMATE OF NUMBER OF DAIRY AND BEEF CATTLE EXCEEDING TEMPORARY PERMISSIBLE LEVELS (TPLs) FOR MILK AND MEAT IN THE CONTAMINATED AREAS OF BELARUS, RUSSIA AND UKRAINE (1992 and 1994)

<table>
<thead>
<tr>
<th></th>
<th>Dairy cows</th>
<th></th>
<th>Beef cattle</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Total number of animals in contaminated areas&lt;sup&gt;a&lt;/sup&gt;</td>
<td>Milk TPLs (Bq/L)</td>
<td>Cows with milk exceeding TPLs</td>
</tr>
<tr>
<td>Belarus</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Collective</td>
<td>393 000</td>
<td>185</td>
<td>3 000</td>
</tr>
<tr>
<td>Private</td>
<td>84 000</td>
<td></td>
<td>25 000</td>
</tr>
<tr>
<td></td>
<td></td>
<td>to 30 000</td>
<td>to 30 000</td>
</tr>
<tr>
<td>Russia</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Collective</td>
<td>200 000</td>
<td>370</td>
<td>15 000</td>
</tr>
<tr>
<td>Private</td>
<td>25 000</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ukraine</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Collective</td>
<td>170 000</td>
<td>370</td>
<td>—</td>
</tr>
<tr>
<td>Private</td>
<td>60 000</td>
<td></td>
<td>—</td>
</tr>
<tr>
<td>Total</td>
<td>932 000</td>
<td>45 000 to 50 000</td>
<td>1 500 000</td>
</tr>
</tbody>
</table>

<sup>a</sup> Defined as areas where depositions of $^{137}$Cs, $^{89}$Sr and $^{239}$Pu fallout exceed 1, 0.15 and 0.01 Ci/km$^2$, respectively (1 Ci = $3.7 \times 10^{10}$ Bq).

<sup>b</sup> Some contaminated districts have lower levels (Gomel region: 37 Bq/L milk and 370 Bq/kg meat.)
2. EXPERIMENTS WITH PRUSSIAN BLUE BOLI

In an introductory series of experiments, dairy cattle in collective farms in each of the three Republics were given single treatments with two, three or four sustained release boli. The boli weighed 250–300 g each, contained 15% PB and in addition barite and beeswax, as described in Ref. [7]. Experiments were performed at the end of the grazing season (October) and during parts of the indoor feeding period; altogether they lasted between 75 and 110 d. The results of experiments with dairy cattle, carried out on a collective farm in the Polyeska region in Ukraine, with initial $^{137}$Cs concentrations of about 160 Bq/L milk, showed that a single treatment with two boli reduced the $^{137}$Cs concentrations in milk by factors of two to four, while a treatment with four boli reduced the $^{137}$Cs levels in milk by factors of two to six (Fig. 1). Decreases in the $^{137}$Cs levels in milk by factors of two to four were observed immediately after treatment, and the reductions were most pronounced during the period from 30 to 50 d after treatment. Thereafter, the effects of the boli gradually diminished and no effects were detectable 120 d after treatment (Fig. 1). Normal milk production was maintained in treated animals. Similar results were obtained in experiments in collective farms in Russia and Belarus.

![Graph showing Caesium-137 concentrations in milk of cattle treated with control, two, or four boli providing PB. Each boli weighed 300 g and contained 15% of Giese-salt, a mixture of 65% ammonium-iron(III)-hexacyanoferrate(II) and 35% ammonium chloride, as the active ingredient. Means and standard deviations, ten animals in each group.](image)
FIG. 2. Caesium-137 concentrations (Bq/L) in milk of dairy cattle on private farms in the village Velikiye Ozera in Ukraine. The cows received three sustained release boli providing PB at the start of the grazing season (May), and additional two or three boli 60 days later. The boli were the same as in Fig. 1. Control animals (15) and treated animals (15) were grazed on the same fields throughout the summer period. Means and standard deviations.

In Ukraine, two demonstration experiments were set up under practical field conditions during the summer of 1991. Data from the village Velikiye Ozera are given in Fig. 2. On each of 15 private farms, one cow was given three boli in May and two or three additional boli 60 d later in the grazing season. Untreated cows (15), which were grazed on the same fields, served as controls. The $^{137}$Cs concentrations in milk were monitored weekly. Treatment with boli resulted in reductions in milk activities by factors of two to six over the observation period of 91 d. The treatment allowed production of milk with less than 370 Bq $^{137}$Cs/L (current TPL) on a pasture which otherwise gave rise to concentrations of between 800 and 1300 Bq $^{137}$Cs/L milk (Fig. 2).

3. USE OF MANURE FROM COWS TREATED WITH PRUSSIAN BLUE BOLI

To study the availability to crops of $^{137}$Cs in the manure of PB treated cattle, 15 cows in the Novozybkov district in Russia were fed rations containing 90–200 kBq $^{137}$Cs/d. Half of the animals were given three sustained release boli
with PB. Manure was collected for 21 d and used in 1991 to fertilize experimental plots (100 m$^2$ each) in a large field with sandy podzolic soil. Manure from treated animals contained 2.8 kBq $^{137}$Cs/kg (wet weight), while that of untreated animals contained 2.9 kBq $^{137}$Cs/kg. The manure was given at a rate of 4 kg/m$^2$ (40 t/ha) and provided 10-11 kBq/m$^2$ in fields where the deposition of $^{137}$Cs was about 400 kBq/m$^2$. Each treatment was given on four replicate plots. Lupin was grown in the first year and used as a green fertilizer for the subsequent crop, which was a mixture of timothy and cockfoot grasses. Two grass harvests were taken in 1992. Mineral fertilizer was given in 1991 and 1992, while manure was applied only in the first year. The transfer parameters for $^{137}$Cs from soil to vegetation were compared by calculating a transfer coefficient (TC) based on $^{137}$Cs concentration in soil, and an aggregated transfer factor ($T_{ag}$) based on the $^{137}$Cs content per square metre of soil (Table II). Compared with unfertilized plots, the plots to which mineral fertilizer had been applied had a markedly reduced $^{137}$Cs transfer from soil to lupin and from soil to grass (Table II). Mineral fertilizer and animal manure in combination reduced the transfer from soil to grass more than mineral fertilizer alone. When manure with PB was applied, both the TC and the $T_{ag}$ from the soil to the lupin crop were reduced by 37% compared with manure from control animals, showing that the $^{137}$Cs in soil fertilized with PB containing manure was less available for plant uptake than the $^{137}$Cs in soil where the manure contained no PB. The PB manure given in 1991 also reduced the $^{137}$Cs uptake in the grass harvested one year later from the same fields (Table II). During the second harvest in 1992, the $^{137}$Cs concentrations in grass were found to be increased on all treatments, probably as an effect of reduced availability of plant nutrients in the second part of the growing season. Application of PB manure was also effective in reducing the transfer parameters in this situation. The results show that PB supplied to the soil through organic manure will reduce the transfer of $^{137}$Cs to crops for at least two years. It is of considerable interest to determine the duration of this sequestration of $^{137}$Cs in soil and the extent to which the added PB also interacts with $^{137}$Cs in the soil solution. Obviously, the long term consequences of PB application to soil will have to be studied further. The present findings nevertheless indicate that PB treatment of cattle can be useful also by reducing the transfer of $^{137}$Cs to plant crops. Through binding to PB, some of the $^{137}$Cs which accumulates in family gardens as a result of the farming practices will be prevented from entering the human food-chain.

4. COUNTERMEASURES BASED ON PRUSSIAN BLUE CURRENTLY USED IN THE CONTAMINATED REGIONS OF BELARUS, RUSSIA AND UKRAINE

Prussian blue has been most widely used in Belarus to control the $^{137}$Cs content of animal products. About 27 t of PB have been or will be used annually to treat
TABLE II. CONTENT OF $^{137}$Cs IN SOIL AND DRIED VEGETATION HARVESTED FROM FIELDS FERTILIZED WITH MINERAL FERTILIZER (MF, 1991 AND 1992) AND MANURE (1991 ONLY)*

<table>
<thead>
<tr>
<th>Soil</th>
<th>Crop</th>
<th>TC soil–veg.</th>
<th>$T_{ag}$ soil–veg.</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Bq/kg)</td>
<td>(Bq/kg)</td>
<td>(m$^2$/kg)</td>
<td>(m$^2$/kg)</td>
</tr>
<tr>
<td>Lupin (1991)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Control</td>
<td>1220 ± 120</td>
<td>405 ± 85</td>
<td>0.32</td>
</tr>
<tr>
<td>MF</td>
<td>1220 ± 60</td>
<td>272 ± 13</td>
<td>0.22</td>
</tr>
<tr>
<td>MF + manure</td>
<td>830 ± 40</td>
<td>182 ± 10</td>
<td>0.22</td>
</tr>
<tr>
<td>MF + PB + manure</td>
<td>920 ± 50</td>
<td>126 ± 14</td>
<td>0.14</td>
</tr>
<tr>
<td>Grass (1992)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>First harvest</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Control</td>
<td>1040 ± 100</td>
<td>419 ± 40</td>
<td>0.40</td>
</tr>
<tr>
<td>MF</td>
<td>1150 ± 70</td>
<td>248 ± 11</td>
<td>0.22</td>
</tr>
<tr>
<td>MF + manure</td>
<td>840 ± 40</td>
<td>127 ± 7</td>
<td>0.15</td>
</tr>
<tr>
<td>MF + PB + manure</td>
<td>1080 ± 40</td>
<td>99 ± 8</td>
<td>0.09</td>
</tr>
<tr>
<td>Second harvest</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Control</td>
<td>970 ± 80</td>
<td>540 ± 61</td>
<td>0.60</td>
</tr>
<tr>
<td>MF</td>
<td>1090 ± 50</td>
<td>376 ± 18</td>
<td>0.35</td>
</tr>
<tr>
<td>MF + manure</td>
<td>840 ± 50</td>
<td>188 ± 12</td>
<td>0.23</td>
</tr>
<tr>
<td>MF + PB + manure</td>
<td>1040 ± 70</td>
<td>118 ± 9</td>
<td>0.14</td>
</tr>
</tbody>
</table>

* Manure was taken from animals fed highly contaminated hay (intake 90–200 kBq/d). Individual plots received manure from cows treated with three PB boli or from animals with no PB treatment (control). Grasses grown were a mixture of timothy (*Phleum pratense*) and cockfoot (*Dactylis glomerata*). Transfer coefficients (TC) were calculated as Bq/kg of dried vegetation per Bq/kg of soil. Aggregated transfer factors ($T_{ag}$) were calculated as Bq/kg of forage dry matter per kBq/m$^2$ of $^{137}$Cs in the plot.

In 1991, the mineral fertilizer was 60 kg/ha of a potassium-phosphorous fertilizer with 60 kg/t of P and K. In 1992, 60 kg/ha of fertilizer was used which provided 90 kg/t of N and 60 kg each of P and K. Manure was applied at a rate of 40 t/ha.
about 30,000 cows during the period 1993–1995. Bolus with PB were produced for treatment of 9,000 cows in 1993 and 16,000 cows in 1994. Simultaneously, 33 t of PB salt licks were provided in 1994. Furthermore, farmers will be provided with PB containing concentrate on a regular basis, starting in 1995. In Ukraine, a total of 6,500 cows were given PB, mostly in the form of bolus (for about 5000 cows). Encouraging practical results have also been achieved in Ukraine by providing private farmers with PB powder. Pre-weighed daily doses of PB powder were used for direct application on feed concentrates and other forms of feeds. Budgetary constraints have not allowed purchase of the required amounts of PB in Ukraine.

In Russia the use of PB caesium binder techniques was delayed by the lack of official permission. Good results have been obtained with a PB preparation (Bifege) with 10% precipitated into cellulose (sawdust). This preparation is considered to be convenient in that it can be spread onto feed for individual cows, both in collective and private farms, at doses of 30–60 g/d. In 1993 and 1994, 9 t and 16 t of Bifege were used to treat about 4,000 dairy cows in the Bryansk region of Russia.

5. CONCLUSIONS

Prussian blue is an effective and convenient caesium binder for use in dairy and beef cattle and may be administered as powder, salt lick or bolus, or mixed with concentrate feed or cellulose. Manure from cows fed PB reduces the incorporation of radiocaesium into vegetation, at least over a period of 2 years. Considerable benefits can be gained from the use of PB as a countermeasure for contaminated regions, since it reduces the internal radiation dose to people and simultaneously assists in continued operation of farms. Administration of PB is the method of choice when $^{137}$Cs levels exceeding TPLs persist after radical soil improvement and in areas where soil countermeasures are not feasible.

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REFERENCES


ASSESSMENT OF WATER PROTECTION OPERATIONS TO MINIMIZE THE RISK FROM USING WATER FROM THE DNIEPER WATER SYSTEM AT VARIOUS STAGES AFTER THE ACCIDENT AT THE CHERNOBYL NUCLEAR POWER PLANT

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Abstract

ASSESSMENT OF WATER PROTECTION OPERATIONS TO MINIMIZE THE RISK FROM USING WATER FROM THE DNIEPER WATER SYSTEM AT VARIOUS STAGES AFTER THE ACCIDENT AT THE CHERNOBYL NUCLEAR POWER PLANT.

Analysis of the series of measures taken to minimize the consequences of contamination of water systems subjected to radiation as a result of the accident at the Chernobyl nuclear power plant in 1986 could provide a unique opportunity for decision makers and others working in the contaminated areas to optimize methods of protecting surface water and groundwater. Most of the engineering and technical measures undertaken within the 30 km Chernobyl zone were concentrated on preventing secondary contamination of surface water and groundwater draining into the Pripyat river and the Kiev reservoir. However, implementation of these measures required vast financial and human resources. The lessons learned from the post-Chernobyl water protection operations therefore need to be carefully analysed. The paper summarizes the results of nine years' work to mitigate the consequences of radioactive contamination of water systems within the 30 km zone. The results show that the effectiveness of countermeasures to reduce the consequences of radioactive contamination of water systems depends not only on the correct use of technology but also on an appropriate strategy for assessing the results of efforts to reduce the potential radiation risk. In view of the present economic situation in Ukraine, schemes for mitigating environmental impact must achieve maximum risk reduction yet also be economically viable, otherwise financial resources could be wasted, leading to serious social problems.
1. **INTRODUCTION**

Numerous research projects conducted since the accident at Unit 4 of the Chernobyl nuclear power plant (NPP) have shown an increase in radioactive contamination levels in most regions of Ukraine, Belarus and Russia and in parts of western Europe as a result of this accident. It is well known that most of the radioactive fallout took place over the catchment areas of the Dnieper basin. The catchment basin around the Chernobyl NPP covers a large area, from which the overland flow contaminated with radionuclides drains into the Pripyat and Dnieper rivers to be conveyed via a whole series of reservoirs to the Black Sea. The surface run-off passing through contaminated land contributes to the migration of radionuclides from the Chernobyl contamination zone to the vast Dnieper region with over 20 million inhabitants whose diet includes drinking water (for about nine million people), fish and irrigated farming products, which are all dependent on water from the Dnieper. These issues are still of critical concern to the population, even nine years after the accident, and controversy over the water protection measures in the Chernobyl zone is continuing. The protection of water systems and the efforts made to decontaminate and rehabilitate the area around the plant already have a dramatic history, which needs to be assessed in order to see what lessons can be learned.

2. **ASSESSMENT OF THE DIFFERENT STAGES OF WATER PROTECTION AFTER THE ACCIDENT AT THE CHERNOBYL NPP**

In the period following the accident, various technical and administrative measures were taken which were aimed at limiting the radiation impact on aqueous ecosystems and other elements of the aqueous environment, in order to protect the population living in the areas along the Dnieper reservoirs. These measures can be classified into three phases, as follows.

2.1. **Emergency phase** (the first two to three months after the accident)

During this period the measures taken were based mainly on administrative decisions and were primarily aimed at monitoring and attempting to control the situation. These measures included: attempts to regulate the flow of contaminated water through the Kiev reservoir by means of the system of gates on the dam of the hydroelectric plant; increasing the use of underground water sources in towns and, wherever possible, reducing to a minimum the use of contaminated surface water; additional purification of drinking water at water treatment facilities in towns and digging of new wells to tap groundwater. Most of these measures were implemented without assessment of their cost efficiency, in view of the lack of time for research, the lack of necessary experience, and the emotional state of the community and of
those taking decisions in the state of emergency after the accident. Most of the countermeasures taken were therefore very expensive and were only of limited value for reducing the radiation risk to the population from the use of water from open reservoirs.

Various examples of preventive measures taken to protect water systems have been discussed previously in Refs [1-5], where typical strategic mistakes committed by decision makers are described. Miscalculations were made because of the lack of adequate information on the processes taking place and the absence of reliable forecasts of the behaviour of radionuclides in catchment basins, the river system and reservoirs, both immediately after the initial radioactive fallout and over a longer period, from several months or years to periods on a geological time-scale.

For example, the lack of experimental data and differences between scientists and decision makers, who had no experience in dealing with such problems, meant that the first estimates of kinetic parameters for the sorption and desorption processes determining the interphase interaction of radionuclides in liquid and solid media were incorrectly performed. The radionuclide wash-out coefficients were put much higher than the values actually occurring, which led to an overestimate of the potential contamination of river water in the accident zone. As a result, many useless or inefficient water protection measures were taken in the first months after the accident. For example, untreated and unpurified zeolite was dumped into the water from barges on the river fairway downstream from Chernobyl, etc.

At the beginning of May 1986, the upper gates of the sluice at the Kiev reservoir dam were opened and the bottom gates closed. It was reckoned that pure water would flow out of the reservoir through the upper spillway while the highly contaminated water, interacting with sediment, would concentrate in the bottom of the reservoir. In order to prevent release of contaminated bed sediment into the tail water of the dam, the lower flood gates were closed. In fact, in the first few weeks after the radioactive release, before complete mixing had occurred, the water in the lower layers of the reservoir was much less contaminated than that in the upper layers because of concentration of atmospheric fallout in the surface layers of the water. The best way of reducing the level of the water in the Kiev reservoir immediately after the accident would have been to open the lower gates and to close the upper ones. In this way the level of radioactivity in fresh water downstream of the Kiev reservoir could have been reduced in the first weeks after the accident when exposure from drinking water occurred. Reference could be made to many other incorrect actions which could have been avoided if there had been better understanding of the situation and of the behaviour of radionuclides in reservoirs.

2.2. Early intermediate phase (from summer 1986 to 1988)

In the summer of 1986, protective dykes were constructed along several kilometres of the river Pripyat in order to hold back contaminated surface run-off
from land around the towns of Chernobyl and Pripyat. These measures were ineffective, as it was impossible to control the run-off from large areas of land.

During the first summer-spring period after the accident, several special pits were dug in the bed of the river Pripyat, in order to increase the cross-sectional area of the river, to reduce the water velocity and thereby to promote more effective settling of the suspended radioactive particles in the flow of water according to Stokes’ law. However, a later study [4] showed that these sediment traps were ineffective, as the suspended radioactive particles were too small to be retained in the strong natural current of a river such as the Pripyat, with large volumes of water and turbulent flow conditions [6].

A major task in the first years after the accident was to try and isolate the contaminated water in the cooling pond of the NPP from the river Pripyat. A special system of drainage shafts was constructed around the pond, in order to prevent percolation of radioactive water. The drainage system has not been brought into operation as yet because of uncertainty as to its possible effects [3, 4]. More recent studies [5] have shown that the pumping back of water from these shafts into the cooling pond could cause problems with the balance of water and dissolved salts in the pond. In any case, this operation would be too costly in terms of construction and maintenance.

One of the actions taken during this early period, in addition to the construction of the series of drainage shafts, was the construction of protective banks along the river. These projects were initiated at a time when insufficient information was available and so the most conservative scenarios were accepted without proof. As additional information was acquired, together with results of research confirming that the concentration and mobility of radionuclides in surface water were significantly less than had been estimated, the projects were scrapped.

During 1986 and the beginning of 1987, construction of over 100 special dykes containing zeolite (a natural mineral sorbent) was completed on minor rivers in order to adsorb radionuclides from the water. A subsequent study of the effectiveness of such dykes in capturing radionuclides showed that only 5-10% $^{90}\text{Cr}$ and $^{137}\text{Cs}$ was retained in the body of the filtering dyke by the zeolite barrier. In addition, as a result of the low adsorption volume of the zeolite in the dykes, much greater problems occurred in the larger rivers with high flow velocities. The filtering dykes were very quickly silted up by river sediment, their permeability decreased, and the water levels in the river rose higher than the dykes, leading to flooding of radioactively contaminated flood plains and additional wash-out of radioactivity into the rivers from the soil in such areas. In 1987, the construction of new dykes was stopped and it was decided to demolish the majority of the existing dykes [1, 4].

In 1987, after evacuation of the local population and extinction of the fire at the reactor, one of the measures taken to mitigate the consequences was the burial at several temporary radioactive waste repositories of contaminated soil, vegetation, construction materials and small buildings from the site of the Chernobyl NPP,
together with highly contaminated 'red trees' from the pine forest (which had died owing to the high radiation level). These measures were considered essential to protect the cleanup personnel and plant workers from high doses of radiation. In order to reduce the exposure of persons working in the Chernobyl zone, the waste was buried in sand pits and trenches, without any special barriers to prevent radioactivity from entering the groundwater. These burials in the 'red forest' area have caused serious local contamination of the groundwater and restoration of the environment has now become a serious long term problem.


The new phase of restoring water resources in the exclusion zone began after the flood in summer 1988. The resulting high water level covered the contaminated flood plain and caused secondary contamination of the river systems with $^{90}$Sr [3, 4, 6]. Modelling of cases of surface water contamination shows that the most dangerous or 'worst-case' scenario which could cause the highest concentrations of radionuclides in rivers could actually occur during a spring flood with maximum flows of up to 2000 m$^3$/s. As a result of modelling the effect of various ways of isolating the contaminated flood lands from contact with river water in the flood scenario and reducing the wash-out of radionuclides from the soils in the flood plain, the concentration of $^{90}$Sr in the river water was reduced by a factor of two to four. Several methods were suggested and the potential effectiveness of these or other measures for reducing the radionuclide concentration was modelled. As a result of the model studies, it was suggested that the best form of protection would be to construct water protection dykes along the contaminated land on the left (eastern) river bank in the area close to the Chernobyl NPP. This measure, combined with soil decontamination on the right river bank (possibly together with dykes), as well as an appropriate method for controlling the percolation of contaminated water from the cooling pond, appeared likely to be capable of reducing considerably the $^{90}$Sr concentration in the lower reaches of the river Pripyat. The events of January 1991 and of the summer of 1993, with inundation of the flood plain, definitively confirmed the validity of the modelling results. The dyke construction was completed by the end of 1992. During the spring flood in 1994, these measures prevented the wash-out of more than 100 Ci of $^{90}$Sr from the flood plain of the Pripyat into the Dnieper cascade.\footnote{1 Ci = 37 GBq.} After an event such as the 1991 flood in the Chernobyl area it became clear that no other measures could be projected without thorough testing and comparison of the planned techniques or without developing a unified strategy, i.e. a 'master plan' for protecting the aqueous environment in the area around Chernobyl. The engineering scheme was partially completed in 1993 and the scientific basis for the whole operation is currently being worked out.
3. CURRENT UNDERSTANDING OF THE PROBLEM

At present, the zone around Chernobyl contains a large quantity of radioactive materials which, as a result of the accident in 1986, entered the soil in the catchment areas and also the water and bottom sediment of lakes and the cooling pond of the Chernobyl NPP. For example, over 10,000 Ci of $^{90}$Sr and approximately 200 Ci of plutonium is concentrated in the flood plains, which are liable to be inundated during the spring flood, and in the soil in the polder areas within the 30 km zone, which are liable to be flooded every year by river water and groundwater during snow melts and in rainy seasons. A large amount of radioactive natural waste is concentrated at waste burial sites in contact with groundwater which flows in the direction of the river Pripyat. Potential sources of secondary contamination of the water system are discussed in detail by the authors of Refs [2, 3, 7].

The first stage in protecting the aqueous environment in the Chernobyl area was based on the realistic assessment that technological measures to control existing sources of radioactive contamination over such large catchment areas have only a very limited effect. The main research previously conducted had shown that operations to protect water systems can only be optimized by comparing the actual doses received by the population (which could have been reduced by countermeasures) with the doses accumulated as a result of work carried out to protect and decontaminate the water systems in the Chernobyl area.

On the basis of these studies, the results of monitoring data and the projected content of radionuclides in Dnieper water up to the year 2056 were used to calculate the potential dose accumulated over a period of 70 years, together with the total external doses of $^{90}$Sr and $^{137}$Cs received by the population of the Dnieper region. The structure of the accumulated dose (over 70 years) resulting from water consumption, as an average for all water consumers along the Dnieper subject to exposure from drinking water, fish and other foodstuffs, works out at 35, 40 and 25%, respectively. The studies have also shown that the dose pattern is different for different regions of Ukraine. It has also been shown that the annual effective internal individual doses vary significantly in different regions of Ukraine. For example, in 1993 the contribution to the total radiation dose of the water component for inhabitants of Kiev was approximately 6%; for people living in the southern regions of Ukraine it was as high as 20–30% [7, 8].

4. ASSESSMENT OF RADIATION RISK TO HEALTH FROM WATER CONSUMPTION

Using the coefficient of nominal risk probability — $7.3 \times 10^{-2}$ Sv$^{-1}$ (ICRP 60) — the number of cancer cases associated with water consumption from the Dnieper was calculated for a 70 year period. This amounted to approximately
200 cases for 21 million inhabitants (70 year period) and approximately 60 cases (for the period 1986-1992). A recalculation of the total dose to the whole population exposed to radiation (approximately 21 million) may show that the average radiation risk to the population from the consumption of Dnieper water would be of an order of magnitude not higher than $1 \times 10^{-5}$. For certain critical population groups the expected risk from water consumption might be reduced by a factor of four to five or even more. In fact, compared with other sources of radiation risk, the aqueous migration path constitutes a very low level of radiation risk for the population. At the same time, the stress component of radiation risk, which reflects the psychological reaction of the population consuming radioactively contaminated water, may in some cases predominate over the pure dose component in its effect on health.

According to our research [8], over 30% of the people surveyed, who had varying levels of education, assumed, in the absence of any special knowledge of radiation protection, that the danger to health from water consumption was greater than that from other sources. However, the mean individual dose from natural radionuclides such as $^{226}$Rn, $^{222}$Rn and $^{238}$U in drinking water in the regions mentioned may be two orders higher than the risk component due to the Chernobyl accident.

Unfortunately, to date we have no clear theory or methods for calculating the total risk from environmental contamination and the partial risk due to water consumption when there is multicomponent contamination of water systems. For example, all of the Dnieper reservoirs are situated in industrial and agricultural regions with a high level of non-radioactive environmental contamination. Toxicological studies have shown that the reservoir water contains a range of other toxic substances with strong carcinogenic and mutagenic properties. In many cases, in contrast to radionuclides, the toxic nature of these substances is unknown or they are not monitored. It is therefore very difficult to specify urgent remedial measures for water protection when the Dnieper reservoir water contains other toxic materials. This is a very important point to consider when assessing the health risks to the population living in the contaminated areas.

5. CONTEMPORARY STRATEGY FOR PROTECTING THE AQUEOUS ENVIRONMENT IN THE CHERNOBYL AREA

Despite the assessment that water-borne radionuclides (mainly from sources located in the Chernobyl area) have only a low impact on the Ukraine population, attention should be given to the problem of contamination of the Dnieper water system. In the case of Chernobyl, where the sources of radioactive contamination of water are known, it was much more preferable to determine a priority set of options for protecting the aqueous environment than to remain inactive. However, no remedial measures could be implemented without demonstrating their real advantages in accordance with the well known ALARA principle. Based on these principles, a technical plan for urgent remedial measures for protecting the aqueous
environment was established in 1993. The primary aims of the countermeasures involved are to curtail further spreading of the effects of the accident by effective controls and by minimization of radionuclide transport via water from the Chernobyl area, and also to develop a radiation monitoring system for surface water and groundwater both inside and outside the Chernobyl area.

As a basic criterion for decision making and for selecting optimal remedial measures, a value of 2 Bq/L has been specified for $^{90}$Sr concentration in the lower reaches of the river Pripyat on the border of the Chernobyl area, which would permit water to be used safely at any point in the Dnieper reservoir system [8].

The following remedial actions have been drawn up to help achieve the above goals:

1. Completion of the embankment of flood lands with extremely high levels of contamination on both sides of the river close to the Chernobyl NPP, in order to protect these areas from potential direct flooding and soil erosion caused by the river flow.

2. Production of a scheme to solve the problems involved in decontaminating the cooling pond deposits after closure of the Chernobyl reactor.

3. Regulation of the water in the flood plains of the Chernobyl area having a very high level of radioactive contamination, in accordance with the scheme adopted in Belarus of flooding the peat bogs as a fire prevention measure.

4. Extension of the monitoring of the groundwater system within the Chernobyl area and around the temporary waste storage sites; and introduction of reliable monitoring and control of the transport of transuranium material by groundwater and surface water beyond the boundaries of the waste storage sites.

5. Prevention of further transport of radionuclides from their present location at the waste storage sites, which could occur as a result of contact with groundwater, by constructing technical and geochemical barriers around the storage sites.

Any other proposals aimed at restoring water resources to their natural state would not be effective at the present time and the funds allocated to finance them should be temporarily frozen.

REFERENCES


TOPICAL DISCUSSIONS

(Session 9)

Chairman

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PROTECTION OF THE NATURAL ENVIRONMENT FROM IONIZING RADIATION

Are specific criteria needed?

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Abstract

PROTECTION OF THE NATURAL ENVIRONMENT FROM IONIZING RADIATION: ARE SPECIFIC CRITERIA NEEDED?

It is generally believed that health protection criteria developed for human exposure to environmental radioactivity will ordinarily provide adequate protection for natural populations of other organisms. Several lines of evidence support this belief. However, there are special situations where adherence to human protection standards may not provide sufficient protection to other species. Furthermore, there is still some uncertainty as to whether reproduction — the biological damage end-point used to develop the basic paradigm — is the most relevant measure to assess the long term fitness of plants and animals. Should more radiosensitive indicators, such as chromosome damage, be used instead? The paper presents some current ideas relating to this question, as well as cases where additional assessment may be required to ensure protection of the natural environment irrespective of the end-point issue. It is recognized that the base of scientific knowledge is not adequate to always make well founded management or mitigation decisions affecting radioactively contaminated habitats. Some areas of needed research are highlighted.

Periodically over the past 20 years, the question has arisen: Do we need specific radiation protection standards for plants and animals in the natural environment, in addition to the criteria already established for humans? This is a reasonable question, since many species are at least as radiosensitive as man and, in the context of environmental contamination with radionuclides, certain organisms may receive higher doses than humans. Furthermore, the human population has both the moral obligation and the practical necessity to protect the other species with which it shares its limited biosphere. The quality of human existence is directly or indirectly dependent on the health and vitality of the environment, and thus of most species of plants and animals.
PROTECTING OTHER SPECIES

Protecting other species implies the maintenance of conditions that permit normal functioning of populations of those species, consistent with the limitations related to environmental factors of a particular locale, such as temperature, moisture, light and nutrient availability. In the context of ionizing radiation, this means allowing routine releases and existing contamination only to the extent that radiation doses do not, alone or in combination with other agents or stress, impair such normal functioning of exposed populations.

For a fruitful examination of the question concerning the need to provide specific criteria for the protection of other species, some boundary conditions have to be specified. First and foremost, it is necessary to focus on specification of the most relevant biological damage end-points. Secondly, it is necessary to deal with the most likely and relevant mode of radiation delivery, for example chronic versus acute doses and high level versus low level exposures.

BIOLOGICAL DAMAGE END-POINTS

It has been the prevailing view for at least 20 years that most non-human populations are primarily valued and appreciated at the population level rather than the individual level. For example, the forest is the relevant unit for measuring environmental quality, rather than individual trees. Individuals in any healthy, robust population are continually meeting tragic fates, but this is offset by the constant recruitment of new, healthy individuals. For the vast majority of populations of plants and animals, it is both practical and adequately conservative to protect the population.

For humans, of course, the protection of individuals is the prevailing philosophy. Our radiation protection guideline permitting an effective dose equivalent rate of 1 mSv/a for the general public is usually judged to provide adequate safety for individuals. At chronic effective dose rates below this level, the probability of a significant health effect is very low for an individual, perhaps of the order of <5 chances in 1000 of a fatal cancer or genetic defect in a lifetime, against perhaps 200 chances in 1000 without any exposure above that from natural background radiation. The philosophy of individual protection could and probably should be applied also to rare and endangered species and/or to those species which are long lived and have very low reproduction rates. One would generally expect such populations to be relatively sensitive to the effects of radiation, and the loss of a few individuals could in some cases impair the ability of the population to maintain itself.

For the vast majority of species that can be adequately protected at the population level, reproduction appears to be the most sensitive yet ecologically relevant end-point, although more debate of this point can be expected. On the basis of com-
prehensive surveys of the literature dealing with the effects of experimentally applied radiation to populations in natural environments, the ability of populations to survive seems most dependent on their ability to reproduce normally. Direct mortality may be more immediately important at higher dose levels, but it is generally believed that, in the long run, survival of the population can be impaired at the lowest dose rates through reductions in the reproduction rate.

At doses much lower than those required to impair reproduction, cellular genomic damage has been observed. Perhaps the most important of this type of damage is the breakage of DNA strands that produce chromosomal alterations. Chromosome damage can lead not only to cell death but also to permanent heritable changes in surviving cells. In somatic cells such changes can lead to carcinogenesis, and in germ cells to genetic mutations, which may affect the progeny of exposed individuals. These types of changes at the level of molecules and cells also occur spontaneously, and most spontaneous changes are qualitatively indistinguishable from radiogenic changes. Those chromosome aberrations occurring in germ cells which are stable and persist long enough to be passed on to the next generation have a high probability of being removed, sooner or later, from the gene pool. For example, a balanced translocation or inversion in a gamete can, after fertilization, result in an individual all of whose cells contain the translocation or inversion. These individuals are known as translocation or inversion heterozygotes, and since only half of their gametes are viable, they are semi-sterile. Even for smaller mutations, the vast majority tend to be harmful and thus they are likely to be removed from the gene pool through reduced reproductive fitness and natural selection. This may be a tragic event for the individual, but the net effect is to restore the pre-irradiation fitness of the population.

RELEVANT MODE OF IRRADIATION

Concerning the mode of radiation delivery to be considered, the normal situations to which this discussion would apply would be controlled, routine releases, and environmental contamination of aquatic and terrestrial environments. Acute releases, such as those which might result from accidents or use of nuclear weapons, will not likely be affected by protection criteria. Both routine releases and environmental contamination with long lived radionuclides produce chronic exposures. At the levels of exposure that might be regulated, we must be concerned with relatively low dose rates. For these reasons, this discussion should apply to chronic exposures at low dose rates. Furthermore, since reproduction is usually considered the principal endpoint of concern for the protection of populations, the appropriate metric should be chronic dose rates to the reproductive organs of plants and animals.
HISTORICAL CONSIDERATIONS

The first position on the need for radiation protection criteria for non-human populations was published by the International Commission on Radiological Protection (ICRP) [1]:

"Although the principal objective of radiation protection is the achievement and maintenance of appropriately safe conditions for activities involving human exposure, the level of safety required for the protection of all human individuals is thought likely to be adequate to protect other species, although not necessarily individual members of these species. The Commission therefore believes that if man is adequately protected then other living things are also likely to be sufficiently protected."

The ICRP position was examined in considerable detail by the IAEA [2]. In this study, the question posed was: If the effective dose equivalent rate of the most exposed human population (the critical group) is limited to 1 mSv/a, considering all environmental pathways, are populations of plants and animals in the same environment adequately protected? First, a comprehensive literature review was performed to determine the minimum chronic dose rates to reproductive tissues of a wide spectrum of aquatic and terrestrial organisms that would impair fecundity. This review focused heavily, but not exclusively, on populations in natural environments. Secondly, the dose rates to the reproductive tissues of representative plants and animals that might result from the same amount of environmental contamination that would produce 1 mSv/a to the critical group of humans were conservatively estimated. Finally, these dose rates were compared with those found in scientific studies to impair reproduction. The basic conclusion of the IAEA study [2] was:

"There is no convincing evidence from the scientific literature that chronic radiation dose rates below 1 mGy/day will harm plant or animal populations. It is highly probable that limitation of the exposure of the most exposed humans (the critical group), living on and receiving full sustenance from the local area, to 1 mSv/year, will lead to dose rates to plants and animals in the same area of less than 1 mGy/day. Therefore, specific radiation protection standards for non-human biota are not needed."

CONSERVATISMS AND CAVEATS

The IAEA conclusion appears to be generally conservative. On the one hand, under the prevailing ALARA ('as low as reasonably achievable') philosophy, it is rare that dose rates of members of the general public approach 1 mSv/a from con-

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1 Gy = 1 J/kg.
trolled releases or anthropogenic contamination. On the other hand, the dose rate calculations for plant and animal tissues were quite conservative. Most calculations probably overestimated the actual doses by one to two orders of magnitude. Finally, most releases and contamination are rather local and, ordinarily, only a very small fraction of the individuals of other populations would receive the higher dose rates.

Despite the conservative nature of the conclusions of the IAEA study, the report did provide some caveats and allusions to cases that should require a specific analysis. The first caveat referred to the existence of rare, endangered, or long lived species with very low reproduction rates. For such species, the philosophy of protecting the population might actually require the more stringent philosophy of individual protection. The second caveat acknowledged the possibility of strong synergistic effects between radiation and other agents or stresses. Little research on this point, particularly that dealing with chronic radiation exposure of natural populations stressed by other factors, has been conducted. The final caveat referred to those cases where exposed populations occupy areas that would not support human beings. Examples might include the ocean depths and local areas of high level contamination at nuclear installations.

RECENT WORK RELEVANT TO THE QUESTION

After completion of the IAEA study, an independent literature review on lower limits of radiosensitivity of a wide variety of organisms was carried out [3]. This review did not reveal any results that would alter the conclusions of the IAEA study. In addition to this, a former nuclear reactor cooling reservoir at the Savannah River Site, South Carolina, United States of America, provided a specific case history relevant to the question about protection criteria for other populations [4, 5]. This reservoir, Par Pond, contaminated with $^{137}$Cs at a level of about 1 MBq/m$^2$, was partially drained in 1991. Site specific data and simple models were used to estimate both human health and ecological risks for hypothetical human residents subsisting on the lake bed and for resident aquatic and terrestrial organisms. Whereas the potential maximum lifetime risk of fatal cancer for humans was about $3 \times 10^{-3}$ (over one order of magnitude above that requiring action under the US Environmental Protection Agency’s ‘Superfund’ Act), the risk to local biota was two to three orders of magnitude below the threshold for reproductive impairment. Actually, in this case, the criterion used for human health protection was more sensitive (by about three orders of magnitude) than the 1 mGy/d criterion for plant and animal populations proposed by the IAEA. Clearly, in this case, and likely in nearly all cases, human health risk, not ecological risk, is probably the limiting factor. Furthermore, no cases were found where radionuclide releases or contamination within allowable limits for human exposure have led to documented effects on other species or on environmental quality.
AN ALTERNATIVE TO PROTECTION CRITERIA FOR OTHER SPECIES

It could be expected that the development, implementation and enforcement of statutory criteria for the protection of other species would be complex, as well as costly. The task would be complicated by the need to establish critical species and dosimetric relationships for a very large number of aquatic and terrestrial habitats. The knowledge base on radionuclide transport parameters, tissue dosimetry and radiobiology is not adequate at present for developing site specific criteria intelligently. Thus, the attempt to do so could be met with various qualifications, exceptions and legal challenges.

Instead of developing standards and criteria for the protection of other species from environmental radioactivity, it would seem more beneficial to use resources to fill critical gaps in our knowledge. One of the more fundamental gaps is the best end-point to use when assessing the effects of radiation on populations. Although the case that reproduction is the key end-point is strong, it can be hypothesized that subcellular damage at lower dose rates, particularly if the dose is maintained for several generations, might lead to a higher equilibrium level of deleterious mutations, which in turn might cause a decrease in population fitness and competitiveness. Few studies have actually attempted to draw connections between molecular damage and population survival at dose rates that are too low to affect reproduction. With all their limitations regarding extrapolation to a population, cellular and molecular studies on plant and animal tissues may lead to a better understanding of this issue. Another area of needed research is that of combined effects of radiation and other stressors on natural populations. It is possible, for example, that radiation damage repair processes could be impaired by other environmental conditions or agents. The ability to accurately estimate the dose to critical biological tissues, especially from alpha and beta particles that may be surficial, could certainly be improved by more research. Finally, many habitats and species have never been experimentally irradiated.

It is possible that additional research could uncover surprises that would argue in favour of the need for specific criteria. Until that happens, however, it seems that we should embrace what is known about radionuclide transport and effects, and increase that knowledge base.

REFERENCES


PROTECTION OF THE NATURAL ENVIRONMENT AND THE NEED TO FORMULATE CRITERIA

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Abstract

PROTECTION OF THE NATURAL ENVIRONMENT AND THE NEED TO FORMULATE CRITERIA.

Protection of the environment from ionizing radiation is usually considered to be achieved if humans living in the same environment are adequately protected. The paper reviews the arguments for and against developing specific criteria for protecting species other than humans in the environment.

1. BIOLOGICAL EFFECTS OF IONIZING RADIATION ON HUMANS AND THE ENVIRONMENT

All living species exist in environments where they are exposed to radiation of natural origin. During the last 50 years this background has been changed on a global scale by fallout from nuclear weapons testing and by accidents in nuclear installations. In some areas this change of the natural background has been significant enough to damage ecosystems. In the long term, there may be adverse effects on ecosystems because of increased chronic exposure resulting from accidents and normal releases.

In principle, there is no difference on a cellular level between the possible effect of radiation on humans and that on other species. Nevertheless, there can be a difference in the protection philosophy because of ethical and other considerations.

2. PROTECTION OF HUMANS

A quantitative system exists for the protection of humans against ionizing radiation, namely the ICRP (International Commission on Radiological Protection) system of dose limitation: justification, optimization and dose limitation. The system focuses on biological effects on humans, taking into account economic and social factors. The objective is to minimize the level of risk to individuals and their offspring. The potential level of harm to populations as a whole by the use of collective dose is also taken into account.
During the last years the concept of the best available technology (BAT) has been introduced into many international conventions. It focuses not only on the effect on humans and because of this it is more widely applicable to environmental protection. The BAT concept has not yet been adequately defined and it is therefore not easy to implement it in a legal framework.

3. PROTECTION OF OTHER SPECIES

In practice, no quantitative criteria exist for the protection of organisms against the risks associated with increased exposure to ionizing radiation. Previously, protection of the environment was in many cases not even taken into account when nuclear installations were commissioned. In most cases, nature has not been affected to a significant degree by normal operations because of the high level of protection provided for humans. This is unfortunately not true for releases of toxic substances other than radionuclides.

On a qualitative basis it has been argued that nature is protected if humans are protected, i.e. humans act as a critical indicator species. This assumption has been widely supported but is still based on limited evidence. It is also assumed that humans are connected with the ecological system under consideration, i.e. that the area where exposure takes place has the maximum radiation level, and that the consumption habits of humans are in one way or another linked to the ecosystem studied.

In some cases a quantitative evaluation based on the increase of the radiation background in an affected area has been used to ensure that species other than humans are protected. It is then assumed, on a qualitative basis, that if the radiological background is only increased by a fraction — preferably by an amount that is within the natural variation — the habitat is not likely to be affected.

4. WHAT IS TO BE PROTECTED?

We can develop criteria for the protection of the natural environment according to our individual preference. However, it appears that a general consensus exists among decision makers regarding the following points:

— Species shall be protected from extinction;
— Mortality of individual organisms is acceptable, but adequate protection of large populations must be ensured;
— In some circumstances, smaller groups of organisms should be protected if they are of particular value.

Sometimes, humans assign an emotionally based value to an organism. None of us want to see unnecessary harm done to individuals such as elephants, gorillas
or big cats. Some of us would also assign to a pet animal a value which exceeds the usual one.

The United Nations Conference on Environment and Development (UNCED), held in Rio de Janeiro in June 1992, placed emphasis on biological diversity. It was declared that biodiversity is "the variability among living organisms from all sources, including terrestrial, marine and other aquatic ecosystems and the ecological complex of which they are part, including diversity within species, between species and of ecosystems". This definition of biodiversity implies that all organisms are to some degree important, not only the publicly 'interesting' species. Thus, the goal for protection of the natural environment should be to uphold biological diversity.

5. THE PRECAUTIONARY PRINCIPLE AND REASONABLENESS

Decision makers as well as the scientific community have argued that our knowledge of potentially adverse effects is incomplete and, accordingly, a precautionary approach should be applied to safety. If this approach were to be applied strictly, it could result, at the extreme, in the phasing out of most activities of humans. Therefore, measures taken to increase safety must be reasonable. A prerequisite is that operational safety criteria must be verifiable.

6. SYSTEM OF PROTECTION

A system for the protection of the environment could be based on a single criterion or on multiple criteria. It could also be based on direct evaluation of the impact on individual organisms or on considerations of the abiotic environment.

6.1. Abiotic environment

If the abiotic environment is 'protected', the ecosystems in this environment will also be protected. This approach is based on the belief that ecosystems have adapted to the natural background. Since the normal concentrations of radionuclides vary in time and space, a limited additional level of contamination (of the normal background, not an extreme background) can be tolerated by the ecosystem. This approach has the benefit of being easily monitored and can be utilized for a first evaluation of a potential detrimental effect caused by releases of toxic materials.
6.2. Humans as indicator species

Another approach is to assume that protection of humans does provide protection of the environment. In the environmental impact assessment it has to be ensured that evaluation of the critical indicator species (humans) takes into account all potential exposure pathways for other species.

6.3. Absolute protection

In order to protect all ecological systems, i.e. populations and communities in an area, an evaluation on an individual level can be carried out. The vast diversity of organisms in even a small area makes it extremely difficult, if not impossible, to identify the critical species/habitat. One way to evaluate the situation is to define a generic organism, such as a plant with radionuclide uptake similar to that of a range of grasses, trees, etc. This generic organism could be selected to represent different trophic levels if accumulation is taken into account.

In some situations, a species existing in the area could be identified and the impact evaluated, in much the same way as for humans, provided that a methodology is available to convert dose to risk. Since data on risk are sparse, it could be relevant to compare calculated doses with the effects of chronic exposure of plants and animals. In this context, it may be noted that dose rates to organisms of less than 0.4 Gy/a are not likely to cause harm to populations of animals and plants. However, in order to apply the precautionary principle, a lower value could be chosen, such as 1–100 mGy/a.¹

7. RECOMMENDATIONS

Up to now, the impact of ionizing radiation on the natural environment has been evaluated indirectly by assuming that, if humans are protected, other species will also be protected. Recently, it has been argued that the detrimental effect of ionizing radiation on biota should be addressed in its own right. However, scientific understanding in this field is limited, particularly with regard to the dose/risk relationship. Nevertheless, it might be worth while to develop quantitative criteria based on dose to individuals for the protection of the natural environment, using data on chronic exposure of plants and animals.

¹ 1 Gy = 1 J/kg.
THE PRECAUTIONARY PRINCIPLE
Where does it come from and where might it lead in the case of radioactive releases to the environment?

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Abstract

THE PRECAUTIONARY PRINCIPLE: WHERE DOES IT COME FROM AND WHERE MIGHT IT LEAD IN THE CASE OF RADIOACTIVE RELEASES TO THE ENVIRONMENT?

It is generally agreed that the origin of the precautionary principle is to be found in the 'Vorsorgeprinzip' in German environmental law. Germany introduced the principle into the preparatory process for the North Sea Ministerial Conferences, which resulted in its explicit articulation in the 1987 Second Ministerial Declaration on the North Sea. From there it found its way into the work of other international forums concerned with the protection of the marine environment. Most notably among these are the Oslo and Paris Commissions, the Meeting of Contracting Parties to the London Convention and the Governing Council of the United Nations Environment Programme. Since then, the principle has found its way into virtually every international treaty and policy instrument concerned with the protection of the environment, both at the global level and the regional level. Examples of such instruments are the Rio Declaration, the Convention on Biological Diversity, the United Nations Convention on Climate Change and the Maastricht Treaty on European Union. Since many, if not most, of these instruments also may be taken to have an implication for the regime applicable to discharges of radionuclides, the conclusion must be that the precautionary principle applies to such discharges. The main problem facing those involved in the development of regimes for the protection of the environment is what substantive measures and procedural instruments may be employed to further precautionary environmental policies. Questions which arise in the process of identifying the implications of the precautionary principle for environmental policy and law may be summarized as follows: (1) What is the intention of the precautionary principle? (How does it relate to the assimilative capacity approach? What are its implications for the relationship between policy and science, technology, economics and law? What is the purport of the varied wording (especially approach and principle) used to refer to the concept of precaution?) (2) Who are the primary addressees of the precautionary principle? (What are its implications for policy makers? What are its implications for environmental standard setting? What are its implications for scientific research?) (3) What are the legal implications of the precautionary principle? (What is its legal status? What are its implications for substantive and procedural aspects of law? What are its implications for the burden of proof? What is its relationship to international liability regimes?) Possible answers to the above mentioned questions are addressed in the paper and related to the regime for radionuclide releases.
INTRODUCTION

It is generally agreed that the origin of the precautionary principle is to be found in the ‘Vorsorgeprinzip’ in German environmental law [1, 2]. Germany introduced the principle into the preparatory process for the North Sea Ministerial Conferences, which resulted in its explicit articulation in the 1987 Ministerial Declaration on the North Sea [3]. From there it found its way into the work of other international forums concerned with the protection of the marine environment. Most notable among these are the Oslo Commission (OSCOM) and the Paris Commission (PARCOM)\(^1\), the Meeting of Contracting Parties to the 1972 Convention on the Prevention of Marine Pollution by Dumping of Wastes and other Matter (London Convention) [4] and the Governing Council of the United Nations Environment Programme (UNEP) [5].

The principle since has found its way into virtually every international treaty and policy instrument concerned with the protection of the environment, both at the global level and the regional level [6]. Examples of such instruments are the Rio Declaration [7], the Convention on Biological Diversity [8], the United Nations Convention on Climate Change [9] and the Maastricht Treaty on European Union [10]. However, the principle has not been included in the Convention on Nuclear Safety, adopted on 20 September 1994 [11].

The initial inclusion of the precautionary principle in legally non-binding documents, such as ministerial declarations and recommendations of international organizations, meant that the principle as such, on the basis of those documents, could not be considered to be a principle of international environmental law. This is not to say that the precautionary principle at that stage was without legal significance. It may, for instance, already at that stage have been relevant in determining the legitimate expectations which the signatories of those documents might have had from each other with respect to, for example, the dumping of radioactive waste at sea.

In law, the subsequent inclusion of the principle in legally binding documents means that, at least for the parties to those treaties, the principle is legally binding. In addition, it has been contended that there is now sufficient evidence to support the argument that the precautionary principle is a principle of customary international law [12]. This would mean that in determining environmental policies, including those involving the use of radioactive materials, States on all occasions would be bound to implement the precautionary principle. However, as is the case with other principles of international law, the question remains what States should do or refrain from doing in order to implement the precautionary principle.\(^2\)

\(^{1}\) See OSCOM Recommendation 89/1 and PARCOM Recommendations 89/1 and 89/2, Ref. [3], pp. 119, 152, 153.

\(^{2}\) On this point, see Ref. [13], p. 213, making the point that similar uncertainties are relevant to the application of the principle of self-determination.
As has been well documented, the precautionary principle has been worded in various terms (see, for example, Ref. [14]). These terms vary from the weak formulation contained in the Bergen Declaration\textsuperscript{3} to a reversal of the normal burden of proof in the Prior Justification Procedure adopted by the Oslo Commission.\textsuperscript{4} The variety of terms, and especially those allowing States a wide margin of discretion\textsuperscript{5}, used in the formulation of the principle, undoubtedly have contributed to the uncertainty as to the precise meaning of the precautionary principle. However, on the basis of the available wordings of the precautionary principle, conclusions can be drawn as to what it intends to achieve. This is the subject which will be addressed in this paper.

The paper is structured as follows. First, some terminological aspects will be addressed. Secondly, the meaning of the precautionary concept will be addressed. Thirdly, the question as to who the addressees of the precautionary principle are will be discussed. Fourthly, some of the possible implications for the development of policy and law related to radioactive materials will be addressed.

**TERMINOLOGY**

The instruments which address the need for precautionary environmental policies do not only use a variety of terms to indicate what precaution may entail, they also refer to the concept of precaution itself in a variety of terms. Some of the terms

\textsuperscript{3} Paragraph 7 of the Bergen Declaration provides as follows: "In order to achieve sustainable development, policies must be based on the precautionary principle. Environmental measures must anticipate, prevent and attack the causes of environmental degradation. Where there are threats of serious or irreversible damage, lack of full scientific certainty should not be used as a reason for postponing measures to prevent environmental damage." See Ref. [15].

\textsuperscript{4} In OSCOM Decision 89/1 on the reduction and cessation of dumping of industrial wastes at sea the contracting parties agree that "... the dumping of industrial wastes in the North Sea shall cease by 31 December 1989, and in other parts of Convention waters by 31 December 1995, except for inert materials of natural origin, and except for those industrial wastes for which it can be shown to the Commission through the Prior Justification Procedure (PJP) both that there are no practical alternatives on land and that the materials cause no harm to the environment." This procedure places the burden of proof on the applicant State; it is to demonstrate that no harm will be caused to the marine environment. See Ref. [3], p. 119.

\textsuperscript{5} See, for example, Principle 15 of the Rio Declaration, which provides that "In order to protect the environment, the precautionary approach shall be widely applied by States according to their capabilities. Where there are threats of serious or irreversible damage, lack of full scientific certainty shall not be used as a reason for postponing cost-effective measures to prevent environmental degradation." (Emphasis added.)
used are ‘precautionary principle’, ‘precautionary approach’ and ‘precautionary measures’ or ‘actions’. As I have contended elsewhere (Ref. [16]), the terms mentioned in fact refer to different aspects of the same concept and no substantial differences in content are intended and, if differences of substance were intended, these have not been pursued systematically.

The intention here is not to suggest that differences in terminology are irrelevant. They are relevant, especially where a concept is concerned which seeks to guide the development of policy and law in a given field. It is with this role in mind that I draw attention to the following terminological aspects.

First, I suggest that a distinction be made between the terms ‘principle’ and ‘approach’. The term ‘principle’, according to the Oxford English Dictionary, means a “source of action”; “a general law or rule adopted or professed as a guide to action” [17]. Thus, “principle” in our case implies a general rule adopted as a guide for developing international environmental policy. According to the same dictionary, the term “approach” refers to “a way of considering or handling something, especially a problem” [18], in our case a way of considering or handling environmental problems.

Secondly, it is useful to define the term ‘measures’. ‘Measures’ might be understood as the course of action to be followed as a result of applying a certain approach to a specific problem, e.g. a decision to terminate incineration of wastes at sea or a decision prescribing that industrial sector y apply the best available technology x by a certain date.

Given these three definitions, the term ‘policy’ could then be used to refer to the combination of principle, approach and measures. Each of the three notions — principle, approach, measures — could be either legally binding or legally non-binding, depending on the status of the instrument in which they are contained or on their status under customary international law. The latter requires one additional remark; the more detailed the nature of a prescription, the more unlikely it is to attain the status of customary international law. As a result, it is unlikely that a detailed measure requiring, for example, the application of certain industrial processes, would be legally binding unless it were contained in an instrument which, according to its form, is binding in law; examples of such instruments are treaties and binding decisions of international organizations.

THE INTENTION OF THE PRECAUTIONARY CONCEPT

As I have submitted elsewhere (see Ref. [16]), the precautionary concept can be characterized as addressing the manner in which policy makers, for purposes of protecting the environment, apply science, technology and economics. The ultimate aim of precautionary policies is to introduce production and consumption patterns which will enable both present and future generations to meet their sustainable needs.
The distinctive feature of the precautionary concept is not that it dictates specific regulatory measures: many different types of measures can be used to implement it. The distinctive characteristic is the way in which, and the time at which, the measures are to be adopted.

The concept assumes that science does not always provide the insights needed to protect the environment effectively, and that undesirable effects may result if measures are taken only when science does provide such insights. Furthermore, it assumes that scarce financial resources may be allocated inefficiently if action is taken only after scientific certainty as to detrimental effects has been provided, particularly when alternative technologies and/or products are available. In fact, the precautionary concept rejects the so-called assimilative capacity approach to environmental policy (Ref. [16], pp. 305 and 306).

The different documents which refer to the precautionary concept provide an indication of the considerations which play a role in attaining the implementation of precaution. Common elements in the texts are that clean methods of production and clean products shall be used, that environmental impact assessments shall be applied, and the need for economic rationale. The first element implies a shift of focus away from trying to determine the level of pollution which the environment can assimilate to the development of research methods and technologies which will eliminate or at least reduce the input of pollutants into the environment — a shift away from policies based on 'dilute and disperse' towards policies based on 'minimization and containment' of substances harmful to the environment. Both technologies resulting in alternative production processes and technologies resulting in alternative products would be included. The second element, environmental impact assessment, likewise implies that policy is not to rely upon the determination of detrimental environmental effects after an activity has taken place and the harm materialized, but instead to attempt to ascertain any possible negative effects prior to the activity taking place and especially prior to the harm having materialized and then to take positive

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6 This notion, for example, is evidenced by the note incorporated in the Decision of the Oslo Commission to prohibit the incineration of wastes at sea (OSCOM Decision 90/2, 23 June 1990, Ref. [3], p. 126). The note provides that "This Decision takes into account concerns expressed about potential implications of incineration at sea, but is not based on evidence that harmful effects have been observed. The Decision is based on the fact that Contracting Parties are in the process of developing methods for the reduction of wastes which result from production processes and that Contracting Parties have developed, or intend to develop, land-based alternatives for recycling and destruction. In most cases of avoidance and recycling, land-based treatment may be cheaper than incineration at sea."

7 See, in particular, D. Fleming, "The economics of taking care: An evaluation of the precautionary principle"; J. Gray, "Integrating the precautionary principle into scientific methods of decision-making"; and L.D. Mee, "Scientific methods and the precautionary principle", in Ref. [6], Chapters 9, 8 and 7, respectively.
action to avoid any possible negative effects. The third element, the need for economic rationale within a precautionary policy, is a concept rather different from traditional cost/benefit analysis; it implies that long term and comprehensive economic accounting methods are to be used which attempt to internalize the cost of possible damage to the environment as well as any related social costs.

On the basis of the above, the precautionary principle and the concomitant approach can be defined as follows:

The precautionary principle means that in order to ensure the protection and preservation of the environment and to attain sustainable development, lack of scientific certainty shall not be used as a reason for deferring measures to enhance the quality of the environment. In order to achieve this, a precautionary approach shall be applied.

The precautionary approach entails the following:

1. Clean production methods, best available technology and best environmental practices must be applied;
2. Comprehensive methods of environmental and economic assessment must be used in deciding upon methods to enhance the quality of the environment;
3. Research, particularly scientific and economic research, that contributes to a better understanding of long term options available must be stimulated; and
4. Legal, administrative and technical procedures that facilitate the implementation of this approach must be applied and, where not available, developed.

From a legal point of view, the most important facet of the principle is that positive action to protect the environment may be required before scientific proof of harm has been provided. The new element is the timing of, rather than the need for, remedial action. Preventive action once damage has been determined is a long standing requirement of international environmental law.\(^8\) In fact, the essence of the precautionary concept, the precautionary principle, is that, once a risk has been identified, the lack of scientific proof of cause and effect shall not be used as a reason for not taking action to protect the environment.

This suggests that the threshold of significant risk has become easier to cross, which from a legal perspective would imply that, once a prima facie case is made that a risk exists, scientific uncertainty works against the potential polluter rather than, as in the past, in his/her favour. The way in which this will be applied to different sectors and to different types of risk, i.e. the measures required to implement the concept of precaution, is still in the process of being developed. However,

\(^8\) Customary international law includes the duty to take preventive action and can be traced back to the Trail Smelter arbitration (Am. J. Int. Law 33 (1939) 182; 35 (1941) 684). See also Ref. [19].
examples of precautionary measures are now becoming more widely available. Examples are the reverse listing system included in Annex II of the 1992 Convention on the Protection of the Environment of the North-East Atlantic (OSPAR Convention) and the introduction in several conventions of the requirement to apply clean technology. In addition, precaution may also entail outright bans of an activity. An example of such a measure is the decision taken, in 1993, by the parties to the London Convention to ban the dumping of radioactive waste at sea (see Ref. [21]).

THE ADDRESSEES OF THE PRECAUTIONARY PRINCIPLE

The primary addressee of the precautionary principle is the policy maker. She or he must address the question as to the type of applied research required — be it economic, technical, scientific, legal or otherwise — and, more importantly, the question as to the degree of risk of harm which should be avoided and at what cost. Answering the latter question is especially difficult, given that the results of scientific research are generally, and rightly so, presented in terms of probabilities.

The precautionary principle does not insist that all risk of harm be avoided. Rather, it requires that society be willing to accept higher costs now in order to avoid the possibility of environmental degradation in future. In legal terms, it prescribes the application of an enhanced duty of care. It thus does not prescribe that all balancing be banned [22], but that environmental considerations be balanced more fully against considerations of an economic nature and that the development of environmentally friendly technologies be stimulated and, where such technologies are available, that they be applied. Neither the precautionary principle nor other principles of international environmental law, or principles of international law, can make such relative determinations. The challenge of making the 'right' decision inevitably must be faced by the policy maker.

Although the primary addressee of the precautionary principle is the policy maker, the principle is not irrelevant for scientists, economists and technologists (see Footnote 7). They may have to alter their methods of research in order to enable more comprehensive assessments of policy options to be made. Thus, instead of

9 22 September 1992. The text of the Convention and its Annexes are reproduced in Ref. [20]. Annex I relates to the prevention and elimination of pollution by dumping or incineration.

10 See, for example, article 2(3)(b) of the OSPAR Convention; article 3(3) of the Convention on the Protection of the Marine Environment of the Baltic Sea Area, Helsinki, 9 April 1992, Yearb. Int. Environ. Law 3 (1992), on the disk accompanying the Yearbook; and articles 3(1)(c) and 3(1)(f) of the Convention on the Protection and Use of Transboundary Watercourses and International Lakes, Helsinki, 17 March 1992, Int. Legal Mater. 31 (1992) 1312.
focusing on the assimilative capacity of the marine environment for a given substance and on trying to determine the cause of environmental deterioration after it has been found, marine biologists may have to concentrate on the determination of natural background levels of substances in the marine environment and on seeking possible effects of human activities in the environment. Economists will have to move away from traditional cost/benefit analysis and find ways of incorporating the long term social costs of human activities. Instead of developing more sophisticated end-of-pipeline technologies, technologists would have to concentrate on the development of no waste or, at least, low waste production processes.

As for lawyers and policy scientists, the precautionary principle should also be taken to have consequences for the focus of their work. In the light of the objectives of the principle, they should aim at the development of procedures which serve to institutionalize caution. In international law, for example, it is well known that many decision making procedures result in the lowest common denominator being the determinant of the decisions taken, instead of resulting in decisions which ultimately move those States which represent the lowest common denominator closer to those States which are able and willing to implement a higher degree of caution. This latter aspect is especially important in a world where vast differences exist in the development standard of States and where the environmental effects of activities may materialize far beyond the boundaries of any one State. These considerations have consequences on two fronts. First, decision making procedures which foster the interests of those States representing the lowest common denominator should be amended. Secondly, provisions for the transfer of funds and know-how to developing countries should be part and parcel of decisions taken (see also Ref. [23]).

Examples of decision making procedures which institutionalize caution are now available in the form of the Prior Justification adopted by the Oslo Commission (see Footnote 4), the waste assessment framework and the prior report procedure adopted within the framework of the London Convention\textsuperscript{11}. In addition, the Convention on Biological Diversity and the Convention on Climate Change are examples of instruments which link implementation by developing States parties to the transfer of funds and know-how by developed States.

**IMPLICATIONS OF THE PRECAUTIONARY PRINCIPLE FOR THE RELEASE OF RADIOACTIVE MATERIALS INTO THE ENVIRONMENT**

The precautionary principle in essence does not raise special considerations for the release of radioactive materials into the environment. The considerations which may apply to the release of radioactive materials by virtue of the precautionary prin-

\textsuperscript{11} For a critical assessment of these procedures with respect to the dumping of radioactive wastes at sea, see Ref. [25].
ciple arise as a result of the nature of the substance at stake and would apply equally to any other substance which may be released into the environment and which may have similar long term and far reaching consequences. This is an important consideration because, traditionally, activities involving the use of radioactive materials have remained outside the scope of ‘normal’ environmental law. Radioactive wastes, for example, are not covered by the Basel Convention on the Control of Transboundary Movements of Hazardous Wastes and their Disposal [27]\(^\text{12}\); with the exception of dumping at sea [24] and disposal on Antarctica [28], radioactive wastes have not been made the object of multilateral legally binding regimes, nor do Chapters 22 or 36 of Agenda 21 call on States to develop such regimes.\(^\text{13}\) Moreover, the 1994 Convention on Nuclear Safety does not apply to sites for the disposal of radioactive wastes [29]. A separate convention is being developed on this topic.\(^\text{14}\)

International regimes concerned with radioactive material traditionally have focused on stimulating the development of peaceful uses of that material and on preventing its proliferation for the development of nuclear weapons — the so-called safeguards regulations.\(^\text{15}\) More recently, after the Chernobyl accident, multilateral legally binding instruments have been developed. These instruments address the notification of nuclear accidents [32], assistance in case of nuclear accidents [33], and the safety of civilian nuclear power plants (for a critical review of the IAEA Convention on Nuclear Safety, see Ref. [34]). In addition, instruments have been available which address civil liability for damage as a result of nuclear accidents [35]. With the exception of the Convention on Nuclear Safety, these instruments are concerned with the aftermath of nuclear incidents and not with their prevention. Such an approach is out of step with the precautionary principle.

\(^{12}\) Radioactive wastes are covered by the Code of Practice on the International Transboundary Movement of Radioactive Waste, a legally non-binding instrument, adopted by the IAEA General Conference on 21 September 1990. For the text of the Code, see IAEA document GC(34)/920.

\(^{13}\) For the documents adopted at the United Nations Conference on Environment and Development (UNCED), see United Nations Doc. A/Conf.151/26 (Vols I–V), 12 August 1992. Chapter 36 calls on States to conclude the ongoing negotiations on a nuclear safety convention in the framework of the IAEA (para. 39.6(b)), while Chapter 22 of Agenda 21 only refers to the desirability of concluding a legally binding instrument in the case of the transportation of radioactive waste (para. 22.5.(a)).

\(^{14}\) Convention on Nuclear Safety, ninth preambular paragraph.

\(^{15}\) The aims of the IAEA as expressed in article 2 of the Statute reflect this approach: “The Agency shall seek to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world. It shall insure, so far as it is able, that assistance provided by it or at its request or under its supervision or control is not used in such a way as to further any military purpose.” For further information see Refs [30, 31].
The lack of evidence of concern for the use of radioactive materials in legally binding international regimes is all the more noteworthy because of the inevitable transnational implications — both in physical space and over time — of incidents that may occur as a result of the use of these materials (see, for example, Ref. [36]). Firstly, an accident involving radioactive materials is likely to have effects beyond the territory of the State where it occurs. Secondly, the risks associated with radioactive materials are likely to transcend the lifetime of present States. These two elements imply that considerations of intergenerational equity [37] are involved and that the traditional concept of State sovereignty which emphasizes the discretion of States does not offer the appropriate basis for regulating the use of radioactive materials. A more functional approach to State sovereignty which emphasizes the duties of States for the benefit of humankind is required [38].

Although it is now, and has been for a long time, generally accepted that States must prevent activities within their jurisdiction from causing transboundary harm or harm to the global commons\textsuperscript{16}, this notion is not sufficiently reflected in the international instruments related to the use of radioactive materials. This is because, in essence, these instruments, with the exception of the Antarctic Treaty and the London Convention, leave the regulation of activities involving radioactive materials to individual States and provide only marginal instruments of control to the international community [34]. In addition, the concept of inter-generational equity implies that States owe a duty of care towards present and future generations within and beyond their territory. As Handl has pointed out (Ref. [19], p. 32), where international environmental law is concerned the notion that sovereignty obliges remains far from having been implemented. This contention applies especially to the use of radioactive materials.

It is known that radioactive materials may have serious detrimental effects over especially extended periods of time, yet international control mechanisms remain scant. Although doubts may remain as to the precise consequences of the release of radioactive materials, the precautionary principle condemns the use of such arguments to postpone regulation.

If precautionary policies were to be implemented, this would require long term assessment of the use of nuclear materials from both an environmental perspective and a socio-economic perspective. For example, with respect to the use of nuclear energy this would entail incorporating into the assessment the subsidies which have been made available, for example, for the development of nuclear energy projects, the long term social cost of the use of radioactive materials and the effects of alternative sources of energy. The latter implies that the use of nuclear energy cannot be considered in isolation; it will have to be assessed against the use of other materials.

Such an approach is not reflected in the present organization at the international and regional levels of decision making, where separate forums, such as the IAEA and Euratom, have responsibilities for the development of the use of nuclear materials.

Applying a precautionary policy to the use of nuclear materials, from a substantive perspective, would, for example, imply that strict safety standards are to be applied; that wastes are to be minimized and, in principle, to be treated at the source; and that, where technologies involving the use of nuclear materials are exported, it should be required that the concomitant safety and waste management technologies are also exported. In this respect, the lack of provisions encouraging the transfer of financial means and technology in the Convention on Nuclear Safety is noteworthy. Consideration should be given to imposing on the exporter and on the exporting State a responsibility for the management of any radioactive waste generated as a result of such export.

From a legal perspective, the precautionary principle should also be taken to have consequences for the decision making processes related to the use of radioactive materials. The conduct of environmental impact assessments of at least a transboundary, but preferably a wider, geographical scope should be required prior to the establishment of any installation involving radioactive materials. Interested parties from outside the State in which such an installation is to be established should be able to participate in such assessments. The Convention on Nuclear Safety only requires that the State conduct an internal environmental impact assessment and that it consult and inform other parties which are likely to be affected by a proposed nuclear installation. 17 The Convention does not provide that the views presented should be taken into account nor is there a provision requiring the conduct of transboundary environmental impact assessments. This is in contrast to the ECE (Economic Commission for Europe) Convention on Environmental Impact Assessment in a Transboundary Context, which requires that, in the region covered, a wide variety of nuclear installations be the subject of such a procedure [39].

The fact that future generations may suffer from the effects of releases of radioactive materials imposes a special duty of care on the present generation. Decision making procedures should provide for the representation of this interest. The latter may be attained through the appointment of ombudspersons by, for example, the United Nations (see Ref. [37], pp. 124–126). The object of these procedures should be to guarantee to future generations at least the level of protection that the present generation deems adequate for the protection of its own interests. In addition, provision should be made for ensuring that future generations have access to information regarding the disposal of radioactive waste. This also implies that radioactive waste should not be dispersed into the environment if the waste is beyond any further measures of control that future generations may wish to introduce.

17 Convention on Nuclear Safety, Article 17(iv).
In order to ensure the implementation of the above mentioned substantive and procedural standards, international supervision is essential. Review procedures enabling the international community at large to participate in the assessment of the implementation of relevant undertakings should be developed. The review procedure included in the Convention on Nuclear Safety does not compare favourably with the review procedures adopted under international environmental treaties (see Ref. [34]), as does, for example, the non-compliance procedure contained in the Montreal Protocol. Any procedure adopted should be transparent and should allow for widespread participation, also of non-governmental organizations. The review process contained in the Convention on Nuclear Safety does not meet these standards. As such, it is confidential, and States themselves may decide which information they wish to regard as confidential, which probably means that it is at the discretion of individual States to determine which information they submit to the review process. Moreover, the fact that consensus among the contracting parties is required, even for the participation of intergovernmental organizations in the review meetings, is not conducive to widespread participation. Nor is the very limited role envisaged for the IAEA Secretariat likely to contribute to thorough review (see Ref. [34]). In fact, the review procedure contained in the Convention on Nuclear Safety is dependent solely on the input from the Member States — this again as opposed to review procedures contained in other environmental conventions.

CONCLUSIONS

The international regime for regulating the use of radioactive materials is far from having implemented the precautionary principle. It leaves too much to the discretion of individual States and, in fact, diverges significantly from the standards developed in international environmental law. This applies where both substantive standards and procedural safeguards are concerned.

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19 Convention on Nuclear Safety, Article 27(3).

20 Convention on Nuclear Safety, Article 27(1).

21 Convention on Nuclear Safety, Article 24(2).

22 The non-compliance procedure adopted under the Montreal Protocol, for example, also enables the Secretariat to play an active role. The Secretariat is not restricted in the sources from which it may obtain its information (non-compliance procedure, paras 3 and 7(b)).
A first requirement which needs to be incorporated into the regime for the use of radioactive materials is that scientific uncertainty is not an argument for the postponement of regulation. The question which may arise at this stage is: "but what is one to regulate for, if one does not know what the consequences may be of a given activity?". The answer which the precautionary principle gives to that query is that regulation entails not only the adoption of substantive standards but also procedural safeguards. Such procedures become particularly important when uncertainties as regards cause and effect are involved. They are to ensure that all interests at stake are entitled to effectively participate in the decision making process and that decision making is not shrouded by a veil of secrecy. It is exactly on this point that both national and international decision making procedures, especially with regard to the use of radioactive material, are deficient. Furthermore, and equally important, decision making procedures should be adopted at the international level; this would shift the burden of proving beyond reasonable doubt that activities involving the use of radioactive materials are safe to the State wishing to engage in, or to approve, such an activity. As a first step, such procedures could be incorporated into the Convention on Nuclear Safety and into the Convention on Nuclear Waste, which is currently being negotiated.

REFERENCES


[26] Convention on Biological Diversity, Article 20(2) and (4), and Climate Change Convention, Article 4(3) and (7).


THE PRECAUTIONARY PRINCIPLE IN A MARINE CONTEXT

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Abstract

THE PRECAUTIONARY PRINCIPLE IN A MARINE CONTEXT.

The precautionary principle has been adopted by a number of international conventions and yet is only loosely formulated. This poses considerable interpretational problems for those with responsibility for managing the marine environment. These problems are discussed in the paper. In particular, the extreme approach by the 'Green Movement', which insists that only harmless substances can be discharged to the sea, is examined. New scientifically based approaches to management are introduced, such as hazard and risk assessment and feedback monitoring. It is argued that adequate protection of the marine environment can be achieved by incorporating such new ideas in management strategies.

1. INTRODUCTION

At the Second International Conference on the North Sea in London the Ministerial Declaration agreed to: "accept the principle of safeguarding the marine ecosystem of the North Sea by reducing polluting emissions of substances that are persistent, toxic and liable to bioaccumulate at source by the use of the best available technology and other appropriate measures. This applies especially when there is reason to assume that certain damage or harmful effects on the living resources of the sea are likely to be caused by such substances, even when there is no scientific evidence to prove a causal link between emissions and effects (the 'principle of precautionary action')".

Acceptance of this principle poses a number of fundamental problems [1]. Firstly, the terms persistent, toxic and bioaccumulatable are not defined. If one adopts the most extreme approach, all elements should be banned from discharge since they are persistent. Likewise, does toxic mean toxic to all marine species, and at what concentration or dose is a substance defined as 'toxic', since all chemicals will become toxic at sufficiently high levels? In fact, aspirin has the same toxicity to man as DDT. Yet DDT unlike aspirin is bioaccumulatable, and lethal doses can accumulate [2]. Some chemicals bioaccumulate naturally and yet cause no stress symptoms in the accumulating organism nor do they present health risks for other
species consuming them. Clearly, it is the combination of toxicity, persistence and bioaccumulatability that needs to be considered.

A second major criticism of the precautionary principle is that suspicion of effects rather than scientific evidence is regarded as being sufficient to introduce discharge controls. The precautionary principle can be invoked to prevent discharges by simply arguing that at some future date a given chemical is likely to have an effect and that discharges to the sea therefore should be banned. Since introduction of most substances into the marine environment will cause at least local disturbances and since effect is not defined, this argument can be and is being used by the ‘Green Movement’ in relation to most sources of direct inputs into the marine environment. Yet, as I will argue later, if science is used within a proper management framework, then objective criteria rather than subjective ones can be used to determine the location and the quantity of substances that can be discharged safely.

In cases where little is known about the chemical that is planned to be discharged or where the biogeochemical cycle and the risks associated with the chemical in the environment are poorly understood, a precautionary approach does have a major role to play. Likewise, it can be applied in environmental management, since it would argue against discharges at all times when concentrations are approaching environmental quality standards and/or critical loads.

Whilst it may seem logical, as is stated in the precautionary principle, that one should employ the best available technology to minimize discharges of toxic chemicals in order to protect the environment, there are problems with adopting this approach. In some circumstances the use of the best available technology may prove to be overprotective (e.g. a small discharge of a substance with limited persistence and toxicity to a large receiving environment); in other situations it may prove to be totally inadequate (e.g. discharge of a highly persistent and toxic substance to a small receiving environment, or discharge of such a substance in smaller amounts but from several different sources to the same area).

The best available technology approach has been adapted to include economic considerations. Britain argues strongly for using the best available technology which does not entail excessive economic costs (BATNEEC) (see p. 138 of Ref. [3]). However, whether or not the adoption of the best available technology should include an assessment of economic costs and/or the consequences for other environmental sectors is a controversial and much debated point.

The sea does have a capacity to cope with waste material and there is very much more sea than land or fresh water; thus, the sea must remain an option for disposal of man’s waste. If one prevents disposal at sea, then the remaining options are disposal on land or in fresh water. Cost–benefit and other relevant analyses assessing disposal options must consider all environmental compartments.

Another argument used in favour of a precautionary approach is that there should be a “reversal of the burden of proof” [4]. Most present legislation on waste discharge requires that scientists prove that there is an effect of a harmful substance
before regulatory measures are applied. This is clearly highly unsatisfactory and not in tune with a precautionary approach. The problem is that an extreme version of the reversal of the burden of proof is now being strongly suggested by the Green Movement, namely that only harmless substances should be discharged. As Stebbing has pointed out [5], the notion is flawed since harmlessness cannot be disproved because no finite number of observations of harmlessness can eliminate the possibility that there will be effects somewhere at some time. As mentioned above, any introduction of a chemical or material will lead to some effect on the status quo of the environment. Thus, compliance of a discharge with certain standards has to be assessed in terms of acceptable effects. There is therefore a need to ensure that the best possible techniques are available for assessing whether or not there are effects on biological systems and to relate these effects to causes.

2. THE ASSIMILATIVE CAPACITY OF THE SEA

Before the precautionary principle was formulated, the most widely accepted view of waste disposal was that the environment had a capacity to cope with discharges of wastes without suffering any deleterious effects on their biological systems. This ability to cope was known as the assimilative capacity [6] or as the environmental capacity [7, 8] of a given area. For the marine domain, the definition of environmental capacity, proposed by the United Nations organization GESAMP (Joint Group of Experts on Scientific Aspects of Marine Environmental Protection), is "a property of the environment, defined as its ability to accommodate a particular activity, or rate of activity, without unacceptable impact" [7]. The concept is based on the assumptions that a certain level of some contaminants may not produce any undesirable effect on the marine environment and its various uses; that each environment has a finite capacity to accommodate some wastes without unacceptable consequences; and that such capacity can be quantified, apportioned to a certain activity, and utilized.

The problems with these assumptions are that there is no definition of what is an undesirable effect nor how it will be measured, or how the capacity of a given area can be predicted or measured. The practice usually adopted is that a concession for a certain discharge is given and then monitoring is done to see that the assimilative capacity is not exceeded with the result that deleterious effects are observed on biological systems. Too often in the past, damage has occurred, and then it is too late to do anything about it. Earll [4] gives as an example mercury in Liverpool Bay, which has been shown to have negative effects on biological systems; yet there is no hope of removing the mercury that was permitted to be discharged.

The key problem with the environmental capacity concept is that the management and legislative systems connected with the environmental capacity have not used all scientific data and knowledge available. Instead, decisions have often been
made with "application factors and other arbitrary rules of thumb" [5] that lead to pollution problems that are "a legacy of a weak scientific basis for consenting procedures and insensitive techniques, or imprecise toxicological data, while the regulators have been obliged not to deny industry and society its traditional access to the environment to dispose of wastes". The failing of management and not science in relation to environmental capacity is also acknowledged by Earll [4]: "In practice, science and its processes have often been totally ignored when development decisions have been taken". Yet science has been mistakenly attacked by Greenpeace as "failing the environment" [9]. It is not science that has failed in the use of ideas such as the precautionary principle and the environmental capacity but the application of science to management issues by managers and legislators.

3. LINKING SCIENCE TO MANAGEMENT

What is needed is a way to combine the spirit of the precautionary principle with the latest scientific findings to give better management. If such a way forward can be found, then it will not be necessary to take the extreme 'green view' that nothing must be discharged into the oceans until we know everything about all chemicals.

The United Nations Environment Programme (UNEP) has developed the Montreal Guidelines [10], which are a set of recommendations for governments to assist them in the development of legislation for the protection of the marine environment against pollution from land-based sources. The Guidelines have taken common elements and principles from existing agreements, such as the Oslo, Paris and Helsinki Conventions, the Athens protocol and the United Nations Convention on the Law of the Sea. In Annex 1 to the Guidelines, there is a section on strategies for protecting, preserving and enhancing the quality of the marine environment. Firstly, there are control strategies such as those based on marine quality standards, emission standards and environmental planning. The Guidelines on marine quality consider standards based on water, sediment, fish or their tissues, health or community composition, but they do not give a framework for deciding how the set of standards should be derived. Dilution and loads are also mentioned, but other important components of hazard assessment are not considered.

The Guidelines suggest that technology standards relating to emission standards should be applied on a sector by sector basis. A distinction is drawn between best practicable technology, which takes into account costs, and best available technology, which does not, but which it is suggested should apply to the most noxious substances.

In relation to planning strategies it is stated that certain activities are incompatible with values or uses of the environment; that uses of the environment will have different quality standards and that the standards should be defined for the specific
uses; that environmental impact assessments are necessary for any activity affecting the environment; and that regional management is needed for such areas as the coastal zone, drainage basins and especially sensitive areas.

The Guidelines give a useful check-list of factors that have to be considered, but they do not provide a logical scientific framework for protection and management of the marine environment. The disposal of radioactive wastes in the marine environment has been based soundly on a framework for risk assessment and risk management. This is not the case for most chemicals. Thus, a framework has been devised which we hope will lead to better management.

Gray et al. [11] have suggested that in the marine environment there are two different basic strategies, depending on whether the discharge is a point source and/or the effect of local disturbance or whether the discharge is diffuse and/or the effect of chronic disturbance. For diffuse sources and/or chronic disturbance it is suggested that surveillance should be undertaken, comparing the levels of contaminants or the disturbances with control sites that are not thought to be contaminated. If the level of one or more contaminants or the state of a physically impacted site are shown to be different from controls, then one proceeds to a hazard assessment. Such surveillance studies should take fully into account the new Beyond BACI techniques [12, 13] for sampling design and be based on power analyses of the ability to detect change [14, 15]. If one tests the means of two sets of data and finds no statistically significant difference this may mean that there is no difference or, alternatively, that the sampling methods were so poor that it would not be possible to detect even a large difference. Power analyses take into account variance and number of samples and allow one to simulate how many samples for a given variance would be needed to detect a significant difference (see Ref. [16] for a thorough description of methods). Power analyses are slowly being incorporated into environmental impact statements and monitoring programmes.

As an example, a baseline study was done in Denmark, using various biological variables to measure the condition of an eelgrass bed. The environmental authorities accepted that a 25% reduction in these variables was an acceptable impact. A power analysis was then done to see whether or not such a change could be detected using these variables. The accepted criterion is that one can detect a change with 80% power. None of the measured variables met this criterion! Figure 1 shows that some only had a 20% power of detecting a change. Further analyses were done to try and improve power. A fivefold increase in sampling effort after the baseline survey made little difference, but a fourfold increase before and after it gave the necessary power to detect a change. This example illustrates that the application of these techniques will greatly improve the chances of detecting trends in monitoring programmes. However, the decision of whether or not to use such techniques rests not with scientists but with managers who fund monitoring programmes. A fourfold increase in the number of samples may not be economically acceptable, yet the consequence of not accepting this is that one probably will not detect a change although it is present.
FIG. 1. Illustration of power analyses as applied to measurement of biological variables used to assess the impact on benthic communities in the Øresund, Denmark (unpublished). A detection power of 80% is usually used as the accepted statistical criterion and can be envisaged as being equivalent to the familiar use of 95% certainty in most statistical analyses.

A hazard assessment is based on data of the expected environmental concentrations and the toxicological properties of the chemical or, alternatively, assessment of the magnitude of an expected physical impact. The process is illustrated in Fig. 2. Where there is a point source and/or a local impact, a knowledge of the production, the discharge patterns, and the loads and sources of potential impacts should be obtained in order to make predictions of which environmental compartment (water, biota or sediment) will be affected. Using data on the physico-chemical properties of the impacts, it is possible using QSARs to predict toxicity and techniques such as the n-octanol/water partition coefficient which can predict bioaccumulation [17]. There are often sufficient literature data on toxicity, persistence and bioaccumulation to enable an evaluative model to be made. At this stage it is usually necessary to incorporate data on the properties of the environment, such as hydrographic conditions and sediment types, in order to develop a proper mass balance. This model will then enable one to make an assessment of likely exposure. Data are needed on the biological systems and this may entail toxicity tests and obtaining data on the biological systems in the area that is likely to be exposed, using the new biological effects techniques mentioned above. With these data it is possible to make an assessment of likely effects. Both the exposure and effects assessments are then combined into a risk assessment. This may result in data which suggest the existence of a problem, and in that case there is a need to solve this by regulatory action, which will lower the predicted levels of impact to acceptable limits. The main purpose of
FIG. 2. An integrated framework for assessing effects of point source or local disturbance (Ref. [11], modified).

The risk assessment is to make quantitative predictions of the limits of effect or concentration. Such predictions use all the available data and models.

Once these predictions have been made by the scientists, the predictions should be fully appraised by a control body. In one of the largest construction works affecting the marine environment in Europe, the Great Belt Link, in Denmark, a control panel was set up involving the environmental authorities, the company doing the
construction, the scientific consultants who had made the predictions and the Green Movement. It was this body which agreed that the predicted impact was acceptable. At this stage it was also agreed a priori what the consequences would be if the limits of the predicted impact were to be exceeded. This took much time and debate, but is in the spirit of the precautionary principle and is a vitally important step. For example, as a result of eutrophication, the Great Belt has low oxygen concentrations in late summer and early autumn of most years. One concern was that excavation of sediment would lead to increased nutrient supply, which would enhance plankton blooms and lead to even lower oxygen concentrations and consequent effects on marine life.

The feedback loop agreed to was that if oxygen fell below 4 mg/L, then automatically, without having to ask the company, the consultants would undertake a wide-scale survey of a wide area in order to ensure whether or not the low oxygen values were limited to the area of the construction. This is illustrated in Fig. 3. This feedback loop was applied in 1991 and it was found that the oxygen concentration was low over the whole area; therefore, it was concluded that the Great Belt Com-

![Feedback Monitoring Diagram](image)

**FIG. 3.** Illustration of feedback monitoring as applied to the criterion of low oxygen concentration in the Great Belt (from Ref. [18]).
pany was not contributing to the low oxygen values. However, had the low oxygen concentration been limited to the near-field, then a feedback loop would automatically have come into operation which would have stopped excavation of sediments until the oxygen values were again above 4 mg/L. This was by no means a trivial agreement, since the company was using the world's largest cutter-dredger and the daily rental fee was extremely high.

This example further illustrates that the conditional hypotheses and feedback loop ideas derived a priori had direct consequences on the construction activities. Other feedback loops involved the risk of enhancing phytoplankton blooms where monitoring involved measuring fluorescence in situ. If the chlorophyll increased by a certain amount over a given time period, then again widespread monitoring was started to check whether or not it was an effect that could be attributed to the construction activities. Again, sediment excavation would stop if a link to the construction activity was found.

Environmental impact assessments (EIA) are now generally required almost worldwide before new activities that may affect the environment are approved. An EIA predicts the likely impact of a planned activity. In order to assess whether or not these predictions are correct, monitoring must be undertaken. Yet, usually, there is little follow-up as to whether the predictions are borne out, nor are there specific tests that have consequences for the constructor or the pollutant discharge if the predictions are incorrect. Often, one has to resort to post hoc remedial action or even litigation, which does not give adequate protection to the environment, a point emphasized by Earll [4]. The feedback monitoring, illustrated in Fig. 3, shows that it is possible to test EIA predictions and that this can have consequences for those making them [18].

4. CONCLUSIONS

From this brief survey it can be concluded that environmental scientists are producing new methods and new data which give a much better possibility of assessing the effects of contaminants on marine biota. In addition, there are a range of new approaches to both EIAs and monitoring that should give a better chance of managing the environment if the scientific techniques are applied within a proper framework.

What will be achieved by adoption of such a framework is that all decisions will be transparent and that the data will be openly available, and that by agreement a priori to remedial action or stopping of a damaging discharge the environment will be protected. With a sound monitoring programme and with agreed limits for a discharge, little or no danger to the environment should result from using only objective scientific criteria to judge whether or not limits have been exceeded.
Having set up the framework, the precautionary principle will be applied, but in the context of using the best available scientific expertise and acknowledging that the natural environment has an assimilative capacity to cope with man's wastes.

Based on a collection of regional surveys, GESAMP [19] has produced a report for the United Nations on the State of the Marine Environment. The report concludes that although man's fingerprint can be found everywhere and chemical contaminants are present from the poles to the tropics, the open ocean is still relatively clean and biological effects there are insignificant. On the other hand, the coasts are significantly affected, but, rather than chemical contamination and pollution effects, habitat destruction was regarded by GESAMP as having the strongest impact. Therefore, in any application of the precautionary principle, man's effects on physical processes also need to be taken into account.

REFERENCES


ENVIRONMENTAL MEASUREMENTS
AND TRANSFER STUDIES

(Poster Session 1)
ENIRONMENTAL MEASUREMENTS AROUND FRENCH NUCLEAR POWER PLANTS

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Electricité de France generates 75% of its electricity in nuclear power plants with pressurized water reactors (PWRs). These plants comprise 34 units of 900 MW and 20 units of 1300 MW, the first of which was connected to the grid in 1977 and the last in 1993. Three other units of 1400 MW are under construction.

The environmental measurements are performed in two complementary ways:

(1) Routine regulatory monitoring carried out by the operator according to a programme and procedures drawn up by the Central Ionizing Radiation Protection Service (SCPRI), which is the State monitoring authority in France. The SCPRI checks the results against those obtained with its own samples. This organization also monitors radioactivity throughout France (in particular through the TELERAY network).

   Around each power plant, the following are monitored:
   — Ambient gamma radiation (continuously) at eight points around the site within a radius of 5 km;
   — Aerosols in the air (once per day) at four points within a radius of 1 km;
   — Rain water and groundwater (monthly);
   — Surface waters (whenever there is a liquid radioactive discharge);
   — Milk and vegetables (monthly) at two points in the area close to the site.

   The plant has an off-site laboratory, two specially adapted vehicles and a team of three chemists. These measurements are quite separate from those conducted on radioactive wastes.

(2) Annual and ten-yearly radioecological measurement campaigns around the sites in order to improve the scientific knowledge of the environmental impact of the plants. The ten-yearly campaign consists of 'radioecological photographs' which are compared with the 'initial zero point'. About 40 samples are taken and various analyses performed (total alpha, beta and gamma spectra, strontium, carbon-14, free and organic tritium). The choice of samples and the places where they are taken
depend on the zero point and the special features of the region. Items sampled include drinking water, irrigation water, ground moss, vegetables, fruit, field crops, field soil, humic gley soil, meadow grass, milk, sewage sludge, wine, sediments, water or marine plants, fish or shellfish.

The annual campaigns around each site enable a picture to be built up of the radiation situation in time and space from gamma spectrometry measurements on 27 samples selected as the most suitable indicators (sediments, bryophytes, fish, moss, fruit, drinking water, milk, wine, soil).

The programme involves around 600 samples and 800 measurements per year and is being conducted, at our request, by the Institute for Radiation Protection and Nuclear Safety (IPSN), which has established the methodology for the sampling and measurements and has the capacity to ensure the continuity indispensable for this type of measurement.

The results of all these measurements are published and communicated to the public through the most appropriate media.

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IMPACT OF THE STRÁŽ URANIUM MINE (NORTHERN BOHEMIA) ON THE RIVER PLOUČNICE BASIN

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The river Ploučnice has been affected by the uranium industry for more than twenty years. Approximately 90% of the radioactive substances were removed by a provisional water treatment plant. The main contaminant was a barium-radium sulphate precipitate with a content of about 3 MBq $^{226}$Ra per kg barium. A detailed study on the contamination of water, sediments and soils in the flood plains was carried out in the period 1986–1992, in co-operation with the Water Board Ohře. A central decontamination station (CDS) has been in operation since 1989.

The contents of $^{226}$Ra and $^{238}$U in water samples have been determined according to official standards (LLD: 0.01 Bq $^{226}$Ra/L and 0.002 mg $^{238}$U/L). Gamma spectrometric analyses have been used for bottom sediments, soil, etc.

Water pollution. A power function of the type $A_v = aL^b$ approximated satisfactorily the relation between the $^{226}$Ra concentration ($A_v$) and the distance from the release point ($L$). A significant improvement of the water quality was detected
after putting the CDS into operation. The average annual $^{226}$Ra concentration in the river Ploučnice Noviny ($L = 3.5$ km) decreased from 0.7 Bq/L in suspended solids and 0.24 Bq/L in dissolved solids in 1986 to 0.08 Bq/L and 0.04 Bq/L, respectively, in 1992. By using the average $^{226}$Ra concentrations for both forms and the mean annual run-off from the river Ploučnice at the sampling points, the total load of $^{226}$Ra was calculated for particular years. The results were adjusted by the difference between the average $^{226}$Ra concentration of the river Ploučnice and that of its tributaries (background). From a comparison of the adjusted amounts of $^{226}$Ra, a distinct uptake of about 60% of $^{226}$Ra, mainly in its suspended form, was apparent in the period before operation of the CDS. Similar results were found for $^{238}$U.

**Bottom sediments.** The highest specific concentration of radioactive substances was found in the section between Mimoň ($L = 10$ km) and Hradčany ($L = 20$ km), where the river meanders and its slope is at a minimum. The maximum concentration observed was about 10 kBq $^{226}$Ra/kg dry sediment. The ratio $^{226}$Ra:$^{228}$Ra is a good indicator of the impact of waste products from the uranium mining industry on bottom sediments or soils in the flood plains of the river Ploučnice. The average value of this ratio for loaded reaches of the river is 32; for its tributaries, which are not affected by the uranium mining industry, it is less than 2.6.

**Transport of radioactive substances.** In the period 1986–1992, the transport of radioactive substances during floods in the river Ploučnice was tested by producing artificial floods using water from the Horka reservoir, situated in the Stráž pod Ralskem municipality. The largest decrease in the amount of transported radioactive substances, particularly in their suspended form (about 80%), caused by sedimentation in the river channel and mainly in the flood plain, was found to occur in the reaches of the river Ploučnice where the highest specific $^{226}$Ra concentrations in bottom sediments were measured.

**Flood plain contamination.** The results of the measurements made in the period 1992–1994 indicate a decrease in the dose rate. The highest value of the dose rate in the contaminated area was about 300 pGy/s. The background dose rate in this area ranges between 15 and 35 pGy/s.

**Conclusions.** A significant improvement in the water quality was found after the CDS was put in operation in 1989. The bottom sediments were decontaminated only in the surface layer. The ratio of the $^{226}$Ra:$^{228}$Ra concentrations in bottom sediments was used to evaluate the impact of the Stráž uranium mine. From the assessment of the $^{226}$Ra loads along the river Ploučnice it follows that $^{226}$Ra, which has accumulated in the basin during the whole operational period of the uranium mining industry, is relatively strongly bound. A decrease in the dose rate has been observed in the flood area; the effective half-life is about ten years.
A program for studying the consequences of radioactive releases of a (hypothetical) nuclear accident has been developed. Atmospheric dispersion, plume depletion by dry-out and wash-out, cloud-shine and ground-shine doses, dose commitments from inhalation and ingestion, and early and late health effects can be computed with this program. Effects of introduction of countermeasures are taken into account.

The actual values of all major parameters (from the core inventory to criteria for countermeasures) can be specified arbitrarily by the user; however, defaults are possible for all data. Meteorological conditions can be user specified (and updated at every step) or selected automatically from a library containing data observed for one year (8760 hours). The program can be used as follows:

- As a tool for education: it is applied in the training of people responsible for introducing countermeasures.
- In the case of a real accident: the program gives fast, easy to understand visual information on the radiological state of the environment and on expected environmental consequences.

The program works as a simulator; various scenarios can be specified (different emissions, meteorological conditions, countermeasures, etc.) and the results obtained for these scenarios can be directly compared with each other. Computations are carried out in one-hour steps for a maximum of 48 hours; meteorological and emission parameters can be newly set, and results for analysis are given at each step. It is possible to step back to earlier phases and to repeat the simulation under different conditions. Simulation is terminated by user request, or 48 hours after the beginning, or when the radioactive plume leaves the border of the country (or another preset region of interest).
A maximum of four (not necessarily adjoining) emission phases can be defined within one run. Two types of countermeasure scenarios can be introduced by the user; intervention criteria may be based on either activity concentration or dose type.

In the case of short term actions (actions that have to be introduced during the simulation period, viz. sheltering, iodine prophylaxis, evacuation), warnings are given when countermeasures should be introduced; the actual decision has to be taken by the user. A unique advantage of the interactive simulator is that the user can execute two parallel runs — with and without countermeasures — and compare the consequences.

Regarding long term countermeasures (relocation of people and prohibition of food consumption, which have to be introduced after the end of direct simulation but whose introduction affects the dose commitment), actions are assumed to be taken automatically if the present levels are exceeded.

Since the program is intended to serve for education and to present quick information for laymen, one of our main requirements was that it had to be easy to handle and the results had to be presented in an easy to understand, demonstrative manner. To fulfill these tasks, the program works under the Windows operating system with the usual menu, with dialogue and message boxes. The results (path of the plume, contaminated area, doses, health effects) are displayed on maps with spectacular symbols and/or in tables showing the computed values. Quantities are computed for about 400 segments, defined by sections of direction sectors and annular rings.

In the version first developed and presented, the emission point is taken to be the site of the Paks nuclear power plant. The maps (four maps with increasing magnification) cover the area of Hungary. All site specific databases are relevant to Hungary (population and food consumption data) or even to the Paks site/reactor (meteorology, core inventory, building dimensions). The segmented structure of the program, however, makes adaptation to another site relatively easy, provided that the data required for that site are available.
USE OF AERIAL SURVEYING FOR DETAILED MAPPING OF THE RADIOACTIVELY CONTAMINATED AREA AROUND THE CHERNOBYL NUCLEAR POWER PLANT

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In 1992, detailed mapping of the radioactive contamination of the region around the Chernobyl nuclear power plant was performed, using the aerial system for remote measurement of radioactive contamination in the case of an accident at a nuclear plant; this system has been developed by the Institute of Radioecology. On the basis of the data obtained with this system, detailed maps of isolines with a scale of 1:25000 were plotted.

Equipment configuration. The configuration of the equipment used for the aerial survey of radiation is shown in Fig. 1.

![Diagram](image-url)

**FIG. 1.** Configuration of the equipment used for aerial survey of radiation. Detectors: HPGe, 20% (portable); NaI(Tl), φ 150 × 100 mm$^2$; viewing angle of the collimator: 50–100°; height of flight: 100 m; speed of helicopter: 100 km/h; sensitivity for $^{137}$Cs contamination of the ground: 0.5 Ci/km$^2$ (20 kBq/m$^2$); minimum detected activity of local sources: natural background 0.005 Ci (200 MBq); Chernobyl area 0.1 Ci (4 GBq); spatial resolution 100 m.
Method of surveying. The data were obtained by a gamma spectrometer system installed on a helicopter. Measurements were done by scanning the territory from a height of 100 m. The velocity of the helicopter was about 100 km/h. Calibration coefficients for evaluating the flight data were measured at various heights for various types of landscape with different distributions of radionuclides in the soil.

The total count rate of the NaI(Tl) detector, the height of the flight and the navigation data were recorded every second. The count rates were used for locating lost sources and small radioactive spots. Spectral information was recorded periodically. The acquisition time of spectra depends upon the sensitivity of the HPGe detector; in the Chernobyl region it was 10–20 s because of the intensity of
radiation. This period permits measurement of a minimum activity of $^{137}\text{Cs}$ contamination at the ground of 0.5 Ci/km$^2$ (20 kBq/m$^2$) with an accuracy of 30%. The type of landscape (forest, water, agricultural area) was observed and indices of the landscapes were recorded by the data acquisition system.

An example of a flight profile is shown in Fig. 2. The high correlation between the total count rate of the detector and the area of the spectrum peak of $^{137}\text{Cs}$ was used to interpolate the density values of $^{137}\text{Cs}$ along profiles.

Figure 3 shows two profiles for which high peaks were registered; these were identified as very high activity 'point' sources. The spectrum and shape of the intensity curve were analysed. On the basis of these data the system response functions and the minimum detected activity were estimated experimentally.

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RADIOBIOLOGICAL EFFECT OF MONAZITE AND BLACK SANDS ON THE SURVIVAL AND SENSITIVITY OF OVOTESTIS OF Biomphalaria Alexandrina SNAILS

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The effect of radiation emitted from the naturally occurring mineral monazite and its mother ore black sand on Biomphalaria Alexandrina snails was investigated by a survival count and an ovotestis sensitivity assay. Monazite — a phosphate of thorium and rare earths with a small amount of uranium — contains all decay products of thorium and uranium, which are alpha, beta and/or gamma emitters. The radiological impact on the fauna and flora in the vicinity of sites where black sands exist and where monazite is produced or processed is the object of a wide research programme, of which the present work is the first part. There are large resources of black sands in Egypt in the Rosetta region where the Rosetta branch of the Nile Delta meets the Mediterranean Sea.

The survival of a group of B. Alexandrina snails, which transmit Schistosoma Mansoni in areas with monazite or black sands, was studied by daily measurement of the mortality of a group of these snails and in comparison with an unexposed control group. Moreover, an examination of the gonads was carried out at time periods of 3, 6 and 9 days of exposure, according to the survival time of the snails.
Laboratory reared *B. Alexandrina* snails were used in this work. Collected snails were maintained in the laboratory in plastic aquaria containing dechlorinated tap water which was changed every three days. The snails were adequately fed with fresh lettuce leaves. The water temperature of the aquaria was kept at 25 ± 2°C throughout the experimental period.

To study the effect of monazite or of black sand on the survival of the snail population, three groups (20 snails each) were used. Two of these groups were put in two aquaria, each containing 200 g monazite or black sand and 1 L dechlorinated tap water. The remaining group was kept as control. The aquaria were provided with air pumps, which allowed regular supply of oxygen.

The snails were inspected daily and the dead ones were counted and removed. Each experiment was repeated twice.

In order to study the effect of monazite and black sand on the gametogenesis of the snails, the hermaphrodite glands of the snails from each group were snipped out of the soft parts and dropped directly into Bouin’s fluid as fixative. The fixed tissues were dehydrated in alcohol, cleared in terpineol and embedded in paraffin.

For histological examination, 5 μm thick sections were stained with Delafield’s haematoxylin, counter-stained with eosin, dehydrated, cleared in xylene and mounted in Canada balsam. The histological features were examined at different times (3, 6 and 9 days).

Results of experiments on the influence of monazite or black sand on the survival of the three groups of *B. Alexandrina* snails are given in Table I. The data obtained show that the maximum survival time of snails exposed to monazite was 15 days; snails exposed to black sand survived up to 30 days. The effect of monazite and black sand on the sensitivity of the ovotestis of the snails is discussed in Refs [1-3].

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Number of survivors out of 20 snails after the following number of days:</th>
<th>Maximum survival (days)</th>
</tr>
</thead>
<tbody>
<tr>
<td>200 g monazite, 1 L water</td>
<td>20 20 18 16 11 6 0</td>
<td>15</td>
</tr>
<tr>
<td>200 g black sand, 1 L water</td>
<td>20 20 18 17 12 6 6 4 2 2 1 0</td>
<td>30</td>
</tr>
<tr>
<td>Control (only water)</td>
<td>20 20 20 19 19 18 18 18 18 17 17</td>
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REFERENCES


IAEA-SM-339/37P

METHOD FOR OPTIMUM ESTIMATION OF THE ENVIRONMENTAL RADIOLOGICAL PARAMETERS AFTER A MAJOR NUCLEAR ACCIDENT

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Hungarian Academy of Sciences,
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Measures taken after major nuclear accidents can affect the living conditions of many people and require a considerable amount of human and material resources. Therefore, decisions in such cases must be based on thorough consideration of all aspects, as well as the most comprehensive knowledge of the radiological situation in the environment. All possible means for assessing the temporal and spatial variation, the isotopic composition and the dose contribution of the radionuclide contamination should be utilized in a complex manner to support decisions regarding measures best suited for the given situation.

Data for decision making are often derived on the basis of transport calculations using emission and meteorological data. Because of the uncertainty of model parameters and the limited knowledge of the initial conditions, predictions can vary by orders of magnitude. A combination of model predictions and corrections of emission data will result in better estimation of the real radiological situation on the investigated site. The computer code OPTIMEST has been developed for complex treatment of collected data using the on-board computer of a mobile nuclear emergency laboratory. The derived optimum estimates of the radiological parameters can be transmitted by radio-telephone to the evaluation centre.
The basic idea behind the calculations with OPTIMEST is the fact that the environment is a complex system with links between its components. Radioecologists study these relationships by observations, experiments and computer modelling, and provide transfer factors (concentration ratios) that are used to describe the transport and/or equilibrium of radionuclides in nature. Some models are dynamic (time dependent), predicting the temporal variation of environmental radioactivity; others are static, giving concentration ratios for steady state conditions. In both cases the transfer parameters vary considerably, depending on a number of factors (knowledge of the initial conditions, meteorology, human activities, etc.), and the model predictions can deviate from reality by orders of magnitude.

OPTIMEST uses a formalism that combines predictions based on transfer factors and real measurements. An initial measurement vector (not necessarily complete) of the major radiological parameters is used to obtain estimates for all other components, using transfer factors provided by modellers. From the set of estimates the optimum one is obtained by averaging with properly chosen weights. Direct measurements are assigned unity, while all other estimates are assigned only fractions of unity, corresponding to a stronger or weaker relationship between the given quantity and the basis of derivation. For example, for calculating the best estimate of the radionuclide concentration in milk, a direct measurement of a milk sample is taken with a weight of 1.0, while the meat (beef) concentration is considered with a fraction of unity which is, however, higher than the concentration of radionuclides in air. On the basis of the best estimates for the radiological parameters, expected annual doses are also computed.

The following major pathways of human exposure are considered in the model used by OPTIMEST:

- Air activity concentration;
- Concentration of ground deposition;
- Concentration of radionuclides in water;
- Concentration of radionuclides in consumable vegetation;
- Concentration of radionuclides in animal feed;
- Concentration of radionuclides in meat (beef, pork, etc.);
- Concentration of radionuclides in milk;
- Dose rate of external irradiation.

The program OPTIMEST provides options for entering and modifying the initial measurement vector, and for loading and saving arbitrary sets of transfer factors, weights and dose conversion factors. An option is also given for calculating new transfer factors on the basis of the best estimate that helps to generate the set of values best fitting to the specific real environment. All data (measured values, derived and best estimates, annual dose for each component) are displayed on a graphic screen.
AQUATIC TRANSPORT AND ADSORPTION BEHAVIOUR OF THE ANTHROPOGENIC RADIONUCLIDES $^{60}$Co AND $^{137}$Cs

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Swiss Federal Institute for Environmental Science and Technology, Dübendorf, Switzerland

Since their installation in the early 1970s, nuclear power plants in Switzerland discharge radionuclides such as $^{60}$Co, $^{54}$Mn, $^{65}$Zn and $^{137}$Cs in low quantities in their waste waters. These discharges represent excellent objects for studying the fate of these radionuclides after their release into the aquatic environment. The list of factors determining the fate of these radionuclides is long. There are factors controlling adsorption of radionuclides by suspended particles of biological, chemical and detrital origin; factors controlling sedimentation and resuspension of particles; factors controlling decomposition of biological matter and redissolution of chemical precipitants; and factors controlling the ionic speciation of radionuclides in solution.

We present results from three different aquatic environments along the course of the river Aare in Switzerland: (1) in the 18 km section between the nuclear reactor Mühlberg, located 13 km west/north-west of Bern, and Lake Biel, (2) in Lake Biel, and (3) in the 7 km section between the nuclear reactor Beznau and the Aare/Rhine confluence. These sections have different physical characteristics and different types and concentrations of suspended particles. All sections have most significant annual variations, particularly regarding the importance of biological particles. In the Aare upstream of Lake Biel, detrital silicates are mostly dominant owing to natural erosion upstream. In the lake, their dominance decreases with distance from the Aare delta; plankton and chemical precipitates play a more dominant role. In the Aare further downstream, plankton continues to be the most important suspended particles because of natural filtration by the lake. These differences allow the above mentioned factors to be studied.

Waste water discharges from the Beznau and Mühlberg reactors occur every week or every second week; the total activities per discharge are several million becquerels of anthropogenic radionuclides. Measurements of $^{60}$Co, $^{65}$Zn, $^{54}$Mn and $^{137}$Cs are made by gamma spectrometry in untreated and filtered water samples, and in particles of different grain size obtained by in situ filtration and continuous flow centrifuging. To evaluate radionuclide transfer into lake and reservoir sediments, we measured the activities in sediment cores downstream of the two reactors.

Anthropogenic radionuclides are adsorbed into both inorganic and organic phases. The following sequence indicates the relative preference of different radionuclides for biogenic particles: $^{54}$Mn > $^{60}$Co > $^{65}$Zn > $^{137}$Cs. The dependence of
adsorption on grain size and mineralogy, observed in laboratory experiments, has been confirmed in the field. When inorganic particles dominate, in particular in the presence of algal colonies and high nitrogen and organic carbon concentrations, adsorption can increase with increasing particle size. Deposition of radionuclides in small reservoirs of hydroelectric plants was studied in two cases: 5 km and 6 km below a nuclear plant for $^{60}$Co. Deposition was found to be <10% of the total discharge. These results have to be viewed in the light of the dominance of grain-size fractions of sand and silt, which are inefficient adsorbers, and in the light of the observed slow natural adsorption rates. Thirty minutes after a discharge, less than 1% of the radionuclides are adsorbed by particles of >1 μm. More than 90% of $^{60}$Co remains 'in solution' after 3–10 hours of reaction time. Even after 2 days of waste water/particle contact, adsorption equilibrium is not reached. In the concentrated waste water, some of the radionuclides are in particulate form (~20% > 0.45 μm for $^{60}$Co) and some are 'in solution'. The radionuclides in solution are entirely cationic. An interesting trend is seen for $^{60}$Co. After 30 minutes of contact with the Aare water, 50% of the $^{60}$Co is found to be anionic, which we explain by complexation with organic ligands.

The behaviour of $^{60}$Co in Lake Biel is controlled by physical and biological aquatic parameters. During summer, while the lake has a stable stratification, the biological factor reaches a maximum. Under this condition the Aare flows into the epilimnion (controlled by temperature), and the water residence time can be as short as 5 days (based on theoretical considerations and a tracer experiment). During winter, total mixing takes place, the water residence time increases and the biological factor is reduced. This striking seasonal variation can be seen in sediment cores. Comparison of the monthly discharge of a nuclear reactor with the sediment content indicates that during summer a significantly smaller portion of $^{60}$Co enters the sediments than during winter.
Canadian utilities which operate CANDU reactors have established extensive radiological monitoring programmes to assess the effects of their releases on the environment and the local population. In order to assess the net impact of a given facility, it is necessary to establish background levels of radionuclides, which arise from natural background radionuclides, and anthropogenic radionuclides from other sources. If the background levels are ignored and the assumption is made that all the radioactivity observed comes from a given facility, then there can be a penalty when doses are estimated from the results. Background levels can be established initially through pre-operational surveys. However, the background levels of artificial radionuclides change with time and the pre-operational data soon become out of date. In addition, monitoring programmes change with operating experience, and radionuclides and pathways of current concern may not have been foreseen at the time when the pre-operational survey was designed. In 1992, a project to establish background levels in the vicinities of CANDU stations was started, with funding from the CANDU Owners Group (COG). The initial approach was to gather the available Canadian environmental radiological data and to assess their usefulness for establishing background levels.

In addition to the monitoring programmes operated by AECL and the COG utilities, environmental monitoring programmes have also been established by the Bureau of Radiation and Medical Devices (BRMD) of Health and Welfare Canada, the Ontario Ministry of Environment and Energy, the Ministry of Labour, and the Bedford Institute of Oceanography of Fisheries and Oceans Canada. Some of these institutions publish their data in reports, usually annually; however, some data are not formally reported.

The BRMD operates the most geographically widespread monitoring programme in Canada. It was decided to divide their data into geographical regions according to the locations of CANDU stations in Canada. These regions include: Southern Ontario (Toronto, Windsor and Ottawa), Eastern Ontario and Quebec (Quebec, Montreal and Ottawa), and the Maritimes (Fredericton, Halifax, St. John's, Summerside, Digby and Greenwood). This allowed calculation of
regional averages for gross beta activity in air and precipitation, $^{90}$Sr and $^{137}$Cs in milk, and exposures measured with thermoluminescent dosimeters. Data obtained by BRMD from other types of measurements, such as tritium in air and $^{90}$Sr and $^{137}$Cs in surface waters, which are made closer to specific CANDU stations, have also been examined. Data collected by other organizations, including background measurements made by the utilities, have also been reviewed and compared with BRMD regional averages.

Doses associated with background levels and the uncertainties in background levels introduced by using regional averages have been estimated using the models and parameters from a Canadian standard [1]. For example, published data for gross beta activity in air for 1987 [2] and 1988 [3] have been examined and the annual averages used to calculate regional averages for both years. The results (Table I) for the different regions are fairly close together, judging from the standard deviations, and the means should give good estimates of the average backgrounds for the regions. Applying an annual regional average background estimate to individual sites within the region could introduce an uncertainty of $\pm 0.05$ to $0.1 \text{ mBq/m}^3$, based on the site to site variation within the regions. If it were assumed that an excess in this range was due to $^{137}$Cs, the estimated dose to an adult through inhalation would be about 3-6 nSv/a. This is obviously a trivial dose compared with average annual natural background doses of 3-5 mSv/a. However, if it were assumed that the excess of 0.05-0.1 Bq/m$^3$ was due to a radionuclide with a higher dose conversion factor, say $^{90}$Sr, then the dose estimates could be several orders of magnitude higher, but, even for $^{90}$Sr, the estimated dose would still be less than 1 $\mu$Sv/a. Thus, the dose penalty due to the uncertainty in background concentrations of gross beta activity in air is quite small. Further results are reported in Ref. [4].

In conclusion, some of the available background data can be used to estimate background levels in the vicinities of CANDU stations in Canada. In many cases the doses associated with background levels or the uncertainties in background levels are trivial. However, there are cases where ignoring the background levels or making conservative assumptions in using the available background data results in dose

<table>
<thead>
<tr>
<th>Region</th>
<th>1987</th>
<th>1988</th>
</tr>
</thead>
<tbody>
<tr>
<td>Southern Ontario</td>
<td>0.40 (0.06)$^a$</td>
<td>0.36 (0.09)</td>
</tr>
<tr>
<td>Quebec</td>
<td>0.35 (0.03)</td>
<td>0.37 (0.05)</td>
</tr>
<tr>
<td>Maritimes</td>
<td>0.28 (0.05)</td>
<td>0.30 (0.05)</td>
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</table>

$^a$ Standard deviation in regional average.
penalties that might be considered unacceptable. The concept of dose penalty provides a useful tool for evaluating the need to precisely estimate background levels and doses around nuclear facilities. Wider application of this concept is encouraged.

REFERENCES


IAEA-SM-339/49P

STUDY OF MARINE RADIOACTIVITY
ALONG THE NORWEGIAN COAST, 1980–1994

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For the study of radionuclides in sea water along the Norwegian coast, brown algae were used as bioindicators. The algae reflect changes in seawater concentrations of radionuclides more rapidly than the water and also concentrate most radionuclides. Analysis of algae is therefore normally more convenient and has lower detection limits than analysis of the sea water itself.

The brown algae *Fucus vesiculosus* have been collected annually at several sampling locations since 1980 (Fig. 1), mainly in August and September [1]. At the location Utsira in the south-west, monthly samples of *Fucus* and sea water have been collected regularly since May 1986. The algae samples were dried and counted by high resolution gamma spectrometry. Time series of this length are very useful in studies related to marine radioecology.
In this 15 year period, the sources of artificial radionuclides in the sea along the Norwegian coast have mainly been the fallout from atmospheric nuclear weapons tests, releases from European reprocessing plants and the fallout from the Chernobyl accident.

In 1980, the radionuclide concentration was highest at the south-western locations Utsira and Lista (Fig. 2). It decreased both along the Skagerrak coast and northwards along the western coast to the Barents Sea. However, between Utsira and the northernmost location Ingøy, a distance of about 2000 km, the concentration decreased by not more than a factor of about two. The concentration of radiocaesium in algae in the period 1980–1994 reflects clearly the decrease in the releases from the reprocessing plant at Sellafield (Fig. 3). The influence of the similar plant at La Hague has been minor in comparison. The concentrations of $^{99}$Tc, $^{134}$Cs and $^{137}$Cs in Fucus compared with the Sellafield releases indicate a transport time from
Sellafield to Utsira of about 3 years, and another 2–3 years to the Barents Sea and the Kara Sea [2].

This study is relevant to the Norwegian–Russian investigations of nuclear materials dumped in the Kara Sea and to the work within the IASAP (international Arctic Seas assessment project) programme of the IAEA, as it gives information on the portion of the radioactive contamination of the Barents Sea and the Kara Sea originating from the European reprocessing plants [3].

The Chernobyl fallout in 1986 caused locally increased concentrations of radionuclides during 1986 and in the first year after the accident. It can be seen in Fig. 3 that the Chernobyl ‘peak’ had already occurred in 1986 at Utsira, but in 1987 at Vardo. This indicates a transport time of about one year from mid-Norway (sampling point 7) — a region which was heavily contaminated by the Chernobyl fallout. The outflow current from the Baltic Sea through the Danish straits and along the Swedish and Norwegian coasts has in later years led to increased radioactivity levels at the Skagerrak locations (Fig. 2). This is due to run-off from land into rivers entering the Baltic Sea. Run-off from large Norwegian rivers into the Oslo Fjord may also have contributed to this increase.

The annual variation at Utsira shows a higher concentration of radiocaesium in Fucus in the spring and summer seasons, with a marked peak in June, and with a concentration of more than twice that found in the period September to March.

Comparison with the radioactivity concentration in water samples from Utsira during 1986-1994 shows a concentration factor for Fucus/sea water (Bq/kg dry weight per Bq/L) of 150-250 (see Fig. 4) [4, 5]. The salinity of the water was 33-35‰.
REFERENCES


IAEA-SM-339/50P

IMPROVEMENT OF PRUSSIAN BLUE
BOUND TO ANION EXCHANGE RESIN BEADS FOR
RADIOCAESIUM DECONTAMINATION OF FOODSTUFFS

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MATFORSK (Norwegian Food Research Institute),
Ås, Norway

Prussian blue bound to anion exchange resin beads devoid of undesired smell and having a minimal release of water soluble materials has been produced. No release of soluble hexacyanoferrate, measured as Fe(CN)$_6^{3-}$, was detected in beads containing ‘insoluble’ Prussian blue. Such beads should be suitable for methods of emergency radiocaesium decontamination of foodstuffs following a major nuclear accident with food shortages in large areas.

Prussian blue was synthesized by consecutive binding of its constituents, Fe(CN)$_6^{4-}$ and Fe$^{3+}$, to macroreticular polystyrene-divinylbenzene anion exchange beads (Amberlite IRA-904, 16–50 wet mesh) [1, 2]. Special washing of the anion exchange resin before Prussian blue synthesis eliminated an unpleasant odour. After various numbers of incomplete cycles (Fe(CN)$_6^{4-}$ treatment last) or whole cycles (Fe$^{3+}$ treatment last) of synthesis the beads were rinsed three times in distilled water and dried. After three whole cycles of synthesis the beads were rinsed up to 12 extra times in distilled water or treated with a meat extract, an artificial meat fluid, 60 mM KH$_2$PO$_4$ or 60 mM K$_4$P$_2$O$_7$, followed by three water rinses. The
FIG. 1. Light absorbance spectra of the first bleeding test fluid of Prussian blue bound to polymer beads, (a) after three whole cycles and (b) after three whole cycles and a half-cycle of Prussian blue synthesis.

FIG. 2. Light absorbance of supernatants from bleeding test of Prussian blue bound to anion exchange polymer beads after one to four whole cycles (filled symbols) and after 1.5, 2.5 and 3.5 cycles (open symbols) of Prussian blue synthesis.
stability of the beads was evaluated in a bleeding test in which the supernatants were investigated by spectrophotometry after seven consecutive 23 h incubations of beads in 0.9% NaCl [2].

For beads from incomplete cycles of Prussian blue synthesis, the bleeding test supernatants were yellow, with reproducible absorbance peaks at 262, 302 and 420 nm, as in a pure $K_3Fe(CN)_6$ solution, whereas beads from whole cycles of synthesis gave colourless supernatants with significant light absorbance below 400 nm only (Fig. 1). The absorbance decreased with an increasing number of bleeding test incubations, resulting in very low values for whole-cycle beads, whereas the values for half-cycle beads were still significant after seven incubations (Fig. 2).

After three whole cycles of Prussian blue synthesis the beads released less light absorbing solutes after six to twelve water rinses, or after treatment with meat extract, artificial meat fluid or $KH_2PO_4$, compared with untreated beads. However, the level of bleeding in the seventh test fluid from extra rinsed or post-synthesis treated beads was roughly the same as in untreated beads. $K_4P_2O_7$ treatment destabilized the beads, as indicated by a high rate of bleeding throughout the bleeding test (not shown).

It is concluded that Prussian blue bound to anion exchange resin beads is more stable after whole cycles of Prussian blue synthesis. The chemical form of Prussian blue in these beads is probably 'insoluble' Prussian blue, $Fe_4[Fe(CN)_6]_3 \sim 15H_2O$. Bleeding can be minimized by extensive rinsing or by post-synthesis treatment with inorganic phosphate.

REFERENCES


Large amounts of airborne particles of dispersed nuclear fuel and condensed particles were released to the atmosphere after the accident at the Chernobyl nuclear power plant (NPP). Condensed particles are the well known form of aerosol in the air of working places in the nuclear industry. Aerosol particles of fragmented nuclear fuel contain fission products and transuranium radionuclides that are strongly bound to the uranium-oxygen matrix.

Nuclear fuel particles are a rare form of airborne radioactivity. Similar aerosol particles have been found in the environment only after nuclear tests; however, they have not been well investigated radiologically. Our hypothesis is that because of the formation of a matrix, nuclear fuel particles have a collective behaviour, both in the environment and in the body. Bound by the matrix, the radionuclides do not escape from nuclear fuel particles until the matrix is chemically destroyed. If one nuclear fuel radionuclide marker is found in a particle of a sample from the environment or from human organs, it must be expected that all other nuclear fuel particle radionuclides are also present in this sample. Their initial normalized activities must be close to those of representative nuclear fuel particles (see Table I) [1].

Nuclear fuel particles are grains produced by fragmentation of fuel pellets. Their uranium-oxygen crystal lattice (matrix) strongly holds fission and transuranium radionuclides. By analogy with the assignment of uranium oxides by the International Commission on Radiological Protection to inhalation class Y, all radionuclides entering the respiratory system as part of fuel particles should be assigned to the same inhalation class. A model for the behaviour of matrix bound radionuclides in the body is presented in Ref. [2]. Our findings confirm the hypothesis that there is a strong bond between the uranium matrix and nuclear fuel particle radionuclides of a different nature, such as caesium and plutonium, and that these radionuclides are present in aerosol particles of the same size.

To test matrix binding of radionuclides in nuclear fuel particles, a special investigation of the kinetics of a dialysis (120 d) of samples from the damaged Chernobyl Unit 4 was carried out. The solubility of over 70% of $^{137}$Cs was found
<table>
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<tr>
<th>Isotope</th>
<th>Normalized activity</th>
<th>Isotope</th>
<th>Normalized activity</th>
<th>Isotope</th>
<th>Normalized activity</th>
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<td>Te-129</td>
<td>$3.3 \times 10^{-2}$</td>
<td></td>
<td>Pr-144m</td>
<td>$1.4 \times 10^{-2}$</td>
<td></td>
<td>Pu-240</td>
</tr>
<tr>
<td>Ru-103</td>
<td>$1.1 \times 10^0$</td>
<td></td>
<td>Te-129m</td>
<td>$4.5 \times 10^{-2}$</td>
<td></td>
<td>Pr-145</td>
<td>$3.4 \times 10^{-2}$</td>
<td></td>
<td>Pu-242</td>
</tr>
<tr>
<td>Ru-105</td>
<td>$9.3 \times 10^{-3}$</td>
<td></td>
<td>Te-131</td>
<td>$1.7 \times 10^{-2}$</td>
<td></td>
<td>Nd-147</td>
<td>$5.2 \times 10^{-1}$</td>
<td></td>
<td>Pu-241</td>
</tr>
<tr>
<td>Ru-106</td>
<td>$3.0 \times 10^{-1}$</td>
<td></td>
<td>Te-131m</td>
<td>$7.6 \times 10^{-2}$</td>
<td></td>
<td>Pm-147</td>
<td>$2.9 \times 10^{-2}$</td>
<td></td>
<td>Pu-243</td>
</tr>
<tr>
<td>Rh-105</td>
<td>$4.1 \times 10^{-1}$</td>
<td></td>
<td>Te-132</td>
<td>$5.0 \times 10^{-1}$</td>
<td></td>
<td>Pm-148</td>
<td>$7.6 \times 10^{-2}$</td>
<td></td>
<td>Am-241</td>
</tr>
<tr>
<td>Rh-106</td>
<td>$3.2 \times 10^{-1}$</td>
<td></td>
<td>I-130</td>
<td>$4.1 \times 10^{-2}$</td>
<td></td>
<td>Pm-148m</td>
<td>$6.9 \times 10^{-2}$</td>
<td></td>
<td>Cm-242</td>
</tr>
<tr>
<td>Pd-109</td>
<td>$5.2 \times 10^{-2}$</td>
<td></td>
<td>I-131</td>
<td>$5.9 \times 10^{-1}$</td>
<td></td>
<td>Pm-149</td>
<td>$4.5 \times 10^{-1}$</td>
<td></td>
<td>Cm-244</td>
</tr>
<tr>
<td>Isotope</td>
<td>Wall of lower large intestine</td>
<td>Lungs</td>
<td>Liver</td>
<td>Bone surfaces</td>
<td>Red bone marrow</td>
<td>Testes</td>
<td>Contribution to committed effective dose (%)</td>
<td></td>
<td></td>
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<tr>
<td>------------</td>
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</tr>
<tr>
<td>Zr-95</td>
<td>2.9</td>
<td>1.9</td>
<td>0.4</td>
<td>0.6</td>
<td>1.2</td>
<td>1.3</td>
<td>1.6</td>
<td></td>
<td></td>
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<tr>
<td>Mo-99</td>
<td>4.0</td>
<td>0.2</td>
<td>0.1</td>
<td>&lt;0.1</td>
<td>0.1</td>
<td>0.2</td>
<td>2.2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ru-103</td>
<td>2.0</td>
<td>0.6</td>
<td>0.1</td>
<td>&lt;0.1</td>
<td>0.2</td>
<td>0.6</td>
<td>1.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ru-106</td>
<td>5.7</td>
<td>10.0</td>
<td>0.1</td>
<td>&lt;0.1</td>
<td>0.2</td>
<td>0.9</td>
<td>5.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ba-140</td>
<td>7.3</td>
<td>1.1</td>
<td>0.1</td>
<td>&lt;0.1</td>
<td>0.3</td>
<td>0.9</td>
<td>3.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>La-140</td>
<td>6.0</td>
<td>0.3</td>
<td>0.3</td>
<td>&lt;0.1</td>
<td>0.4</td>
<td>1.4</td>
<td>3.2</td>
<td></td>
<td></td>
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<tr>
<td>Ce-144</td>
<td>17.0</td>
<td>26.0</td>
<td>16.0</td>
<td>1.1</td>
<td>6.6</td>
<td>2.0</td>
<td>16.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Np-239</td>
<td>9.7</td>
<td>0.5</td>
<td>0.1</td>
<td>0.2</td>
<td>0.3</td>
<td>0.5</td>
<td>5.2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu-238 +</td>
<td>&lt;0.1</td>
<td>9.0</td>
<td>31.0</td>
<td>35.0</td>
<td>30.0</td>
<td>28.0</td>
<td>9.2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu-239</td>
<td>&lt;0.1</td>
<td>6.7</td>
<td>38.2</td>
<td>53.3</td>
<td>47.0</td>
<td>53.3</td>
<td>12.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu-241</td>
<td>&lt;0.1</td>
<td>14.2</td>
<td>8.5</td>
<td>6.8</td>
<td>6.0</td>
<td>4.5</td>
<td>3.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cm-242</td>
<td>0.1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total</td>
<td>55</td>
<td>71</td>
<td>95</td>
<td>97</td>
<td>92</td>
<td>94</td>
<td>63</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* AMAD = 15 μm, standard geometric deviation = 2.8.
to be weak, with a leaching constant of $0.002 \pm 0.001 \text{ d}^{-1}$. For the alpha emitters, the dialysis constant was $0.005 \pm 0.002 \text{ d}^{-1}$. The difference between the leaching constants for $^{137}\text{Cs}$ and for the total alpha emitters is not statistically significant ($p > 0.05$) and the effective constant for radionuclide leaching from nuclear fuel particles is estimated at $0.003 \pm 0.002 \text{ d}^{-1}$.

The activity median aerodynamic diameter (AMAD) of Chernobyl nuclear fuel particle aerosol for the first days after the accident was not measured. The only way to obtain AMAD values is to use indirect methods based on an investigation of the AMAD dependent behaviour of nuclear fuel particle radionuclides in bodies of persons who inhaled that aerosol at the time of the accident. Two groups of staff of the Chernobyl NPP and firemen who witnessed the accident were investigated.

The first test group was chosen from the workers involved in operations on 26–27 April 1986 and who had been measured with a semi-conductor whole-body counter. It was found that, even 700–800 d after the accident, the $^{137}\text{Cs}$ activity in these persons was more than 100 nCi,$^1$ i.e. the average value for 1987–1988 for persons working at Chernobyl who had not been involved in emergency operations in April–May 1986. A median half-life of $^{137}\text{Cs}$ of 330 d for this test group was found. It corresponds to the $^{137}\text{Cs}$ behaviour in the body after a single inhalation of a nuclear fuel particle aerosol with an AMAD of $16 \pm 2 \mu\text{m}$.

The second test group included workers at the Chernobyl NPP and firemen who died of acute radiation sickness in 1986. The results of post-mortem studies on the $^{239}\text{Pu}$ distribution in their organs [3] were used for validation of the AMAD of nuclear fuel particles. From these data, the mean AMAD of fuel particles was estimated at $12 \pm 2 \mu\text{m}$. This estimate proved to be in good agreement with estimates of the $^{137}\text{Cs}$ behaviour in living witnesses to the Chernobyl accident, with the evaluated nuclear fuel particle AMAD for the plutonium distribution in the bodies of residents of the Gomel region [2], and with the results of an examination of soil samples collected in Germany in May 1986 [4].

A special computer code was developed which was based on the biokinetic model for the behaviour of nuclear fuel particle radionuclides in the human body after acute inhalation [1]. The code was used for investigating the dosimetric properties of nuclear fuel particles. The results of an evaluation of the contribution of the main radionuclides of nuclear fuel particles to the committed doses to different organs are presented in Table II.

The studies showed the unique properties of the Chernobyl nuclear fuel particle aerosol and confirmed the assumption that nuclear fuel particle radionuclides are in aerosol particles of the same size and have a similar chemical speciation, being matrix bound. The findings should be considered in planning protective measures and in assessments of health effects following a major accident at a nuclear power installation.

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$^1$ 1 Ci = 37 GBq.
REFERENCES


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TRANSFER OF RADIOCAESIUM FROM SOIL TO SOME CROPS IN SEMI-ARID AREAS OF SYRIA

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Radioactive contamination of soil resulting from releases of radionuclides and their transfer to plants is one of the most important sources of human exposure. An investigation of radionuclides in crops produced in the Syrian Arab Republic has been performed. The concentration of $^{137}$Cs in soil was quite low (<20 Bq/kg), but this concentration led to measurable $^{137}$Cs levels in some crops.

Table I gives data for transfer factors (TFs) of $^{137}$Cs for different crops in the Syrian Arab Republic. These data show the variation of TFs, which might be due to variation of species, soil types and weather conditions. We have investigated the transfer of radionuclides to different crops in typical soils in the Syrian Arab Republic.

A field experiment was designed for this purpose; about 36 m$^2$ of land were contaminated with $^{137}$Cs (~5000 Bq/kg) to a depth of more than 30 cm. Soil labelling was carried out after saturation of the soil with water. The radionuclides were diluted in about 50 L to obtain a concentration of about 37 MBq/L and the mixture was applied by spraying. A surface contamination dosimeter was used to ensure homogeneity of radionuclide distribution on the surface. When the area was nearly dry, the soil was ploughed ten times. The homogeneity of the radionuclide distribution was evaluated by gamma spectrometry of 13 soil columns in the contaminated area.
### TABLE I. TRANSFER FACTORS OF FALLOUT $^{137}$Cs FROM SOIL TO SOME CROPS

<table>
<thead>
<tr>
<th>Crop</th>
<th>Location</th>
<th>Sampling date</th>
<th>$^{137}$Cs in soil (Bq/kg dry weight)</th>
<th>$^{137}$Cs in crops (Bq/kg dry weight)</th>
<th>TFs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spinach</td>
<td>Alkamishly</td>
<td>April 1993</td>
<td>21 ± 3</td>
<td>0.3 ± 0.1</td>
<td>0.014</td>
</tr>
<tr>
<td>Vine leaves</td>
<td>Deer Alzor</td>
<td>June 1993</td>
<td>10 ± 2</td>
<td>0.4 ± 0.2</td>
<td>0.04</td>
</tr>
<tr>
<td>Vine leaves</td>
<td>Tartous</td>
<td>June 1993</td>
<td>44 ± 3</td>
<td>0.6 ± 0.4</td>
<td>0.013</td>
</tr>
<tr>
<td>Tomatoes</td>
<td>Tartous</td>
<td>June 1993</td>
<td>14 ± 2</td>
<td>0.9 ± 0.3</td>
<td>0.06</td>
</tr>
<tr>
<td>Tomatoes</td>
<td>Tartous</td>
<td>June 1993</td>
<td>8.3 ± 1</td>
<td>0.4 ± 0.2</td>
<td>0.04</td>
</tr>
<tr>
<td>Broad beans</td>
<td>Tartous</td>
<td>April 1993</td>
<td>40 ± 4</td>
<td>1 ± 0.2</td>
<td>0.05</td>
</tr>
</tbody>
</table>

### TABLE II. TRANSFER FACTORS OF $^{137}$Cs FROM CONTAMINATED SOIL TO EDIBLE PLANT PARTS

<table>
<thead>
<tr>
<th>Crop</th>
<th>Part</th>
<th>Drying factor</th>
<th>Irrigation water (mm)</th>
<th>Fertilizer (kg/ha)$^a$</th>
<th>TFs $^{137}$Cs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chick-peas</td>
<td>Grain</td>
<td>0.33</td>
<td>455</td>
<td>92 125 27.5 27.5</td>
<td>$1.2 \times 10^{-2}$ $\pm 6 \times 10^{-4}$</td>
</tr>
<tr>
<td>Beans</td>
<td>Grain</td>
<td>0.56</td>
<td>455</td>
<td>184 75 27.5 27.5</td>
<td>$1.0 \times 10^{-2}$ $\pm 6 \times 10^{-4}$</td>
</tr>
<tr>
<td></td>
<td>Pod</td>
<td>0.38</td>
<td></td>
<td></td>
<td>$1.4 \times 10^{-2}$ $\pm 1 \times 10^{-3}$</td>
</tr>
<tr>
<td>Tomatoes</td>
<td>Fruit</td>
<td>0.05</td>
<td>455</td>
<td>46 80 64.5 64.5</td>
<td>$2.0 \times 10^{-2}$ $\pm 3 \times 10^{-3}$</td>
</tr>
<tr>
<td>Sorghum</td>
<td>Grain</td>
<td>0.93</td>
<td>455</td>
<td>46 80 57.5 57.5</td>
<td>$1.2 \times 10^{-2}$ $\pm 5 \times 10^{-4}$</td>
</tr>
<tr>
<td>Water melons</td>
<td>Flesh</td>
<td>0.06</td>
<td>455</td>
<td>92 100 27.5 27.5</td>
<td>$1.0 \times 10^{-2}$ $\pm 1.5 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

$^a$ P and K were added at seeding.

$^b$ N was added in two stages, at seeding and one month after seeding.
After assurance of homogeneity, the land was sown with different kinds of crop: tomatoes, beans, chick-peas, melons and sorghum. Irrigation and fertilization were carried out as usual, and the TF was calculated as the ratio between the radiouclide activity in dry edible plant parts and that in the soil. The results obtained (Table II) are in good agreement with those reported for other investigations.

IAEA-SM-339/64P

STUDY OF TURBULENT ATMOSPHERIC DISPERSION OF POLLUTANTS AT LOW WIND VELOCITIES

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Low wind velocities coupled with strong atmospheric stability constitute complex diffusion situations, surface layer modelling of which proves to be difficult at present because of lack of theoretical bases and experimental data [1]. Under these conditions, diffusion of a pollutant on the vertical plane is governed by thermal phenomena. On the horizontal plane, according to site orography, fluctuations in the wind direction may determine the mixing process [2, 3]. These fluctuations depend on flow velocity [4] as well as on undulatory phenomena generated by the relief of the area and by exchanges with the synoptic layer [5].
We report here the first results obtained during a campaign carried out by IPSN and Electricité de France in August 1993 at the Salon de Provence airfield (France). Five atmospheric tracing trials using SF₆ were performed at night, with a wind velocity of less than 2 m/s at ground level. The evolution in space and time of the concentrations 1 m above the ground was measured by means of 40 air sampling devices distributed over three concentric sections. At all stations, samples were taken during successive periods of one quarter of an hour, for five hours, from the beginning of the release (11 kg SF₆ emitted at a constant rate for half an hour, 2 m above ground level). Analyses of the concentrations were made every quarter of an hour; also determined were the atmospheric transfer coefficients (ATCs), which correspond to the total content at each sampling point divided by the mass of emitted tracer. The available meteorological apparatus allowed the characteristics of the wind to be determined during the tracing trials. Point A was equipped with three 3-D sonic anemometers placed at heights of 6, 11 and 30 m, a 3-D Doppler sodar, a dish anemometer at a height of 11 m, and temperature, humidity and radiation sensors at a height of 1 m. At point B, a fourth sonic anemometer was placed at a height of 3 m. Point C was equipped with a dish anemometer at a height of 3 m, and a temperature and humidity sensor at a height of 2 m. Point D comprised a dish anemometer at a height of 30 m and a system for determining temperature deviations over the first thirty metres.

The parameters of the wind sampled at high frequency by the sonic anemometers were recorded by a micro-computer during the tracing period. This information allows the turbulence of the wind field to be characterized and, subsequently, the stability of the atmosphere to be deduced. The temperature deviation over the first thirty metres suggested high stability. Every twenty minutes, the Doppler sodar provided profiles of the deviations and means of the three components of wind at a height of 50-600 m above the ground. The data from the other sensors were presented in the form of means and standard deviations over 6 min or 10 min periods.

An analysis of the mean components of the meteorological data and the concentrations shows inconsistencies in space and time in the wind field. These could be the localized results of abnormally high concentrations and alterations of the plume, which conventional Gaussian type models — based on the hypothesis of a homogeneous field — cannot reproduce. Analyses of related statistics on wind velocities at the different sampling stations are now being processed, with the aim of characterizing these inconsistencies with great precision. The very high sensitivity of the sonic anemometers to light winds allowed verification of the fact that, despite the considerable wind variations in time and space, the overall distribution of the isoconcentrations was mainly determined by the distribution effect of the wind directions observed during the release. Thus, considering the mean values of the wind during tracer emission, the ATCs calculated by Gaussian models did not excessively deviate from the experimental ATCs. However, the predicted ATCs
were generally higher than the observed values and, correspondingly, the predicted widths of the plume were considerably smaller than the experimental widths.

At the present stage of the study it appears that, for situations of low wind velocities, the prediction of impact in terms of collective doses using ATCs provided by a Gaussian type computer code is reasonable. However, the difficulty in predicting the more random evolution of concentrations in time and space with this type of model can result in critical situations regarding individual doses that exceed the permissible dose for exposure to radiation. Numerical modelling using simple adapted models or use of a 3-D model that can take into account wind variations in time and space will complete this experimental analysis.

REFERENCES

Introduction. Annual radioecological monitoring of gamma emitter radio-nuclides in the environment of the 19 French nuclear power plants (NPPs) has been conducted since 1992. We review here the results of analyses of terrestrial mosses (1992 and 1993), which are considered to be the best indicators of atmospheric radioactive contamination. Our intention is to trace the different origins of radionuclides present in the terrestrial environment of these French NPPs: fallout from atmospheric nuclear weapons tests (\(^{137}\text{Cs}\)), fallout from the Chernobyl accident (\(^{134}\text{Cs}, {^{137}\text{Cs}}\)) and fallout from atmospheric wastes (\(^{58}\text{Co}, {^{60}\text{Co}, {^{134}\text{Cs}, {^{137}\text{Cs}}}\)) from NPPs (Fig. 1).

Method. Locations of sampling points were determined in areas around French NPPs according to local wind conditions, i.e. two or three sampling points in the prevailing wind direction (affected area) and one sampling point elsewhere (unaffected area). Results are analysed on the basis of the \(^{137}\text{Cs}/^{134}\text{Cs}\) ratio. More precisely, the isotopic ratio measured (RI\(_m\)) at a given date is compared with the theoretical isotopic ratio (RI\(_t\)) at the same date, assuming that both \(^{137}\text{Cs}\) and \(^{134}\text{Cs}\) result totally from the Chernobyl accident.

Results. (1) Mosses are contaminated only by \(^{137}\text{Cs}\) and \(^{134}\text{Cs}\); (2) The spatial distribution of \(^{134}\text{Cs}\) in mosses in 1992 and 1993 is similar to that of \(^{134}\text{Cs}\) deposits after the Chernobyl accident in 1986 (Fig. 2); and (3) the RI\(_m\) in the affected area is not significantly different from that in the unaffected area (Fig. 3).
FIG. 1. Average composition of gaseous wastes from French NPPs (St. Laurent des Eaux, 1992 and 1993).

(\textsuperscript{60}Co 27\%  
\textsuperscript{58}Co 21\%  
\textsuperscript{134}Cs 16\%  
\textsuperscript{137}Cs 21\%  
Others 15\%)

FIG. 2. (a) Spatial distribution of \textsuperscript{134}Cs for average deposits on soils (Bq/m\textsuperscript{2}) after the Chernobyl accident in 1986. (b) Average concentrations of \textsuperscript{134}Cs (Bq/kg dry weight) in mosses in 1992 and 1993.
FIG. 3. Comparison of $R_{I_m}$ values in affected areas (AA) and in unaffected areas (UA) in 1992 and 1993.

FIG. 4. Average isotopic ratio ($R_{I_m}$) of radiocaesium in mosses, compared with the theoretical isotopic ratio ($R_{I_t}$) in 1992 and 1993.
Conclusions. (1) The atmospheric radioactive wastes from the French NPPs have no visible effect on mosses: $^{58}$Co and $^{60}$Co are absent and the $\text{RI}_{m}$ of radio-caesium is independent of the sampling area. (2) With regard to the other two origins of radiocaesium, the French territory can be divided into two areas, i.e. east and west of four degrees east of the Greenwich meridian. In the western area, the mean $\text{RI}_{m}$ is significantly higher than the $\text{RI}_{t}$; in the eastern area, such a difference exists, but to a lesser degree (Fig. 4). These values from 1992 and 1993 indicate that the influence of fallout from atmospheric nuclear weapons tests (increasing the $\text{RI}_{m}$) is greater in the western area. Considering that this fallout is constant over the whole territory, it can be concluded that the impact of the Chernobyl accident is more significant in the eastern area than in the western area. This conclusion is in agreement with the comparison between the $^{134}$Cs deposits on soil in France in May 1986 and the $^{134}$Cs concentration in mosses in 1992 and 1993 (Fig. 2).

**IAEA-SM-339/66P**

**RADON EXHALATION FROM URGEIRIÇA URANIUM TAILINGS**

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The Urgeiriça uranium mine is one of the most significant ore deposits in the continental temperate and dry zone of Beira Alta.

Exploitation of the mine started in 1913 and it was found to be one of the most important uraniferous deposits in Europe. Activities in this mine were exclusively dedicated to radium production until 1944, when uranium production became more important. In 1951 a chemical treatment unit for production of poor $\text{U}_3\text{O}_8$ concentrates was built and in 1967 it was transformed into a new unit with a treatment capacity of about 150 t of ore per day [1]. The mine is now almost exhausted, but its facilities are still used for the treatment of ore from other mines.

Milling wastes have accumulated in this region, near the mine, and the tailings piles extend over an area of about $11 \times 10^4 \text{ m}^2$, with a total deposit of four million tonnes. Some of the piles are covered with vegetation to avoid erosion.

The Urgeiriça mine is surrounded by small farms and country houses, and most of the local population lives in a village (Canas de Senhorim) about 2 km from the mine.
A surveillance programme was started in 1993 to assess the radon exhalation rates from the tailings piles around the mine. Several tailings were monitored in order to determine the radon exhalation rates from tailings covered with different materials.

Determination of radon flux was based on an accumulation method. Nuclear track detectors, with Kodak LR115 films, were exposed in special devices with a volume of about 10 L and an open end, which were placed above the tailings to be monitored. Integrated measurements were performed during 24 hours; this period of time for experimental in situ testing was chosen in order to avoid buildup to a level at which the radon concentrations will be reduced by back-diffusion into the measured area [2]. The radon concentration in the device is proportional to the flux rate and to the area of the device in contact with the tailings surface.

Several devices were distributed over the tailings (top and asents) in order to obtain detailed data on the actual radon exhalation rates from the area. Since the radon exhalation rates are strongly dependent on the meteorological conditions, several series of measurements were performed during different periods of the year.

Five tailings piles of different ages (piles with covers and without covers) have been considered: the tailings piles T1, T2 and T3 are covered with bushes, eucalyptus and pine trees, with T1 being the oldest pile; pile T7 is covered with a gravel layer of about 1 m thickness; and pile T8 (without cover) is still receiving milling wastes. The results obtained are presented in Fig. 1.

![Graph showing average radon exhalation rates from different tailings piles.]

**FIG. 1. Average radon exhalation rates from different tailings piles.**
Higher exhalation rates were found in the spring measurements for all tailings piles except T1. The lower values are for pile T7, which is covered by gravel. The highest values were found for T1, the oldest pile, which contains milling wastes from processing and treatment of ore for uranium recovery with a low yield.

REFERENCES


IAEA-SM-339/73P

TRANSFER OF $^{232}$Th, $^{230}$Th, $^{228}$Th AND $^{238}$U, $^{234}$U TO PLANTS VIA THE ROOT SYSTEM AND ABOVE-GROUND PARTS

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Warsaw, Poland

The transfer of thorium and uranium isotopes to plants used as fodder (alfalfa, grass) and food (cereals, vegetables, potatoes) has been studied. Plants were grown in an open field (exposure to dry and wet deposition, resuspension and soil adhesion) and in a polyethylene tent with an underground irrigation system (isolation from wet deposition and from water splash on soil). A sandy loam soil was used which contained $^{232}$Th, $^{230}$Th, $^{228}$Th, $^{238}$U and $^{234}$U at average concentrations of 19.5 ± 1.7, 19.7 ± 1.4, 21.1 ± 1.9, 18.8 ± 1.3, 19.1 ± 1.6 Bq/kg dry weight, respectively. The experiments were carried out in 1993–1994.

The above-ground parts of the plants were washed in distilled water. The isotopes $^{232}$Th, $^{230}$Th, $^{228}$Th and $^{238}$U, $^{234}$U were determined in the soil, in the washed plants, in the rinse and in insoluble residue separated from the rinse. Thorium and uranium isotopes were determined radiochemically in the presence of $^{229}$Th and $^{232}$U tracers, which were added to the ashed samples. The radionuclides were deposited on stainless steel discs and their activity was measured by alpha spectrometry.
### TABLE I. AVERAGE CONCENTRATIONS OF $^{232}$Th, $^{230}$Th AND $^{228}$Th (mBq/kg d.w.) IN PLANTS GROWN ON AN OPEN FIELD AND IN PLANTS GROWN IN A TENT

<table>
<thead>
<tr>
<th>Plant</th>
<th>$^{232}$Th</th>
<th></th>
<th>$^{230}$Th</th>
<th></th>
<th>$^{228}$Th</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Field</td>
<td>Tent</td>
<td>Field</td>
<td>Tent</td>
<td>Field</td>
<td>Tent</td>
</tr>
<tr>
<td>Alfalfa</td>
<td>54.2 ± 8.7</td>
<td>35.0 ± 5.4</td>
<td>63.0 ± 9.5</td>
<td>33.0 ± 2.9</td>
<td>112.2 ± 13.1</td>
<td>38.1 ± 5.7</td>
</tr>
<tr>
<td>Grass</td>
<td>68.1 ± 8.7</td>
<td>54.1 ± 8.4</td>
<td>84.6 ± 9.3</td>
<td>56.4 ± 8.6</td>
<td>146.3 ± 13.7</td>
<td>120.3 ± 14.2</td>
</tr>
<tr>
<td>Barley straw</td>
<td>37.9 ± 5.8</td>
<td>29.0 ± 4.0</td>
<td>40.2 ± 5.9</td>
<td>30.5 ± 4.2</td>
<td>156.2 ± 13.4</td>
<td>120.6 ± 11.7</td>
</tr>
<tr>
<td>Barley grain</td>
<td>7.6 ± 2.6</td>
<td>6.2 ± 2.4</td>
<td>9.0 ± 2.4</td>
<td>7.0 ± 2.6</td>
<td>36.4 ± 4.3</td>
<td>34.0 ± 3.8</td>
</tr>
<tr>
<td>Spinach</td>
<td>80.4 ± 12.1</td>
<td>55.2 ± 13.2</td>
<td>91.3 ± 10.5</td>
<td>46.0 ± 11.1</td>
<td>167.1 ± 14.4</td>
<td>91.9 ± 17.7</td>
</tr>
<tr>
<td>Radish leaves</td>
<td>273.5 ± 29.9</td>
<td>39.3 ± 6.4</td>
<td>276.9 ± 26.7</td>
<td>51.1 ± 9.4</td>
<td>481.0 ± 23.4</td>
<td>114.2 ± 15.0</td>
</tr>
<tr>
<td>and stems</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Radish roots</td>
<td>75.2 ± 12.4</td>
<td>81.1 ± 10.2</td>
<td>82.3 ± 9.4</td>
<td>85.7 ± 10.0</td>
<td>120.2 ± 12.6</td>
<td>119.9 ± 13.6</td>
</tr>
<tr>
<td>Potato tubers</td>
<td>46.9 ± 6.3</td>
<td>21.5 ± 5.4</td>
<td>45.6 ± 5.2</td>
<td>21.5 ± 6.4</td>
<td>67.5 ± 7.9</td>
<td>40.9 ± 9.1</td>
</tr>
</tbody>
</table>
TABLE II. AVERAGE CONCENTRATIONS OF $^{238}$U AND $^{234}$U (mBq/kg d.w.) IN PLANTS GROWN IN AN OPEN FIELD AND IN PLANTS GROWN IN A TENT

<table>
<thead>
<tr>
<th>Plant</th>
<th>$^{238}$U</th>
<th></th>
<th>$^{234}$U</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Field</td>
<td>Tent</td>
<td>Field</td>
<td>Tent</td>
</tr>
<tr>
<td>Alfalfa</td>
<td>82.3 ± 9.9</td>
<td>27.8 ± 4.6</td>
<td>87.0 ± 10.2</td>
<td>31.8 ± 4.9</td>
</tr>
<tr>
<td>Grass</td>
<td>88.2 ± 7.4</td>
<td>72.5 ± 9.1</td>
<td>87.2 ± 6.1</td>
<td>65.3 ± 8.5</td>
</tr>
<tr>
<td>Barley straw</td>
<td>79.1 ± 8.0</td>
<td>45.3 ± 5.2</td>
<td>81.9 ± 8.8</td>
<td>53.2 ± 7.7</td>
</tr>
<tr>
<td>Barley grain</td>
<td>24.4 ± 3.1</td>
<td>23.6 ± 3.1</td>
<td>23.1 ± 2.4</td>
<td>20.0 ± 2.7</td>
</tr>
<tr>
<td>Spinach</td>
<td>113.5 ± 15.1</td>
<td>63.2 ± 9.9</td>
<td>132.2 ± 13.3</td>
<td>63.2 ± 9.9</td>
</tr>
<tr>
<td>Radish leaves</td>
<td>338.0 ± 32.3</td>
<td>64.9 ± 10.7</td>
<td>372.6 ± 34.6</td>
<td>58.8 ± 6.4</td>
</tr>
<tr>
<td>and stems</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Radish roots</td>
<td>192.1 ± 15.7</td>
<td>78.0 ± 8.7</td>
<td>204.0 ± 19.3</td>
<td>83.2 ± 8.6</td>
</tr>
<tr>
<td>Potato tubers</td>
<td>57.3 ± 6.0</td>
<td>34.1 ± 3.3</td>
<td>63.0 ± 6.9</td>
<td>40.0 ± 4.15</td>
</tr>
</tbody>
</table>

The concentrations of thorium and uranium isotopes in the washed plants and in the plant roots are given in Tables I and II, respectively. For the majority of plants grown in the field the radionuclide concentrations were significantly higher than those of plants grown in the tent. Much higher radionuclide concentrations for the open field experiment were also found on the surface of the above-ground parts. The radionuclides on the plant surface were mainly in the form of insoluble residue and their amount usually exceeded that in the plants.

The difference between the activity concentrations in plants on the field and plants in the tent was taken as a measure of transfer via the above-ground plant parts. The ratio of this difference to the total radionuclide content under field conditions represents the contribution of the transfer via above-ground plant parts. This contribution varied from nil for thorium in radish roots to 75% for thorium in radish leaves and stems. Typically, the contribution for the above-ground plant parts and that for the roots was similar; this suggests the occurrence of translocation of thorium and uranium.

Transfer of radionuclides from soil to plants was expressed by the ratio of the radionuclide concentration in the plants to that in soil. The transfer factors (TFs) were calculated only for the tent conditions. The TFs for the two uranium isotopes were similar, ranging from 0.001 (barley grain) to 0.004 (grass). Similar TF values were found for $^{232}$Th and $^{230}$Th (from 0.0003 for grain to 0.004 for radish roots). The TF for $^{228}$Th was higher by a factor of 1.4–5 than that for $^{232}$Th.
A quantitative measurement method for monitoring the radioactive contamination of ground level air has been elaborated by the Central Laboratory for Radiological Protection. Collection of aerosols from an air volume of approximately 100 000 m$^3$ permits accurate spectrometric measurement of the radiation of natural and artificial radionuclides in a wide range of concentrations, starting at 0.5 μBq/m$^3$.

The ASS-500 type sampler is a stand-alone all-weather instrument for continuous collection of aerosols in air (Fig. 1). The Petrianov FPP-15-1.5 type filter with high collection efficiency is routinely used. The gamma and beta count rates are continuously measured for 'on-line' control of the filter activity. Infrared heaters are installed above the filter to keep it dry during the sampling period, which depends...
on the contamination of the atmospheric air. In normal situations, a weekly sampling period is routinely accepted. After sampling, the filter is removed from the filter cartridge and pressed into a cylinder of standard geometry with a diameter of 51 mm.

High resolution gamma spectrometry using a HPGe detector is applied for the quantitative measurements. For shielding purposes, the detector is placed in a low background housing so that the gamma background is reduced. Each sample is measured twice. A preliminary measurement of the sample, lasting 3000 s, is performed directly upon termination of sampling. The aim of this measurement is the detection of artificial radionuclides in air, with a lower limit of detection of about 20-50 $\mu$Bq/m$^3$. The main measurement is made two days after the end of sampling to allow decay of radon daughters.

On the basis of the main measurement, the concentration of each radionuclide in air is determined. Reports on the measurement results are prepared weekly.

IAEA-SM-339/77P

USE OF NATURAL AND MODIFIED SORBENTS TO REDUCE THE UPTAKE OF MAN-MADE RADIONUCLIDES BY FARM ANIMALS

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Central Radiation Protection and Toxicology Laboratory,
Sofia, Bulgaria

One way of obtaining food products containing reduced amounts of man-made radionuclides in the event of releases of radioactive substances to the environment is to introduce into the food cycle of farm animals substances which reduce their uptake [1, 2]. Accordingly, in order to create a technology for reducing the uptake of $^{137}$Cs by animals, we undertook the following:

(1) Investigation of the capacity of various mineral substances from natural industrially worked Bulgarian deposits (zeolites, clays, bentonites, mordenites and kieselguhrs) for sorbing man-made radionuclides from liquid media as a function of the pH and the composition of the medium, the presence of competitive ions and the time and conditions of contact [3]. When sorbent particles of less than 0.315 mm in diameter were placed in a medium containing fission and activation products, with a ratio of solid to liquid phase of 1:100, pH = 7 and a contact time of 24 h (at rest), we obtained the following mean radionuclide retention levels:

Mn-54  49.9% (from 0.0 to 94%)
Fe-59  83.7% (from 47.1 to 98.6%)
Co-60  43.8% (from 7.6 to 93.5%)
Zn-65  49.5% (from 10.3 to 96.2%)
Sr-90  71.2% (from 18.2 to 95.9%)
Cs-137  92.1% (from 77.8 to 98.5%)

Under the above model conditions the most promising agents for sorption of $^{137}\text{Cs}$ were zeolites of the clinoptilolitic type with 97.0% (from 95.4 to 98.1%), clays with 94.3% (from 93.7 to 94.8%) and bentonites with 89.5% (from 82.5 to 94.1%).

(2) Increasing the selectivity and ion exchange capacity of natural sorbents with respect to $^{137}\text{Cs}$ and $^{90}\text{Sr}$ with the aid of chemical or chemico-mechanical activation [4]. We obtained modified zeolites in which the sorption capacity was increased several fold, providing a basis for effective technological application (see Table I).

(3) Determination of the effect on uptake of $^{137}\text{Cs}$ of natural and modified ion exchangers as a function of their quantity in the fodder of experimental animals. The results, which are shown in Table II, were obtained from comparative investigations on poultry and piglets which had sorbents added to their feed (6%). It can be concluded that these sorbents could be used in the feed of farm animals in the event of radioactive contamination of the environment to reduce the content of $^{137}\text{Cs}$ in the meat by a factor of 1.66 to 2.47.

### TABLE I. CHEMICAL OR CHEMICO-MECHANICAL ACTIVATION OF ZEOLITES

<table>
<thead>
<tr>
<th>Zeolites (deposit)</th>
<th>Activation method</th>
<th>Sorption capacity (meq/g)</th>
<th>$^{137}\text{Cs}$</th>
<th>$^{90}\text{Sr}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beli Plast</td>
<td></td>
<td></td>
<td>0.95</td>
<td>0.29</td>
</tr>
<tr>
<td>Zhelezni Vrata</td>
<td></td>
<td></td>
<td>1.00</td>
<td>0.32</td>
</tr>
<tr>
<td>Zh. Vrata</td>
<td>Dry chem.</td>
<td></td>
<td>0.78</td>
<td>0.95</td>
</tr>
<tr>
<td>Zh. Vrata + B. Plast (2:1)</td>
<td>Dry chem.</td>
<td>0.95</td>
<td>0.88</td>
<td></td>
</tr>
<tr>
<td>Zh. Vrata + B. Plast (2:1) 1991</td>
<td>Dry chem.-mech. KLS-5</td>
<td>0.95</td>
<td>0.42</td>
<td></td>
</tr>
<tr>
<td>Zh. Vrata + B. Plast (2:1) 1992</td>
<td>Dry chem.-mech. KLS-5</td>
<td>0.88</td>
<td>—</td>
<td></td>
</tr>
<tr>
<td>Zh. Vrata + B. Plast (1:1)</td>
<td>Dry chem.-mech. KLS-10A</td>
<td>0.87</td>
<td>0.66</td>
<td></td>
</tr>
<tr>
<td>Zh. Vrata + B. Plast (1:1)</td>
<td>Wet chem.-mech. KLS-10AM</td>
<td>1.30</td>
<td>0.57</td>
<td></td>
</tr>
</tbody>
</table>

*Clinosorbent.*
TABLE II. EFFECT ON UPTAKE OF $^{137}$Cs OF NATURAL AND MODIFIED ION EXCHANGERS

<table>
<thead>
<tr>
<th>Type of animal</th>
<th>Type of sorbent and content</th>
<th>Reduction in uptake of $^{137}$Cs (%)</th>
<th>Factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Boiler</td>
<td>Natural zeolite, 6%</td>
<td>39.8</td>
<td>1.66</td>
</tr>
<tr>
<td>Piglet</td>
<td>Natural zeolite, 6%</td>
<td>46.5</td>
<td>1.86</td>
</tr>
<tr>
<td>Piglet</td>
<td>Mod. KLS-5, 6%</td>
<td>48.9</td>
<td>1.95</td>
</tr>
<tr>
<td>Piglet</td>
<td>Mod. KLS-10AM, 6%</td>
<td>59.5</td>
<td>2.47</td>
</tr>
</tbody>
</table>

REFERENCES

Bulgaria operates six Soviet designed nuclear power units with an installed capacity of 3538 MW(e) at the Kozloduy nuclear power plant (NPP) (four WWER-440 units, model 230, and two third-generation WWER-1000 units). The first WWER-440 unit reached power in 1974 and the last one in 1982; the two WWER-1000 units were built in 1988 and 1992. The study of environmental radioactivity in the region of Kozloduy began in 1968. The objects of the studies are the gamma ray background, atmospheric fallout, and contamination of soil, grass, milk and other foods, river water and bottom sediment. At present, radioactivity monitoring near Kozloduy is being carried out by three independent authorities: the NPP operator, the Ministry of the Environment and the Ministry of Health.

The NCRRP has carried out regular radioactivity monitoring of the environment in the region of Kozloduy since 1972; the two first years of pre-operational survey were used for collection of reference data. Samples of water and bottom sediments of the Danube and other rivers, and samples of soil and grass are being collected three times a year at 13 control points, located at distances of 6–90 km from the NPP. For determination of man-made radionuclides in environmental samples, measurements of total beta activity, high-resolution gamma ray spectrometry, and radiochemical separation of strontium and caesium are applied. The analytical quality is verified by regular participation of the NCRRP in international intercomparisons performed by the World Health Organization and the IAEA.

The main results for the last 13 years (1982–1994, since the start-up of the fourth WWER-440) have been presented as one-year mean values of the $^{90}$Sr and $^{137}$Cs content for the Danube (four sampling points), for other rivers (four points), for the area < 12 km from the NPP (three points), for the 12–30 km area (three points) and for the area > 30 km from the NPP (seven points). Except for 1986, the only man-made gamma emitters detected are $^{134}$Cs and $^{137}$Cs. The results have been compared with those for the pre-operational period (1972–1974). As a rule, the corresponding values for the period 1982–1985 are lower than the results for the pre-
operational period; this can be attributed to fallout from atmospheric nuclear weapons tests in the northern hemisphere. The period after 1987 is characterized by reduction of environmental contamination from the Chernobyl accident. The latest results (after 1991) show a reduction in $^{90}\text{Sr}$ to pre-Chernobyl levels; the reduction in $^{137}\text{Cs}$ is much slower (it is more rapid in the aquatic ecosystem than in the terrestrial one) and varies over a wide range. No distinct differences between the results for areas near the NPP and areas far from the NPP have been observed for the whole period.

Generally, it can be concluded that radioactive effluents and atmospheric releases from the Kozloduy NPP have no impact on the environmental radioactivity of the region caused by global fallout and by the Chernobyl accident.

IAEA-SM-339/89P

DETERMINATION OF $^{90}\text{Sr}$ AND $^{239,240}\text{Pu}$ IN ENVIRONMENTAL SAMPLES FROM THE SURROUNDINGS OF THE JASLOVSKÉ BOHUNICE NUCLEAR POWER PLANT

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Comenius University,
Bratislava

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Jaslovské Bohunice,
Trnava
Slovakia

Solvent extraction methods for separation of $^{90}\text{Sr}$ and $^{239,240}\text{Pu}$ have been developed for rapid and reproducible determination [1, 2] (Fig. 1). Since 1992, the methods have been applied for systematic measurements of both radionuclides in environmental samples from the surroundings of the nuclear power plant (NPP) Jaslovské Bohunice and evaluated with other radionuclides determined by gamma measurement.
Soil, sediment, vegetation

Posters: Ashing

Add tracers ($^{236}$Pu, $^{243}$Am) and carriers (Ba, Pb, Sr, Y)

Acid leaching with 8M HNO$_3$-H$_2$O$_2$ (autoclave, 180°C)

Adjustment of the plutonium oxidation state (NaN$_2$)

Evaporation

Dissolution in HNO$_3$

Extraction with TBP

Organic Y Aqueous phase

Stripping with H$_2$C$_2$O$_4$-HF (HCl-HF)

Precipitation of oxalates ($\text{pH} = 2-3$)

Beta counting of $^{90}$Y ($^{90}$Sr)

Organic Pu Aqueous phase

Stripping with $\text{H}_2\text{C}_2\text{O}_4$-HF (HCl-HF)

Microprecipitation with NdF$_3$

for alpha spectrometry

Alpha spectrometry of Pu

FIG. 1. Separation scheme.

Environmental samples have been collected at different locations close to the NPP and from the Manivier waste water channel, which enters the Dudváh river 5.2 km away from the NPP. We have analysed inorganic and organic samples (soils, sediments, aerosols, grass, clover, corn, wheat, barley, sugar beet, water plants) from different sites (Table I).

The results of measurements of $^{90}$Sr and $^{239,240}$Pu in environmental samples over two years have shown that the radioactivity of the samples is low (Table II).
<table>
<thead>
<tr>
<th>Samples</th>
<th>Site</th>
<th>Frequency of sampling per year</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aerosols</td>
<td>NPP</td>
<td>12</td>
</tr>
<tr>
<td>Fallout</td>
<td>NPP</td>
<td>12</td>
</tr>
<tr>
<td>Soil</td>
<td>NPP, V. Kostolany, Žlkovce, Kášovce, Trnava</td>
<td>1</td>
</tr>
<tr>
<td>Grass, clover</td>
<td>From ten sites</td>
<td>2</td>
</tr>
<tr>
<td>Sediments</td>
<td>Recipient (Žlkovce), Dudváh river (Bučany, V. Kostolany), Váh river (Sered)</td>
<td>1</td>
</tr>
<tr>
<td>Water plants</td>
<td>Bučany, V. Kostolany</td>
<td>1</td>
</tr>
</tbody>
</table>
TABLE II. RESULTS OF THE DETERMINATION OF $^{90}\text{Sr}$ AND $^{239,240}\text{Pu}$

<table>
<thead>
<tr>
<th>Samples</th>
<th>Depth (cm)</th>
<th>Ash (g)</th>
<th>$^{239,240}\text{Pu}$ (Bq/kg)</th>
<th>$^{137}\text{Cs}$ (Bq/kg)</th>
<th>Ratio $^{239,240}\text{Pu}/^{137}\text{Cs}$</th>
<th>$^{90}\text{Sr}$ (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Sediments</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>V. Kostolany</td>
<td>0-5</td>
<td>20</td>
<td>0.28 ± 0.08</td>
<td>15.7 ± 0.6</td>
<td>1.8 x 10$^{-2}$</td>
<td>&lt;1.2</td>
</tr>
<tr>
<td>V. Kostolany</td>
<td>0-5</td>
<td>20</td>
<td>0.21 ± 0.03</td>
<td>25.7 ± 0.4</td>
<td>8.2 x 10$^{-3}$</td>
<td>7.2 ± 1.0</td>
</tr>
<tr>
<td>Žlkovce</td>
<td>0-5</td>
<td>20</td>
<td>0.28 ± 0.05</td>
<td>45.7 ± 1.9</td>
<td>6.1 x 10$^{-3}$</td>
<td>&lt;1.2</td>
</tr>
<tr>
<td>Bučany</td>
<td>0-5</td>
<td>20</td>
<td>0.43 ± 0.05</td>
<td>221.8 ± 8.9</td>
<td>1.9 x 10$^{-3}$</td>
<td>2.9 ± 0.8</td>
</tr>
<tr>
<td>Sered</td>
<td>0-5</td>
<td>20</td>
<td>&lt;0.11</td>
<td>45.9 ± 1.9</td>
<td></td>
<td>4.8 ± 1.0</td>
</tr>
<tr>
<td><strong>Water plants</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bučany</td>
<td>20</td>
<td></td>
<td>0.43 ± 0.05</td>
<td>108.0 ± 4.36</td>
<td>4.0 x 10$^{-3}$</td>
<td>0.7 ± 0.3</td>
</tr>
<tr>
<td>V. Kostolany</td>
<td>20</td>
<td></td>
<td>&lt;0.11</td>
<td>2.6 ± 0.18</td>
<td></td>
<td>&lt;4.9</td>
</tr>
<tr>
<td>Samples</td>
<td>Depth (cm)</td>
<td>Ash (g)</td>
<td>$^{239,240}\text{Pu}$ (Bq/kg)</td>
<td>$^{137}\text{Cs}$ (Bq/kg)</td>
<td>Ratio $^{239,240}\text{Pu} / ^{137}\text{Cs}$</td>
<td>$^{90}\text{Sr}$ (Bq/kg)</td>
</tr>
<tr>
<td>-------------</td>
<td>------------</td>
<td>---------</td>
<td>-------------------------------</td>
<td>--------------------------</td>
<td>---------------------------------------------</td>
<td>--------------------------</td>
</tr>
<tr>
<td><strong>Soils</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Žlkovce</td>
<td>0–5</td>
<td>20</td>
<td>0.22 ± 0.06</td>
<td>30.8 ± 1.3</td>
<td>7.1 $\times 10^{-3}$</td>
<td>&lt;1.2</td>
</tr>
<tr>
<td>V. Kostolany</td>
<td>0–5</td>
<td>20</td>
<td>0.11 ± 0.04</td>
<td>11.1 ± 0.48</td>
<td>9.9 $\times 10^{-3}$</td>
<td>&lt;1.2</td>
</tr>
<tr>
<td>Radosovec</td>
<td>0–5</td>
<td>20</td>
<td>0.16 ± 0.02</td>
<td>10.9 ± 0.29</td>
<td>1.5 $\times 10^{-2}$</td>
<td>2.4 ± 0.6</td>
</tr>
<tr>
<td>Pecenady</td>
<td>0–5</td>
<td>20</td>
<td>0.31 ± 0.08</td>
<td>10.6 ± 0.24</td>
<td>2.9 $\times 10^{-2}$</td>
<td>&lt;1.2</td>
</tr>
<tr>
<td>Ninná</td>
<td>0–5</td>
<td>20</td>
<td>0.23 ± 0.07</td>
<td>9.9 ± 0.25</td>
<td>2.3 $\times 10^{-2}$</td>
<td>1.6 ± 0.5</td>
</tr>
<tr>
<td>Kátlovce</td>
<td>0–5</td>
<td>20</td>
<td>0.29 ± 0.05</td>
<td>7.7 ± 0.38</td>
<td>3.8 $\times 10^{-2}$</td>
<td>&lt;1.2</td>
</tr>
<tr>
<td>Krakovany</td>
<td>0–5</td>
<td>20</td>
<td>0.14 ± 0.04</td>
<td>12.7 ± 0.57</td>
<td>1.1 $\times 10^{-2}$</td>
<td>&lt;1.2</td>
</tr>
<tr>
<td>Žlkovce</td>
<td>0–2</td>
<td>20</td>
<td>0.22 ± 0.03</td>
<td>30.8 ± 1.3</td>
<td>7.4 $\times 10^{-2}$</td>
<td>1.6 ± 0.9</td>
</tr>
<tr>
<td>NPP</td>
<td>0–2</td>
<td>20</td>
<td>&lt;0.18</td>
<td>29.8 ± 1.2</td>
<td>—</td>
<td>2.5 ± 0.5</td>
</tr>
<tr>
<td>Trnava</td>
<td>0–2</td>
<td>20</td>
<td>0.15 ± 0.03</td>
<td>25.7 ± 0.4</td>
<td>5.8 $\times 10^{-3}$</td>
<td>5.7 ± 0.6</td>
</tr>
</tbody>
</table>
TABLE III. TYPICAL VALUES FOR DIFFERENT GROUPS

<table>
<thead>
<tr>
<th></th>
<th>LDA $^{90}\text{Sr}$</th>
<th>Measured value (min.-max.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aerosols</td>
<td>930 nBq/m$^3$</td>
<td>930–1460 nBq/m$^3$</td>
</tr>
<tr>
<td>Soils, sediments</td>
<td>1.2 Bq/kg</td>
<td>1.6–5.7 Bq/kg</td>
</tr>
<tr>
<td>Fallout</td>
<td>99 mBq/m$^2$</td>
<td>99–110 mBq/m$^2$</td>
</tr>
<tr>
<td>Grass, clover, water plants</td>
<td>0.3 Bq/kg</td>
<td>0.35–3.7 Bq/kg</td>
</tr>
<tr>
<td>Corn, maize, sugar beet</td>
<td>0.17 Bq/kg</td>
<td>0.17–25.2 Bq/kg</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>LDA $^{239,240}\text{Pu}$</th>
<th>Measured value (min.-max.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aerosols</td>
<td>11.5 nBq/m$^3$</td>
<td>11.7–42.9 nBq/m$^3$</td>
</tr>
<tr>
<td>Soils, sediments</td>
<td>0.31 Bq/kg</td>
<td>0.31–0.43 Bq/kg</td>
</tr>
<tr>
<td>Fallout</td>
<td>5.9 mBq/m$^2$</td>
<td>5.9–15.9 mBq/m$^2$</td>
</tr>
<tr>
<td>Grass, clover, water plants</td>
<td>9.1 mBq/kg</td>
<td>9.3–19.2 mBq/kg</td>
</tr>
<tr>
<td>Corn, maize, sugar beet</td>
<td>2.7 mBq/kg</td>
<td>2.9–3.5 mBq/kg</td>
</tr>
</tbody>
</table>

The occurrence of these radionuclides in soils, sediments and water plants can be explained by global fallout and by the impact of releases from the previous NPP A-1, which was closed in 1979. The values of radioactivity in aerosols and fallout were so low that they were below the detection limit and were detected only exceptionally. The radioactivity of the food samples did not reach the detection limit. Among all environmental samples collected, those from the Manivier channel exhibited the highest levels of radioactivity. The values for $^{239,240}\text{Pu}$ were correlated with those for $^{137}\text{Cs}$ (see Table III).

REFERENCES


The Nuclear Regulatory Authority (NRA) of Slovakia has no official documents, such as guides or practices, for the evaluation of the environmental impact of releases from nuclear facilities. The general formulation of laws and regulations has forced the NRA to reconsider them. Three examples of NRA activities regarding the impact of releases are presented below.

(1) According to the basic Act on State Supervision of the Nuclear Safety of Nuclear Facilities (1984), the NRA has approved 'limits and conditions' for discharges from nuclear power plants (NPPs). They are formulated in the Pre-operational Safety Report, which is a basic document for licensing of NPPs by the district environmental office, after approval by the NRA. Basic conditions for the limitation of discharges from NPPs were provided by national radiation protection bodies: the maximum equivalent dose to individuals must be less than 200 $\mu$Sv/a from discharges to the atmosphere and less than 50 $\mu$Sv/a from liquid releases.

By analysis of source terms, the values of releases were estimated. The equivalent dose was calculated from those values, according to a normative-technical document [1] that is valid in Member States of the former Interatomenergo (Association of east- and middle-European countries in the field of nuclear energy). Release limits were verified by comparison of the results with the above mentioned basic conditions.

According to a decision of the former Czechoslovak Atomic Energy Commission, NPP operators have evaluated the consequences of actual releases from NPPs and the values of the equivalent dose to individuals by utilizing the standardized computer code RDOJE [2]. The values for individuals belonging to the critical group of the population around NPPs (Jaslovské Bohunice, PWR, 4 $\times$ 440 MW, and the decommissioned HWGCR, 150 MW) are shown in Table I.

At present, the NRA is preparing documents with the required limits of NPP releases according to new metrological rules and practices in industrialized countries.

(2) The banks of the river Dudváh and the discharge channel of Jaslovské Bohunice were contaminated several years ago. The soils were contaminated mainly by $^{137}$Cs and $^{90}$Sr, but $^{60}$Co and $^{239,240}$Pu were also determined [3]. The maximum activity of $^{137}$Cs is approximately 20 kBq/kg, and the existence of 'hot spots' along
TABLE I. EFFECTIVE EQUIVALENT DOSES DUE TO RELEASES FROM JASLOVSKÉ BOHUNICE IN 1992

<table>
<thead>
<tr>
<th>Age of persons</th>
<th>IDE$^a$ (µSv)</th>
<th>IDE for atmospheric releases (µSv)</th>
<th>IDE for liquid releases (µSv)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0-1</td>
<td>1.24</td>
<td>0.40</td>
<td>0.84</td>
</tr>
<tr>
<td>1-8</td>
<td>1.10</td>
<td>0.34</td>
<td>0.76</td>
</tr>
<tr>
<td>8-12</td>
<td>0.90</td>
<td>0.27</td>
<td>0.63</td>
</tr>
<tr>
<td>12-17</td>
<td>0.71</td>
<td>0.26</td>
<td>0.45</td>
</tr>
<tr>
<td>Adults</td>
<td>0.36</td>
<td>0.15</td>
<td>0.21</td>
</tr>
</tbody>
</table>

$^a$ Individual dose equivalent.

the discharge channel (250 kBq/kg) has been demonstrated. From a detailed investigation, the total activity of $^{137}$Cs has been estimated at 110 GBq and that of $^{90}$Sr at 1.8 GBq [4]; 90% of the activity was found in the 20 cm soil profile. In spite of the fact that the calculated equivalent dose for individuals did not exceed 1 µSv/a, it was decided to remove about 4300 m$^3$ soil and to put it into an isolated concrete basin. The consequences of a 'maximum accident' (loss of quality of isolating barriers; loss of retention quality of soils, etc.) were evaluated and a dose of 0.08 µSv/a from the critical radionuclide $^{90}$Sr was correlated.

(3) At present, the NRA is involved in licensing of the low and intermediate level radwaste disposal facility at Mochovce. Basic equivalent dose limits have been given for performance analysis and for control of the operation of the facility: (a) limit for 'releases' (drainage water): equivalent dose to individuals from the critical group: 50 µSv/a; and (b) limits for performance analysis: 100 µSv/a for normal operation and 250 µSv/a for intrusive operation.

A monitoring programme for checking the operation of the disposal facility has been proposed by the facility operator; this is being reviewed by the NRA.

REFERENCES

High activity wastes from the radiochemical production facility at the Mayak nuclear industrial complex were continuously released into the river Techa from 1949 to 1956. The volume of effluent was 76 million m$^3$ with a total activity of 2.76 million Ci.\(^1\) Approximately 95\% of the total activity was released during the period from March 1950 to November 1951, when the average daily release was 4300 Ci/d, with almost 25\% of the activity consisting of the long lived radionuclides $^{90}$Sr and $^{137}$Cs. Restriction of the releases in 1951 did not lead to a marked reduction in the concentration of radionuclides in the river water. Bottom sediments and the flood bed of the river in the upper reaches had become strong sources of secondary contamination. Since 1964, after the construction of the last dam, the river Techa has been going through a process of natural decontamination.

An integrated radioecological study of the rivers Techa and Iset is taking place in the Kurgan region. Sampling has been carried out at ten main measuring stations of the rivers Techa, Iset and Miass. The location of the latter has been selected so as, firstly, to isolate pollutants entering the river's ecosystem from the neighbouring regions (Sverdlovsk and Chelyabinsk) and, secondly, to determine how the state of the river Iset is influenced by its main Techa and Miass tributaries in the Kurgan region. Samples of water, bottom sediments and river vegetation were taken at each measuring station. Samples of bottom sediments were taken at 15 additional measuring stations of the river Techa and water samples were taken at another 12 stations.

\(^1\) 1 Ci = 37 GBq.
The $^{90}$Sr concentration in the water of the rivers Techa, Iset and Miass is within the range 0.12–9.1 Bq/L. The concentration of $^{239}$Pu in all water samples is less than 0.005 Bq/L. The $^{90}$Sr concentration in the water of the Iset along its entire length is much higher after the confluence with the Techa than before the confluence. The $^{137}$Cs content is between 0.0013 and 0.31 Bq/L.

The $^{137}$Cs concentration in the samples of water vegetation from the Iset are within the range of 4–15 Bq/kg. Before the confluence of the Iset with the Techa, the $^{90}$Sr content in various species is within the range 7.8–22 Bq/kg. After the confluence, the $^{90}$Sr content in those species is within the range 113–965 Bq/kg.

The $^{137}$Cs content in cladophora from the Miass is two to three times higher than that in cladophora from the Iset, which may indicate higher medium term concentrations of this element in the water of the Miass compared to those in the Iset. At the same time, the $^{90}$Sr concentration in fennel pondweed from the upstream measuring station of the Miass and that of the Iset are comparable (12 and 21 Bq/kg).

Macrophytes in the Techa have the highest $^{137}$Cs content (200–350 Bq/kg) and $^{90}$Sr content (1000–2000 Bq/kg) compared to macrophytes from other rivers. This is due to the high activity levels of bottom sediments in the Techa and the relatively high concentration of radionuclides in the water of the Techa.

The $^{137}$Cs content in fish from the Techa, Miass and Iset was analysed and the highest concentrations were found in fish from the Techa, namely 26.48 Bq/kg (6.83 Bq/kg in the bones).

The $^{137}$Cs concentration in bottom sediments from the Iset does not vary much along the river and is within the range 1.2–10 Bq/kg. The $^{90}$Sr content in the same samples exceeds the $^{137}$Cs concentration in many cases by a factor of ten and varies from 14 to 60 Bq/kg. The same applies to the Miass bottom sediments. Their $^{137}$Cs content ranges up to 10 Bq/kg, and the $^{90}$Sr content up to 50 Bq/kg.

In the bottom sediments of the Techa the $^{137}$Cs content varies from 120 to 905 Bq/kg, and in the majority of cases it slightly exceeds the $^{90}$Sr content in the same samples, except for the last 5–10 km.

Soil samples were taken at 5 cm intervals down to a depth of 30 cm, and the area of each soil sample was determined. In the majority of cases the main radionuclide content was in the first 5–10 cm. The $^{90}$Sr content of the samples varied from 100 to 2000 Bq/kg. The $^{137}$Cs content was in the range from several hundred to 1500 Bq/kg.

The $^{137}$Cs contamination level of the Techa, which is $3 \text{ Ci/km}^2$, taken over its course as a whole, drops to $1 \text{ Ci/km}^2$ at the border of the Kurgan and Chelyabinsk regions. In the upper reaches, the $^{90}$Sr contamination amounts to $2.1 \text{ Ci/km}^2$. Near the confluence of the Techa and the Iset, the contamination level reaches $11.7 \text{ Ci/km}^2$.

Different plant species accumulate $^{90}$Sr differently. The biological transfer coefficient for nettles is 12–17 and the $^{90}$Sr concentration in nettles is 6000–7000 Bq/kg. For coltsfoot the biological transfer coefficient is 0.2–0.3 and the $^{90}$Sr concentration
is 90–120 Bq/kg. The biological transfer coefficient for $^{137}$Cs in various other species is 0.01–0.03.

As in the case of $^{137}$Cs content in bottom sediments, the variation in the exposure rate in the flood bed of the Techa along the course of the river is well described by the decreasing exponential function:

$$C_g = \exp (3.75 - 0.012x)$$

where $C_g$ is the gamma dose rate.

A study has been made of the different species and the most important biological characteristics of the main groups of aquatic organisms which ensure the functioning of the river ecosystems and influence the water quality. The ichthyofauna was examined to assess the potential of the waters for fishing use. A comparative analysis of biological indicators for various groups of aquatic organisms was performed to assess the living conditions in the rivers and to determine the nature and extent of the effect of the contaminated environment on biological organisms. The parts of the river ecosystems which are the most resistant to industrial contamination were identified.

The hydrogeological and hydrochemical conditions of the area were studied. The characteristics of the hydrological regime as well as the hydraulic and morphometric characteristics of the rivers and the river beds were determined. The average long term and minimum water discharges were assessed with a 95% confidence level for the reference points and monitored sections.

Studies were made of the water regulating capacity of forests, and of the aquaphysical drainage regulating capacities of soils in the valleys of the Techa, Iset and Miass.
Permanent monitoring of radiation exposure and health surveillance have been performed in the Russian Arctic, from the Kola peninsula to Chukotka, at the request of the Ministry of Health and the State Committee on Sanitary and Epidemiological Surveillance. All factors of the food-chain of reindeer breeders in the Arctic have been studied: the contents of radionuclides \( {^{137}}Cs, {^{90}}Sr, Po-Pb \) and others) in soil, lichens, reindeer meat and fish, and the contents in the human body (see Fig. 1). Environmental factors connected with radiation and other sources have also been evaluated. The average values of \({^{137}}Cs\) in the food-chain are presented in Table I.

On the basis of the obtained data, the structure of the average individual effective dose (mSv/a) has been estimated (Table II). Whereas the radiation levels from
TABLE I. MEAN VALUES OF $^{137}$Cs (mCi) IN THE FOOD-CHAIN$^a$

<table>
<thead>
<tr>
<th>Region</th>
<th>Lichens (per kg dry weight)</th>
<th>Reindeer meat (per kg)</th>
<th>Human excrements (over 24 h)</th>
<th>Human body (per kg)</th>
<th>Dose for humans (mSv/a)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Murmansk</td>
<td>1.1</td>
<td>2.1</td>
<td>0.6</td>
<td>1.0</td>
<td>11.0</td>
</tr>
<tr>
<td>Nenetsk</td>
<td>0.8</td>
<td>1.0</td>
<td>0.2</td>
<td>0.4</td>
<td>4.4</td>
</tr>
<tr>
<td>Yamal</td>
<td>0.7</td>
<td>1.4</td>
<td>0.2</td>
<td>0.4</td>
<td>4.5</td>
</tr>
<tr>
<td>Taimir</td>
<td></td>
<td>0.8</td>
<td>0.3</td>
<td>0.5</td>
<td>5.5</td>
</tr>
<tr>
<td>Yakutia</td>
<td>0.7</td>
<td>0.5</td>
<td>0.06</td>
<td>0.15</td>
<td>1.7</td>
</tr>
<tr>
<td>Chukotka</td>
<td></td>
<td>0.7</td>
<td>0.2</td>
<td>0.4</td>
<td>4.4</td>
</tr>
<tr>
<td>Mean values</td>
<td>0.8</td>
<td>1.1</td>
<td>0.3</td>
<td>0.5</td>
<td>5.0</td>
</tr>
</tbody>
</table>

$^a$ For 1 mCi/km$^2$ and 0.5 kg lichens per square metre (1 Ci = 37 GBq).

TABLE II. STRUCTURE OF THE MEAN INDIVIDUAL EFFECTIVE DOSE (mSv/a)

<table>
<thead>
<tr>
<th>Radiation sources</th>
<th>Effective dose (external and internal radiation)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cosmic rays</td>
<td>0.30</td>
</tr>
<tr>
<td>Potassium-40</td>
<td>0.30</td>
</tr>
<tr>
<td>Uranium-238 series</td>
<td>2.85</td>
</tr>
<tr>
<td>Thorium-232 series</td>
<td>1.06</td>
</tr>
<tr>
<td>Medical exposure</td>
<td>0.50</td>
</tr>
<tr>
<td>Radioactive fallout</td>
<td>0.22–0.66</td>
</tr>
<tr>
<td>Total</td>
<td>5.23–6.07</td>
</tr>
</tbody>
</table>

Sources 1, 2 and 5 (Table II) in the various zones do not differ by more than 15%, the doses from the other sources are subject to regional variability. The most detailed information available in this regard is that for source 6 (see Fig. 1), and there are also reliable data for source 3.
FIG. 2. Mean individual effective dose from $^{137}$Cs in the food-chain (mSv/a).

The dynamics of the mean individual effective dose from $^{137}$Cs is illustrated in Fig. 2. A slight increase in $^{137}$Cs after the Chernobyl accident has been found in the eastern regions.

In general, a trend toward a decrease in the exposure levels and the effective dose from west to east and regional differences in the radiation situation have been observed, with similar changes in the mortality structure. Detailed analysis of epidemiological data indicated that the increase in cancer incidence in some population groups does not correspond to the radiation levels and could be correlated with other factors, such as severity of climate, ethnogenetic variations and dietary habits (Table III). The higher level of cancer morbidity and mortality and possibly other pathological conditions compared with Russian statistical data could not be explained by the impact of radiation (radioactive fallout). When deciding on activities to improve the health situation in the northern regions, other environmental factors should be taken into account.
TABLE III. CORRELATION COEFFICIENTS ($r$) OF MORTALITY INDICES ($X$) AND SOME ENVIRONMENTAL CHARACTERISTICS ($Y$)

<table>
<thead>
<tr>
<th>Region a</th>
<th>Average annual mortality index for 10 000 persons (1961-1982)</th>
<th>Average vegetation period (days)</th>
<th>Average contents of $^{210}$Pb in reindeer bones (Bq/kg)</th>
<th>Maximal contents of $^{137}$Cs ($10^3$ Bq)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>From all causes</td>
<td>From cancer</td>
<td></td>
<td>Human organisms</td>
</tr>
<tr>
<td>N</td>
<td>$X_1$</td>
<td>$X_2$</td>
<td>$Y_3$</td>
<td>$Y_4$</td>
</tr>
<tr>
<td>(1)</td>
<td>124</td>
<td>16</td>
<td>100</td>
<td>551</td>
</tr>
<tr>
<td>(2)</td>
<td>101</td>
<td>10</td>
<td>90</td>
<td>111</td>
</tr>
<tr>
<td>(3)</td>
<td>115</td>
<td>11</td>
<td>90</td>
<td>278</td>
</tr>
<tr>
<td>(4)</td>
<td>208</td>
<td>30</td>
<td>40</td>
<td>426</td>
</tr>
<tr>
<td>(5)</td>
<td>191</td>
<td>32</td>
<td>60</td>
<td>351</td>
</tr>
<tr>
<td>(6)</td>
<td>203</td>
<td>41</td>
<td>75</td>
<td>577</td>
</tr>
</tbody>
</table>

$X_1$: from all causes, $X_2$: from cancer.

$-0.86 \pm 0.12$, $+0.56 \pm 0.28$, $-0.67 \pm 0.22$, $-0.84 \pm 0.12$ for $X_1$.

$+0.63 \pm 0.26$, $-0.68 \pm 0.22$, $-0.35 \pm 0.34$, $-0.79 \pm 0.16$ for $X_2$.

a (1) Murmansk region, (2) Komi region, (3) Nenets region, (4) Taimir region, (5) Yakutiya region, (6) Chukotka region.
THE NEW FILM SORBENT

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Investigating radionuclide propagation at great depth in the layer adjacent to the sea-bed involves taking samples of aqueously dispersed matter under adverse conditions. The investigation is complicated by the imperfection of the existing sampling techniques. The new film sorbent (FS), developed by the authors for sampling aqueously dispersed matter, allows the efficiency of the investigation to be increased significantly.

The FS has a photo-film support the surface of which is covered with a thin layer of a new sorbent substance. When the FS is placed in a dispersed medium, it provides an equal absorption rate of particles (including colloidal particles) within the dimensional range R = 0–40 μm, regardless of their chemical composition, their phase (liquid or solid) and the polarity of their charge.

The optical transparency of the FS allows a structural (microscopic) analysis of samples to be made. When the surface of the FS is large enough, radionuclide

![Graph](graph.png)

**FIG. 1.** Kinetics of the saturation of the FS at different concentrations of the dispersed particles.
and chemical analyses of samples can also be made. Furthermore, using the characteristics of the kinetics of saturation, it is also possible to assess the particle concentration in the medium under study by measuring the time of exposure of the FS and the weight of absorbed particles (Fig. 1).

In the course of many tests conducted in a laboratory flume as well as in the sea at a depth of 2000 m, a number of new characteristics of the layer adjacent to the sea-bed were obtained. A vertical concentration gradient of the dispersed particles in the layer was found by using a vertically positioned FS in the form of a long strip.

It was discovered that the layer adjacent to the sea-bed comprises three sublayers with distinct differences in concentration. The lower sublayer \( L \) (Fig. 2), which has the properties of a solid body, is stationary and consists of fast, bound, dispersed particles with an average concentration \( C_L = 900 \text{ g/dm}^3 \). The medium sublayer \( M \), which has the properties of a viscous liquid, is semi-stationary. It consists of loosely bound particles with an average concentration \( C_M = 50 \text{ g/dm}^3 \). The upper sublayer \( U \) has properties similar to those of water and consists of unbound dispersed particles with an average concentration \( C_U = 3 \text{ g/dm}^3 \).

All three sublayers are involved in the process of radionuclide propagation. To study the role of each sublayer in this process, radionuclides of various densities in the form of dispersed particles were added to the sediment composition of the sea-bed, and radiographic analysis was performed.

As can be seen from the diagrammatic sketch of a radioautogram (Fig. 3(a)), radionuclides of low density \( (p \leq 5 \text{ g/cm}^3) \) concentrate in sublayer \( M \) in the form

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**FIG. 2.** Variations in the concentration of dispersed particles with the height of the layer adjacent to the sea-bed.
FIG. 3. Distribution of radionuclides of different densities in FS samples.

of suspension (the intensity of cross-hatching in Fig. 3 shows the degree of blackening of the X ray film after its contact with the FS sample). The suspended radioactive particles move together with sublayer M. All radionuclides of high density ($p \geq 10 \text{ g/cm}^3$) deposit on the surface of the lower sublayer L (Fig. 3(b)), but do not penetrate into it. In this case, the radioactive particles can only move if they are detached from the surface and drawn along it by the flow.

The above described characteristics of the three sublayers are not universal and correspond only to the specific state of the sea-bed sediment. They are also not constant in time and depend on a number of external variables, such as flow velocity, sea-bed roughness, turbulent flow intensity, temperature and electromagnetic radiation.

Numerous tests of the FS have proven that, despite the simplicity of its design and of the sampling technique, the FS has a large number of applications. The FS makes it possible to study dispersed systems with a wide range of particle dimensions and concentrations, to carry out sampling on a continuous in-depth basis, and to make structural, chemical and radionuclide analyses of samples.
The discovery of a sandstone uranium ore deposit at Stráž pod Ralskem in Northern Bohemia in the late 1960s resulted in extensive exploration, development and mining activities. During the first approximately 15 years, no adequate measures were adopted to reduce the environmental impact of these activities. This was due to the then valid system of exemptions from legislative regulation, for strategic reasons [1]. Because of the delayed development of appropriate water treatment measures, radionuclides and accompanying toxic heavy elements entered the river Ploučnice, resulting in extensive contamination of its flood plain sediments. The initial distribution of the contaminating substances in the sediments was also affected by the canalization of approximately 14 km of the river valley along the slope of the river, which was performed in stages between 1976 and 1987.

Only since 1989 has it been possible to determine systematically the extent of the contamination (only radioactive). The first systematic data were provided in a report [2]; these data were based on the measurement of the gamma dose rates in approximately 100 cross-sections of the flood plain; the distance between the sections was approximately 460 m. Approximately 40 km of the river valley were reported to be contaminated at different levels (see Table I).

It was no sooner than in 1990 that systematic detailed gamma dose rate measurements in regular grids of the localities with the highest hazards for the population could be started by DIAMO [3, 4]. The measured localities were selected according to the results of gamma spectrometric surveys with airborne devices. The order of measurements was determined by means of a qualitative priority assessment, based on: (1) the distance from human settlements, (2) the number of inhabitants in the immediate surroundings, (3) the ease of access, (4) the current and future utilization of the area, (5) the existence of water management and supply systems, and (6) the water level in the soils.

The first results of these surveys showed that the distribution of radionuclides is very irregular, and even the survey with airborne devices gives only a very rough picture of the radioactive contamination. An adequate display of the actual distribu-
TABLE I. AREAL EXTENT AND ESTIMATED RADIUM CONTENTS OF FLOOD PLAIN SEDIMENTS

<table>
<thead>
<tr>
<th>Range (nGy/h)</th>
<th>Average (nGy/h)</th>
<th>Area (m²)</th>
<th>$^{226}$Ra (Bq/g)</th>
<th>$^{226}$Ra (GBq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0-200</td>
<td>100</td>
<td>3,377,000</td>
<td>0.21</td>
<td>59.5</td>
</tr>
<tr>
<td>200-600</td>
<td>400</td>
<td>2,215,000</td>
<td>1.21</td>
<td>393.3</td>
</tr>
<tr>
<td>600-2500</td>
<td>1550</td>
<td>1,126,000</td>
<td>5.06</td>
<td>893.6</td>
</tr>
<tr>
<td>&gt;1500</td>
<td>50,000</td>
<td></td>
<td>22.95</td>
<td>182.8</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td></td>
<td>6,768,000</td>
<td></td>
<td>1529.2</td>
</tr>
<tr>
<td><strong>Total above background</strong></td>
<td></td>
<td>3,391,000</td>
<td></td>
<td>1469.7</td>
</tr>
</tbody>
</table>

*The background radioactivity is assumed to be about 110 nGy/h [1].*
TABLE II. COMPARISON OF CONTAMINATED AREAS ESTIMATED FROM CROSS-SECTIONS AND DETERMINED BY DIRECT DETAILED MEASUREMENTS OF GAMMA DOSES (INCLUDING BACKGROUND RADIATION)

<table>
<thead>
<tr>
<th>Contamination level (nGy/h)</th>
<th>A. Estimated from cross-sections</th>
<th>B. From detailed measurements</th>
<th>Comparison 100 B/A</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt;200</td>
<td>65 598.0 m²</td>
<td>13 362.0 m²</td>
<td>10.87%</td>
</tr>
<tr>
<td>200 &lt; D &lt; 600</td>
<td>180 960.0 m²</td>
<td>7 129.5 m²</td>
<td>1.35%</td>
</tr>
<tr>
<td>&gt;600</td>
<td>246 558.0 m²</td>
<td>2 430.5 m²</td>
<td>3.88%</td>
</tr>
<tr>
<td>&gt;200</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

in random sections of the valley. Detailed measurements of contaminated areas by means of airborne gamma spectrometry revealed that the radioactivity varies greatly over very short distances. Thus, for exploration of the area, it is necessary to perform field measurements of gamma doses in grids with node distances as short as a few metres (2-10 m).

Planned remediation work based on the methodology of Ref. [2] would require full removal and disposal of all sediments from the contaminated part of the flood plain area (about 600 ha, depth 4-12 m, in about 40 km of the river valley, i.e. more than 48 million m³; this request was actually made) or removal of the contaminated upper 50 cm layer of soil (about 600 ha, depth 50 cm, i.e. 3 million m³). The same net benefit, however, can be achieved by remediation work only in the areas with maximum concentrations of contaminants, as determined by field measurements in detailed grids. In this case, the soil volume to be removed will probably be several per cent (2-5%) of the figure given in Ref. [2].

REFERENCES


The knowledge acquired after the Chernobyl accident has shown the need to improve the quantification of some parameters affecting radionuclide transfer along the food-chain. In order to obtain reliable values for different soil-plant systems, four experiments were designed in a joint project (‘Transfer of accidentally released radionuclides in agricultural systems — TARRAS’), supported by the radiation protection programme of the European Commission.

These experiments were based on the deposition of a thermogenerated aerosol on bare soils (sandy loam and sandy soils) which had been sown with lettuce and pea
TABLE I. TRANSFER COEFFICIENTS FOR THE SOIL–PLANT SYSTEMS STUDIED

<table>
<thead>
<tr>
<th>Soil</th>
<th>Growth stage</th>
<th>Transfer coefficients</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>kg soil/kg plant (d.w.)</td>
<td>m² soil/kg plant (d.w.)</td>
</tr>
<tr>
<td>Cs-134</td>
<td>Lettuce</td>
<td>Sandy loam Seedling</td>
<td>0.705 ± 0.374</td>
<td>0.006 ± 0.003</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Mature</td>
<td>0.199 ± 0.120</td>
<td>0.002 ± 0.001</td>
</tr>
<tr>
<td></td>
<td>Sandy</td>
<td>Seedling</td>
<td>3.603 ± 2.992</td>
<td>0.026 ± 0.020</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Mature</td>
<td>1.407 ± 1.331</td>
<td>0.010 ± 0.009</td>
</tr>
<tr>
<td>Pea plant</td>
<td>Sandy loam</td>
<td>Seedling</td>
<td>0.140 ± 0.094</td>
<td>0.0008 ± 0.0005</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Leaves + stems</td>
<td>0.595 ± 0.133</td>
<td>0.0042 ± 0.0013</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Fruit</td>
<td>0.087 ± 0.037</td>
<td>0.0006 ± 0.0002</td>
</tr>
<tr>
<td></td>
<td>Sandy</td>
<td>Seedling</td>
<td>1.069 ± 0.577</td>
<td>0.0051 ± 0.0025</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Leaves + stems</td>
<td>0.963 ± 0.511</td>
<td>0.0051 ± 0.0029</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Fruit</td>
<td>0.311 ± 0.110</td>
<td>0.0016 ± 0.0006</td>
</tr>
<tr>
<td>Sr-85</td>
<td>Lettuce</td>
<td>Sandy loam Seedling</td>
<td>7.499 ± 2.539</td>
<td>0.063 ± 0.026</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Mature</td>
<td>7.338 ± 1.577</td>
<td>0.061 ± 0.015</td>
</tr>
<tr>
<td></td>
<td>Sandy</td>
<td>Seedling</td>
<td>40.57 ± 32.28</td>
<td>0.288 ± 0.202</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Mature</td>
<td>26.78 ± 18.20</td>
<td>0.204 ± 0.160</td>
</tr>
<tr>
<td>Pea plant</td>
<td>Sandy loam</td>
<td>Seedling</td>
<td>1.134 ± 0.251</td>
<td>0.0062 ± 0.0015</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Leaves + stems</td>
<td>3.082 ± 1.089</td>
<td>0.0212 ± 0.0052</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Fruit</td>
<td>0.533 ± 0.296</td>
<td>0.0036 ± 0.0016</td>
</tr>
<tr>
<td></td>
<td>Sandy</td>
<td>Seedling</td>
<td>22.777 ± 9.717</td>
<td>0.1119 ± 0.0447</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Leaves + stems</td>
<td>72.776 ± 21.619</td>
<td>0.3922 ± 0.0859</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Fruit</td>
<td>2.180 ± 0.802</td>
<td>0.0118 ± 0.0034</td>
</tr>
<tr>
<td>Ag-110m</td>
<td>Lettuce</td>
<td>Sandy loam Seedling</td>
<td>1.869 ± 0.876</td>
<td>0.015 ± 0.006</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Mature</td>
<td>0.274 ± 0.184</td>
<td>0.002 ± 0.002</td>
</tr>
<tr>
<td></td>
<td>Sandy</td>
<td>Seedling</td>
<td>7.837 ± 5.925</td>
<td>0.056 ± 0.038</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Mature</td>
<td>0.948 ± 0.402</td>
<td>0.007 ± 0.004</td>
</tr>
<tr>
<td>Pea plant</td>
<td>Sandy loam</td>
<td>Seedling</td>
<td>0.109 ± 0.059</td>
<td>0.0006 ± 0.0003</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Leaves + stems</td>
<td>0.444 ± 0.101</td>
<td>0.0031 ± 0.0006</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Fruit</td>
<td>0.517 ± 0.272</td>
<td>0.0035 ± 0.0017</td>
</tr>
<tr>
<td></td>
<td>Sandy</td>
<td>Seedling</td>
<td>0.506 ± 0.227</td>
<td>0.0024 ± 0.0009</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Leaves + stems</td>
<td>1.217 ± 0.417</td>
<td>0.0066 ± 0.0027</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Fruit</td>
<td>0.279 ± 0.157</td>
<td>0.0014 ± 0.0006</td>
</tr>
</tbody>
</table>
POSTER SESSION 1

plant seeds. The soils are described elsewhere [1]. The aerosol simulated an accidental source term and contained $^{134}$Cs, $^{85}$Sr and $^{110}$Ag [2]. Following contamination processes, vegetation samples were collected at two stages of plant growth: seedling stage and mature plant stage. In the case of pea plants, fruits and leaves plus stems were sampled separately at the mature stage. Soil samples were taken for the complete profile in each sampling of vegetation. This design permitted the study of the influence of several parameters, such as soil properties, type of plant, growth stage or radionuclide in soil to plant transfer.

Soil to plant transfer coefficients are usually calculated from the ratio between the specific activities of plants and soil. Transfer of radionuclides may also be expressed as the ratio between plant specific activities and soil activity, expressed in surface unities. Furthermore, either the total activity of soils or the activity of the available radionuclide fraction (obtained by extraction of MgCl$_2$, 1 mol/L) may be used for calculations. The way of expressing radionuclide transfer is of paramount importance, as the values obtained will hardly be comparable and could even lead to different conclusions.

Table I shows transfer coefficients for lettuce and pea plants. In general, the results agree with those obtained by the authors, using samples contaminated by soluble or ‘fallout’ radionuclides [4]. Nevertheless, there is a wide range of transfer values in the literature.

The transfer increased with growth stage of pea plants, whereas a decrease in transfer was observed for lettuce. This behaviour can be explained by the different strategies of nutrient uptake in each case. In pea plants, radionuclide transfer to the fruits was always much lower than that to leaves and stems.

Radionuclide transfer was always higher in sandy soil than in sandy loam soil, as could be expected because of the soil properties. When the transfer coefficients were calculated using the total soil specific activity, the values obtained for Cs and Ag were lower than those for Sr. However, when the transfer coefficients were calculated using radionuclide exchangeable activity, higher values were obtained for Cs, especially in sandy loam soil. To explain this behaviour, the exchangeable status of Ca and K in the soil should be taken into account [5].

REFERENCES


Radionuclide partitioning and downward migration in soils, as well as the nutrient status in the soil solution (mainly Ca, Mg, K and NH$_4$) are the most significant factors to be used in predicting and explaining radionuclide soil to plant transfer factors [1, 2]. Migration patterns may give information on radionuclide solubility, although in some cases this information is not significant, considering the ploughing practices by which the top 20 cm layer is homogenized. Besides, the dynamics of radionuclide distribution in soils, the study of which allows the changes with time of available and fixed fractions to be evaluated, may become a key factor in explaining differences with time of soil to plant transfer factors.

A qualitative and operational approach, based on radionuclide desorption using sequential extractions, may be applied in order to obtain information on distribution dynamics [3, 4]. Despite the relationship between desorption yields and experimental conditions, such methods are useful tools for studying the time dynamics of radionuclide partitioning and predicting the long term behaviour of radionuclides.

The scheme provides for sequential application of the following extractant reagents: H$_2$O, NH$_4$Cl or KCl or MgCl$_2$, HAcO, NH$_2$OH·HCl and H$_2$O$_2$-NH$_4$AcO. From sequential extraction, it is possible to define several radionuclide fractions associated with different soil phases, the most significant ones being the exchangeable radionuclide fraction (associated with plant uptake of available radionuclides) and the radionuclide fraction related to mineral soil phases (fixed radionuclide).

Radionuclide migration and ageing has been studied in two Mediterranean agricultural soils, in the frame of a collaborative project called 'Transfer of accidentally released radionuclides in agricultural systems — TARRAS', supported by the
radiation protection programme of the European Commission. One type of soil is a sandy loam soil (SL), from the experimental fields of the University of Barcelona (Spain); the other type is a sandy soil (S), from Belleville (Aix-en-Provence, France). The main characteristics of both types of soil have been determined (summarized in Ref. [5]). Both types of soil have a low organic matter content (SL 2.4% and S 0.2%) and a relatively high illite proportion in the clay fraction (SL 79.0% and S 65.4%). The cation exchange capacity is lower in sandy soil, which is due mainly to its high coarse sand fraction (SL 36.8% and S 86.2%).

The soil samples were placed in lysimeters of 50 cm × 40 cm × 25 cm and subsequently contaminated using a thermogenerated aerosol containing $^{134}$Cs, $^{85}$Sr and $^{110}$Ag$^m$ [6]. One day after contamination, an area of 10 cm × 10 cm was sampled, to a depth of 2 cm, which allows the 'zero time' stage to be defined. After sampling, the lysimeters were kept at room temperature and manually irrigated (3.75 L/m$^2$, every ten days). Subsequently, different samples were taken, to the depth of the complete soil profile, divided into different layers.

To study radionuclide ageing, a sequential extraction scheme has been applied to all samples from the different sampling stages (one day, three months and nine months after contamination) in order to evaluate changes with time and to study the possibility of reaching a distribution that is in 'pseudo-equilibrium' and for which no further significant variation can be expected.

FIG. 1. Exchangeable fractions of radionuclides in sandy soil and sandy loam soil.
Figure 1 shows the exchangeable fractions of the three radionuclides in sandy soil and sandy loam soil, obtained from samples from the initial stage. The extractant reagent used to predict these fractions and to distinguish radionuclide solubility in both soils, as well as the different desorption yields obtained for the radionuclides are the main factors.

The results obtained are expected to be used in evaluations of the transfer factors calculated for plants growing in the soils studied, during a period similar to the one used for the ageing studies.

REFERENCES

ASSESSMENT OF THE ACTIVITY CONCENTRATION OF HT AND HTO IN AIR

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Long term measurements of HT and HTO in the open air have been made at different sites in the Vienna area. Such measurements have also been carried out (for about 40 weeks) in the air of the hall of the TRIGA research reactor in Vienna. The measuring equipment has been tested and the necessary modifications have been made. The results obtained complement routine measurements of HTO in precipitation. The origin of some unidentifiable increases in the activity concentration has been investigated.

The measuring technique is based on the adsorption of HTO on charcoal, subsequent catalysis of HT to HTO and desorption. Figure 1 is a schematic diagram of selective sample preparation. After desorption, the samples are counted by liquid scintillation counting.

Results of the measurements of HT and HTO in the air on the hall of the TRIGA reactor are presented below. Applying data from the International Commission on Radiological Protection for dose estimation, annual doses of 7 pSv for HT and 600 nSv for HTO have been obtained (see Fig. 2).

\begin{center}
\includegraphics[width=\textwidth]{fig1}
\end{center}

\textit{FIG. 1. Schematic diagram of selective sample preparation.}
**FIG. 2.** Activity concentration of HTO in the air of the TRIGA reactor hall and reactor operation time per week.

**FIG. 3.** Activity concentrations of HT and HTO in the open air.
Measurements of HT and HTO in the open air are being continued; Fig. 3 shows recent results. The annual doses due to inhalation are estimated as being of the order of 0.2 pSv for HT and 4 nSv for HTO. These doses are well below the limits. It is pointed out that additional measuring equipment should be installed at different sites in order to identify releases of HT.

MODELLING OF NORMALIZED CONSEQUENCES OF $^{137}$Cs DEPOSITION IN VARIOUS AQUATIC ENVIRONMENTS

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L. MONTE
ENAE, CRE Casaccia,
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A compartmental modelling approach for several European freshwater ecosystems has been used to predict dynamically the concentrations of $^{137}$Cs in lake water and in food-chains. The predicted doses from $^{137}$Cs depositions have been further normalized, i.e. converted to dose per equal deposition for each freshwater environment considered. Improved confidence in the model predictions has been achieved through comparisons with the results of measurements carried out in several countries after the accident at the Chernobyl reactor.
FIG. 1. Conceptual model used.

Conceptually, the model used for freshwater ecosystems accounts for the drainage area, the lake reservoir, the sediments and the dynamic fish-chain connected with the lake reservoir (Fig. 1). Transfer of $^{137}$Cs is modelled by calculating the partition between solids and water and by using the known mass rates for estimating the total transfer rates. The conceptual model accounts for the primary source term, i.e. deposition on lake water, and the secondary source term, i.e. leaching from the drainage area after a certain time. The dynamic fish model is based on a realistic description of the chain non-predatory fish — intermediate — predatory fish. Estimation of the relative amounts of plankton and different types of fish populations and their living habits has provided an opportunity to follow the activity rates of this important aquatic food-chain.

Different types of lake environments, located in southern, central and northern parts of Europe, have been modelled and analysed in the study. The modelling study was part of an exercise in the international VAMP programme co-ordinated by the IAEA [1]. The numerical calculations have been performed by employing the computer code DETRA of VTT Energy.

The specific features of each lake environment are accounted for by the data used in the calculations of the model predictions. Therefore, drainage areas, precipitation rates, water exchange rates of lakes, sedimentation rates, trophic levels of lakes, etc., are relevant to the specific lake considered.
The objective of this study was to investigate the normalized consequences for the lakes analysed. After the Chernobyl reactor accident, various lake environments were exposed to different $^{137}$Cs depositions. However, the consequences can be normalized, e.g. per equal deposition, to see whether there is any consistency between the results for the different lake environments. The temporal behaviour of normalized concentrations of $^{137}$Cs in the dynamic fish-chain is an example of the results which can be obtained (Fig. 2).

By comparing the model predictions with observations, it is possible to increase further the reliability of the conceptual models and to test their applicability. In this study the normalized consequences of $^{137}$Cs deposition have been considered, accounting also for the observations available for the lake environments analysed. Preliminary results of the study indicate that, in many cases, consistency has been obtained in the temporal behaviour of the responses of different aquatic environments after an accidental deposition of radionuclides.

**REFERENCE**

Preliminary observation of the radioecological conditions and a dose assessment have been carried out at the candidate site of the first nuclear power plant in Muria, Central Java. The radioecological conditions were evaluated by examining the $^{137}$Cs concentrations in some environmental samples. Fish and shrimp, which are consumed by the local people, were collected and their $^{137}$Cs content was measured. Sea water was also collected at ten different places around the site.

The seawater samples were acidified and added to a caesium carrier. These samples were then radiochemically analysed and their $^{137}$Cs content was counted with a gamma spectrometer. The collected fish and shrimp were dried at 105°C and ashed at 450°C. The ash was measured with a gamma spectrometer.

The concentration of $^{137}$Cs in sea water ranged from undetectable amounts to 1.09 mBq/kg; the concentration in fish ranged from undetectable amounts to 0.29 Bq/kg and that in shrimp ranged from undetectable amounts to 0.09 Bq/kg.

From data collected by the Indonesian Central Bureau of Statistics is was found that the quantities of fish and shrimp consumed annually were 1.157 kg and 0.583 kg, respectively [1, 2]. By using a $^{137}$Cs dose coefficient of $1.3 \times 10^{-8}$ Sv/Bq for the committed effective dose and of $1.5 \times 10^{-8}$ Sv/Bq for the committed organ (uterus) dose, the committed effective doses for the population around Muria were estimated to be 0–0.218 µSv and 0–0.034 µSv for fish and shrimp, respectively, and the committed organ doses were estimated to be 0–0.252 µSv and 0–0.039 µSv, respectively.

REFERENCES

Laboratory experiments were made at the end of autumn and the beginning of winter in two aquariums with a volume of 100 L each. Each aquarium contained natural water of various quality and the same number of carp fry (Cyprinus carpio, L.). The water temperature ranged from 8 to 12°C throughout the whole experiment. The fish were not fed.

Water samples were taken from the river Vltava at two places (approximately 65 km away from each other). The river Vltava will be affected by liquid radioactive effluents from the Temelín nuclear power plant (PWR type).

Water type 1 (eutrophic, contaminated by municipal sewage water) was taken not far from Temelín, below the town Týn n. Vltava. Water type 2 (less eutrophic than water type 1) was taken in the middle of the stream, below the Orlik dam. The physical, chemical and bacteriological characteristics of both types of water were analysed. A list of some of these characteristics is given on the next page.

At a certain time ($t_0 = 0$) the same amount of the radionuclides $^{54}$Mn, $^{60}$Co, $^{65}$Zn, $^{85}$Sr and $^{137}$Cs was mixed with the water in each aquarium. The activity concentrations of the individual radionuclides were ordinarily $10^1$ Bq/mL. At certain intervals, samples of water and fish were taken. The water was filtered with 0.4 μm membrane filters. The activities of the fish, the filtered water and the filters were measured by semiconductor gamma spectrometry.

The results obtained for the time courses of the activity concentrations of individual radionuclides are as follows.

1. The time course of the volume activity of filtered water is analogous for the two types of water. Only the decreases in $^{54}$Mn, $^{60}$Co and $^{65}$Zn are significant, while the initial volume activities for $^{85}$Sr and $^{137}$Cs have changed only little during the experiment.

2. The time course of the specific activities of fish reflects the described course of the volume activity of water. A steady state of the system was not reached during the experiment. For $^{54}$Mn, $^{60}$Co and $^{65}$Zn there was a relatively fast increase between the eighth day and the tenth day; for $^{85}$Sr and $^{137}$Cs the increase was much
smaller, but it continued during the whole period of the experiment. The concentration factors are practically of the same magnitude for both types of water; for the individual radionuclides they are similar to values given in the literature (see, for example, Refs [1-3]). The speed of radionuclide accumulation is somewhat higher for water type 2, which is cleaner, than for water type 1.

(3) The time course of the activity of suspended particles related to the unit volume of water differs somewhat for the two types of water. For $^{54}$Mn, $^{60}$Co and $^{65}$Zn the activity of suspended particles from water type 2 decreases quickly after an abrupt increase during the first day; in the case of water type 1 this decrease is slower.

The experiment showed different kinetics of sorption/desorption of the above mentioned radionuclides in suspended particles in various types of natural water (taken from various localities) and certain small differences in the accumulation of these radionuclides in carp living in these waters.

<table>
<thead>
<tr>
<th>Analysis</th>
<th>Water type 1</th>
<th>Water type 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Undissolved matter (105°/600°)</td>
<td>10/7</td>
<td>3/2</td>
</tr>
<tr>
<td>Dissolved matter (105°/600°)</td>
<td>250/130</td>
<td>240/120</td>
</tr>
<tr>
<td>pH</td>
<td>7.1</td>
<td>7.1</td>
</tr>
<tr>
<td>Total hardness (mval/L)</td>
<td>0.95</td>
<td>0.99</td>
</tr>
<tr>
<td>Permanganate demand</td>
<td>15.5</td>
<td>11.0</td>
</tr>
<tr>
<td>Biochemical oxygen demand</td>
<td>3.4</td>
<td>2.4</td>
</tr>
<tr>
<td>Sodium (mg/L)</td>
<td>14.0</td>
<td>15.5</td>
</tr>
<tr>
<td>Potassium (mg/L)</td>
<td>8.0</td>
<td>9.0</td>
</tr>
<tr>
<td>Zinc (mg/L)</td>
<td>0.01</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Manganese (mg/L)</td>
<td>&lt;0.05</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>Calcium (mg/L)</td>
<td>30</td>
<td>27</td>
</tr>
<tr>
<td>Magnesium (mg/L)</td>
<td>3.9</td>
<td>6.8</td>
</tr>
<tr>
<td>Ammonium (mg/L)</td>
<td>0.75</td>
<td>0.3</td>
</tr>
<tr>
<td>Nitrite (mg/L)</td>
<td>0.03</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Nitrate (mg/L)</td>
<td>5.0</td>
<td>6.5</td>
</tr>
<tr>
<td>Humin matter (mg/L)</td>
<td>1.5</td>
<td>3.4</td>
</tr>
<tr>
<td>Total phosphate (mg/L)</td>
<td>0.87</td>
<td>0.46</td>
</tr>
<tr>
<td>Conductivity ($\mu$S/cm)</td>
<td>243</td>
<td>239</td>
</tr>
</tbody>
</table>
REFERENCES


IAEA-SM-339/144P

ANALYTICAL APPROACH TO MEASUREMENT OF RADIONUCLIDES IN A CONTAMINATED FORMER NUCLEAR WEAPONS TESTING AREA

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Recently, many countries of the former Soviet Union have expressed to the IAEA their deep concern about the radiological situation in areas that were once used for nuclear related activities, such as uranium mining/processing and weapons development, manufacture and testing. The independent Republic of Kazakhstan asked the IAEA for assistance in re-evaluating the radioactivity contamination in and around the former nuclear weapons testing site at Semipalatinsk in order to assess the health risks to the population. The IAEA established a special project through its Department of Nuclear Energy and Safety [1]. With the assistance of the Department of Technical Co-operation, two expert missions were sent to Semipalatinsk to

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make a preliminary assessment of the environmental contamination in that area. The second mission consisted of experts from France, Kazakhstan, the Russian Federation, the United Kingdom, the United States of America and the IAEA’s Department of Research and Isotopes. Both missions collected environmental samples, but only the sampling and analytical operating procedures used in conjunction with the IAEA Laboratory in Seibersdorf during the second mission (15–30 July 1994) are described here.

Sampling protocols, which had been worked out in detail before the start of the second mission according to guidelines recommended by the IAEA [2], were followed carefully, with only minor deviations. Fresh milk was collected from individual farms in new fluoro-plastic 1 L screw-cap bottles with metal tags; the milk was kept frozen without addition of preservative and then deep-frozen in liquid nitrogen just before air shipment in an insulated box. Grass/vegetation samples, typically about 300 g, were cut from 1 m² pastoral areas with a light-weight grass trimmer; the fresh weight was determined on the site, using an electronic hanging balance, and the specimens were air dried. Soil cores of 5 cm diameter and about 20 cm depth were removed from selected sites with a steel pipe coring tool. In most cases, the intact soil cores were carefully extruded, wrapped in aluminium foil and put into plastic shipping tubes in the field. All of the milk, grass/vegetation and soil core samples were transported in secured containers, shipped by air freight to Vienna, and retrieved by the IAEA Laboratory. Reasonable precautions were taken to preserve sample integrity throughout the mission. Several duplicate samples were placed in the custody of the team members from Kazakhstan.

Team members collected milk, grass/vegetation and soil samples in farms and villages around the former test area (the so-called ‘Polygon’) and also within it. Six milk samples were taken from farms/villages; five grass/vegetation samples were taken from farms/villages and five samples from the Polygon area; ten soil core samples were taken from farms/villages and seven samples from the Polygon area.

Samples from the inhabited areas were assigned the highest priority for analysis. Analysis of these materials provides unique information about the distribution and concentration of radionuclides in the environment. The reasons for this are as follows:

— The activity concentrations of gamma emitting radionuclides can be determined more precisely under controlled laboratory conditions (e.g. \(^{137}\)Cs concentration in milk, long lived artificial and natural gamma emitters in grass and soil profiles) than under field conditions.
— The activity concentrations of \(^{90}\)Sr and Pu can be determined in the samples after radiochemical separation by liquid scintillation counting and alpha spectrometric measurements, respectively.

A work plan with established analytical procedures was prepared, following an approach such as, for example, that which was used in the case of the analyses
of samples from the United Nations Special Mission to Iraq [3]. The general strategy was to direct the sample flow through non-destructive measurements by high resolution gamma ray spectrometry before any destructive radiochemical operations were started. Sample preparation was initially limited to drying and homogenization for the non-destructive assays. This allowed a well characterized gamma spectrometer with a HPGe well-type detector to be used.

After non-destructive measurements, the sample materials were ashed at 600°C. Depending on the amount of remaining ash, all or part of it was taken for $^{90}$Sr and Pu radiochemical analyses [4, 5]. Generally, the $^{90}$Sr and Pu analyses were carried out sequentially from the same ash sample after aggressive decomposition in the presence of added Sr carrier and $^{236}$Pu tracer, using HF, NHO$_3$ and HCl.

During the analyses, quality control was maintained by measurements/analyses of selected reference materials and blanks, careful documentation, taking precautions against sample contamination, and use of approved, well tested analytical methods and procedures. The results obtained can be compared with other existing data and could provide a reliable input (but necessarily limited in quantity) into models used for making critical radiological assessments.

REFERENCES

COMPARATIVE EVALUATION OF ANALYTICAL METHODS FOR THE DETERMINATION OF $^{90}$Sr

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For rapid determination of $^{90}$Sr, different methods have been applied widely, such as those based on solvent extraction of yttrium, followed by analysis of $^{90}$Y, which is assumed to be in secular equilibrium with $^{90}$Sr. Tributyl phosphate (TBP) is one of the most favoured solvents, and the Cerenkov counting technique has the best reputation for detection of low level activities [1]. Recently, a novel, selective extraction chromatographic material, consisting of a supported crown ether (commercially available as Sr. Spec from EiChrom Ind. Inc.), has been developed by Horwitz et al. [2] for separation of strontium. Following the chemical separation procedure, $^{90}$Sr is counted directly by a beta measuring technique, most frequently by liquid scintillation counting. The new analytical method is being utilized in a rapidly increasing number of applications, including the analysis of environmental samples [3], and different procedures have been developed and adopted. The 'classical' method, based on the separation of strontium by a set of semi-selective precipitations, is still the only accepted/standard procedure in many countries that is applied in radioactivity monitoring networks.

Two basically different 'rapid' methods have been compared with each other and with the 'classical' method, regarding both theoretical and practical aspects.

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### TABLE I. DETERMINATION OF $^{90}$Sr IN DIFFERENT STANDARD REFERENCE MATERIALS

<table>
<thead>
<tr>
<th>Sample</th>
<th>Strontium extraction by crown ether</th>
<th>Yttrium extraction by TBP</th>
<th>Reference value</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{90}$Sr</td>
<td>$^{90}$Sr</td>
<td>$^{90}$Sr</td>
</tr>
<tr>
<td></td>
<td>Sample weight, ash (g)</td>
<td>Average recovery (%)</td>
<td>Activity concentration (Bq/kg d.w.)</td>
</tr>
<tr>
<td>Soil-6, soil</td>
<td>10</td>
<td>75</td>
<td>28.53</td>
</tr>
<tr>
<td>IAEA-321, milk</td>
<td>10</td>
<td>79</td>
<td>3.13</td>
</tr>
<tr>
<td>IAEA-373, grass</td>
<td>1–2</td>
<td>70</td>
<td>1387&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>A-12, animal bone</td>
<td>10</td>
<td>52</td>
<td>47.4&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

<sup>a</sup> The result is corrected for the Th content of the sample.

<sup>b</sup> The result is corrected for the Sr content of the sample.
Experiments have been performed to analyse a set of standard reference materials of 'known' $^{90}$Sr content and of different matrix composition. As an example, results obtained by analysis of soil, milk, grass and animal bone samples are shown in Table I.

Major considerations regarding the characteristics of the chemical procedures and the measuring techniques include sensitivities, accuracies/uncertainties, chemical recoveries, selectivities/possible interferences, time and labour demands, cost and quality control, as well as fields of application. Regarding most of the parameters, the two methods are comparable. Because all of the longer lived strontium and yttrium radionuclides, i.e. $^{89}$Sr, $^{90}$Sr, $^{90}$Y and $^{91}$Y, are pure beta emitters and their differentiation by nuclear measuring methods is restricted, the final measuring source must have high purity. According to our measurements, this purity is usually provided by the crown ether method as a result of its relatively high selectivity with regard to strontium. Possible interferences due to potassium, barium or other radionuclides can be minimized or avoided. The major interference in the TBP procedure comes from thorium, so that its separation has to be included in the procedure. Omitting the correction for thorium, e.g. in the case of the soil-6 sample (Table I), results in overestimating the $^{90}$Sr activity by a factor of two. Suitable purity is provided by the 'classical' method, but the procedure is extremely time and labour consuming. The fact that the 'classical' procedure must be performed only by highly skilled personnel cannot be neglected in routine analytical laboratories.

The final conclusion is that both 'rapid' methods are reliable, accurate and competitive with the 'classical' procedure. The analyst must base his choice upon consideration of the various parameters outlined above.

REFERENCES

TIME DEPENDENT CHANGES IN MOBILITY OF RADIONUCLIDES IN SOIL–WATER–PLANT SYSTEMS

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Information on transformation processes within the affected ecosystems is needed for assessing the short term and long term consequences of authorized or accidental releases of radionuclides from nuclear installations. The mobility and bioavailability of radionuclides will depend largely on their physico-chemical forms. After the Chernobyl accident, significant fractions of radionuclides were deposited in the form of uranium oxide fuel particles, especially at sites close to the reactor. After deposition, transformation processes can alter the original distribution of species. Radionuclides can be mobilized owing to weathering; ionic species can interact with mineral and biological components in soils or they can be taken up by vegetation, and both colloidal and ionic species can be transported to other parts of the ecosystem with run-off or percolating water.

In the terrestrial ecosystem, soil represents the major reservoir for deposited radionuclides. Hence, processes taking place within the soil will play a major role in controlling the observed concentrations and the ecological half-lives of radionuclides in other parts of the system. The transfer of $^{137}$Cs and $^{90}$Sr in natural soil–water–plant ecosystems is discussed here, with particular emphasis on the mobility, physico-chemical forms and transformation processes of deposited radionuclides. The data presented are based on samples collected in Norway between 1989 and 1994, as well as soil samples collected at sites in Belarus and Russia between 1991 and 1994.

Information on the fraction of radionuclides present in a mobile or readily labile form (where labile means being able to change chemically) was obtained by NH$_4$Ac extraction of soils. Changes in the mobility of $^{137}$Cs and $^{90}$Sr were studied as a function of time (1989–1994) and season. Sequential extraction [1] was carried out to provide more detailed information on the association of radionuclides with soil
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FIG. 1. Sequential extraction of (a) $^{137}$Cs and (b) $^{90}$Sr in Norwegian soil samples (1989–1992, 12 samples per year).

components. Soil to plant transfer coefficients were compared with the fraction of radionuclides in labile form. At one site in Norway, the labile fraction of $^{137}$Cs in soil decreased from a mean of 13% to 4% between 1989 and 1992, approximating a half-life of 1.8 years. No significant change in $^{90}$Sr was observed during this time, and between 49 and 75% of the radionuclides were found to be in a labile form (Fig. 1). Changes with time in the fraction of radionuclides present in a labile form indicate that transformation processes have occurred which affect the mobility and bioavailability of radionuclides. Some evidence of seasonal variations in $^{137}$Cs mobility was found in both Norwegian and Russian soils. This may reflect microbial activity. In extractions of Norwegian soils it was found that the NH$_4$Ac extractable fraction decreased with depth, from between 3 and 11% in the 0–1 cm layer to between 1 and 4% in lower layers. This probably reflects the higher organic content in the upper soil layers.

Radionuclide distributions in the soil–plant–water system were compared with the distributions of naturally occurring stable Cs and Sr. This provided valuable information on the ‘equilibrium status’ of the deposited radionuclides within the ecosystem and can be used in predicting future transfer levels. The labile fraction of stable Cs in all soils (from <0.05% to 1%) was less than that of $^{137}$Cs, indicating that the mobility of $^{137}$Cs may be further reduced with time. Furthermore, the soil to plant concentration ratios for $^{137}$Cs were up to a factor of ten higher than those observed for stable Cs. It is expected that, in the course of time, when $^{137}$Cs
is transported to lower parts of the soil profile and bound to inaccessible sites in the clay matrix, the lability and the soil to plant concentration ratio for $^{137}\text{Cs}$ will approach those of stable Cs. The rate of these processes, together with the physical half-life of $^{137}\text{Cs}$, will determine the ecological half-life of $^{137}\text{Cs}$ in food-chains.

The distribution of $^{90}\text{Sr}$ and stable Sr between sequential extraction fractions was similar in Norwegian and Russian soils, but some of the Byelorussian soils showed considerable differences in this distribution, with the labile fractions of $^{90}\text{Sr}$ being as low as 15% and 25%. Reduced availability of $^{90}\text{Sr}$ in near-field soils is thought to reflect the influence of fuel particles [1, 2]. From comparisons with the lability of stable Sr, we find that our results indicate that the mobility of $^{90}\text{Sr}$ and the soil to plant transfer may increase in the future.

REFERENCES

First the Commissariat à l'énergie atomique and then Cogéma have been operating a uranium mill, located at Ecarpière (Loire Atlantique), from 1957 to 1991. The total output was nearly 15 000 t U in yellow cake; neutralized mill tailings totalling more than seven million tonnes were allowed to settle in a pond behind a dyke built with the coarse fraction of the residues; and nearly four million tonnes of acid heap leaching wastes were stockpiled. Remediation of the uranium tailings pond at Ecarpière is being performed by Cogéma [1].

Characterization studies of the mill tailings (mineralogy and geochemistry [2], geotechnical characteristics) have demonstrated an interesting natural evolution: a real diagenesis is occurring, which could further reduce the naturally low solubility of radium (results of normalized leaching tests) and lead to self-confinement. Local materials have been used for the cover; a layer of coarse, compacted heap leaching wastes restricts erosion, and final control of radioactivity is provided by a compacted layer of altered barren rock.

Remediation includes dismantling of the mill, re-sloping of the dike, placement of the cover, as well as drainage of the whole site and water collection. The water treatment plant is adapted to the remaining waste water flow after remediation.

During and after remediation, monitoring goes on, using a complete network on the site and in the neighbourhood. Evaluation of the environmental impact implies the following:
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— Annual radiological measurements of the main pathways during and after remediation;
— Use of similar values for assessment of the natural background (a method is described in Ref. [3]);
— Description of the critical group;
— Calculation of the added total annual exposure (ATAE), which is the sum of the different exposures due to mining, with deduction of the initial natural exposure;
— Application of the regulatory value, which is the sum of the exposures, rated to their equivalent maximum recommended value of 5 mSv. The added total annual exposure rate (ATAER) must be less than 1.

Table I gives data for two stations. The exposure and the ATAER of the critical group are evaluated with the following parameters:

— Annual residence time: 7000 h;
— Standard breathing rate: 0.8 m$^3$/h;
— Daily amount of ingested water: 2.2 L of downstream water (this figure includes water ingested through food consumption).

Hautegente is the nearest station in the environment of the impoundment and Bel Air was set up in 1990 for assessing the radiological background of the granitic region. Although the values in Hautegente are slightly higher than the background levels, the global impact of the site (ATAER = 0.12) is limited.

### TABLE I. DATA FOR HAUTEGENTE AND BEL AIR

<table>
<thead>
<tr>
<th>Station</th>
<th>External exposure</th>
<th>Internal exposure</th>
<th>Water pathway</th>
<th>TAER</th>
<th>ATAER</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Gamma rays (nGy/h)</td>
<td>$^{220}$Rn (nJ/m$^3$)</td>
<td>$^{222}$Rn (nJ/m$^3$)</td>
<td>Dust (mBq/m$^3$)</td>
<td>$^{226}$Ra (Bq/L)</td>
</tr>
<tr>
<td>Hautegente</td>
<td>200</td>
<td>18</td>
<td>51</td>
<td>1</td>
<td>0.11</td>
</tr>
<tr>
<td>Average, 6 stations</td>
<td>242</td>
<td>17</td>
<td>55</td>
<td>1</td>
<td>0.11</td>
</tr>
<tr>
<td>Average, 13 stations</td>
<td>211</td>
<td>15</td>
<td>45</td>
<td>1</td>
<td>0.13</td>
</tr>
<tr>
<td>Bel Air</td>
<td>150</td>
<td>13</td>
<td>35</td>
<td>1</td>
<td>0.12</td>
</tr>
</tbody>
</table>
Figure 1 presents the calculated total exposures according to French regulation (Decree 90-222, with ICRP-26 equivalents of 5 mSv).

Because of the natural variability of radioactivity, including also the uncertainty of the measurement, maximum and minimum individual measurements (quarterly or monthly values for external exposure and alpha potential energy, respectively) show a great variation for both Hautegente and Bel Air. The resulting annual exposures are given in Fig. 1; they range from 1 to 2.5 mSv for Hautegente and from 0.8 to 1.4 mSv for Bel Air, i.e. a maximum difference of 1.5 mSv for Hautegente and of 0.6 mSv for Bel Air. This difference is reduced to 0.5 mSv for Hautegente if the annual maximum and minimum values are used for calculation of the variability of total exposure. Both of these figures are near to 1 mSv, which is the new limit for the ATAE recommended in ICRP-60.

The following points should be mentioned:

(a) The ICRP recommendations are very difficult to comply with when dealing with natural radioactivity and consequently they are difficult to adapt to the mining industry;

(b) Compliance with the ICRP dose limits requires:
   — The use of annual means of measurements (because seasonal variability is too great);
   — Better definition of the critical group and of the scenario used for calculation of the total exposure (only an approximate evaluation of the environmental impact can be made on the basis of a 7000 h residence time);
— Better definition of how and where to measure, in the natural environment, the different parameters used for calculation of the ATAER;
— Special attention to the choice of measuring equipment used for monitoring the environment (active dosimeters).

REFERENCES


IAEA-SM-339/158P

BIOLOGICAL RELEVANCE OF HOT PARTICLES INGESTED BY DOMESTIC ANIMALS

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A major fraction of radionuclides deposited in the zone near the Chernobyl nuclear power plant was associated with hot particles. On the surface soil, hot particles were still identifiable several years after the accident. The collection of hot particles by the Ukrainian Institute of Agricultural Radiology showed a large variation in size, activity levels, activity ratios and structure. In general, the particles are
depleted in volatile elements; the varying activity ratios of refractory elements are attributed to fuel with varying burnup.

Initially, in soils contaminated with hot particles the uptake of radionuclides by vegetation and consequently the transfer to animals was low. However, because of weathering of particles, radionuclides will be mobilized with time. Depending on soil type and composition, released Cs isotopes may become fixed to clay minerals, while mobilized $^{90}$Sr may be available for plant uptake. Increasing uptake of $^{90}$Sr by vegetation and transfer to river systems with time has also been observed in the Chernobyl zone.

Hot particles associated with vegetation and surface soil may be taken up by grazing animals, as observed in 1989 in Norway when a fistulated goat grazed contaminated pastures. However, detailed studies of the uptake of radionuclides released from hot particles do not seem to be available. For most models, no release of radionuclides is expected to occur in the gastro-intestinal (GI) tract and the transit time is believed to be relatively short (2–4 days in goats).

In the present work, six particles from the Chernobyl hot particle bank of the Ukrainian Institute of Agricultural Radiology were characterized with respect to gamma emitter composition (Ge detector) and structure (electron microscopy with X-ray microanalysis). The particles ranged from 50 μm to several hundred micrometres, and contained uranium and mixed fission products (e.g. Cs isotopes, $^{144}$Ce, $^{103}$Ru, $^{124}$Sb). After characterization, the particles were placed in gelatine capsules and administered to lactating goats, using a stomach tube placed in the rumen (one particle per animal). The goats were placed in metabolic cages, and samples of milk, urine and faeces were collected daily. Because of the high activity of gamma emitters in the particles, the transfer through the animals could be followed by live monitoring (NaI detector). When the particles were excreted, they were re-examined with respect to gamma emitters and structure.

The results show that the retention time of five particles varied from 2 to 10 days. One particle was, however, retained for about 3.5 months. This animal was slaughtered and the particle was found to be attached very strongly to an epithelial fold in the reticulum. No structural changes in the retained particle were found.

From a comparison of the radionuclide activity of the particles before ingestion with that after excretion it is found that the release of radionuclides varied (up to 60% of Cs isotopes) and seemed to be closely related to the structure. On the basis of activity measurements of milk and urine samples, the maximum uptake of Cs isotopes was estimated to be about 25% of those initially present in the particle. The excretion of Cs isotopes in milk and urine approximated that which would be expected after a single oral dose of radionuclides. Electron microscopy of particles excreted in faeces showed that structural changes also occurred (disintegration of particle aggregates).

These findings should be of relevance for dose assessments, for the following reasons: (a) the retention of hot particles in the GI tract may be significantly longer
than expected; (b) the dose from a retained point source is different from the dose from whole-body absorption; (c) release of radionuclides from hot particles may occur during digestion in the GI tract, followed by subsequent uptake of Cs isotopes.

IAEA-SM-339/170P

BEHAVIOUR OF $^{125}$Sb, $^{134}$Cs AND $^{137}$Cs IN FOREST ECOSYSTEMS

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In the framework of the research project "Cycling of $^{137}$Cs and $^{90}$Sr in natural ecosystems", funded by the European Union, several coniferous and deciduous forests have been investigated in Southern Bavaria. Since 1987, $^{134}$Cs, $^{137}$Cs and $^{125}$Sb have been observed in different forest soil layers of a coniferous stand at Hochstadt near Lake Ammersee.

In order to investigate the locations of the mycelia of fungi, the time dependence of the $^{137}$Cs/$^{134}$Cs ratio and the absolute activity of $^{134}$Cs, based on Bq/kg dry weight, have been analysed and compared with these quantities measured in different soil horizons. Both the $^{137}$Cs/$^{134}$Cs ratio and the absolute $^{134}$Cs activity measured in fruit bodies of *Lepista nebularis* can be explained by assuming that this species takes up radiocaesium from the L-horizon and/or the Of-horizon (see Figs 1 and 2).

![Graph showing $^{137}$Cs/$^{134}$Cs ratios in different soil horizons](image)

*FIT-L
FIT-Of
FIT-Oh
FIT-Ah
Rat-LN

**FIG. 1.** Fitted $^{137}$Cs/$^{134}$Cs ratios in the L-, Of-, Oh- and Ah-horizons compared with measured $^{137}$Cs/$^{134}$Cs ratios in *Lepista nebularis* (corrected for 1 May 1986).
FIG. 2. Measured $^{134}$Cs activities in Lepista nebularis (corrected for 1 May 1986) compared with a fit of $^{134}$Cs activities found in samples from the L-horizon ($TF = 0.7$, corr. coeff. = 0.79).

FIG. 3. Specific $^{125}$Sb activities measured in samples from the L-horizon (corrected for 1 May 1986). An exponential fit of the data is shown for comparison, using an initial $^{125}$Sb activity of 150 Bq/kg d.w. (corr. coeff. = 0.49).

By using a compartment model, ecological half-lives for radiocaesium and $^{125}$Sb have been derived. First results show that the residence times for $^{125}$Sb in different soil horizons are similar to or longer than the residence times for radiocaesium (Fig. 3).
Several measuring programmes have been carried out in the Austrian part of the river Danube. Some of them were concerned with long term investigations of radionuclide concentrations in water and dose assessment [1, 2]. One programme was concerned with a detailed investigation of radionuclide concentrations in fish [3]. The present project complements the previous programmes by in situ investigations of the environmental behaviour of radionuclides in sediments and suspended matter under the hydrological conditions obtaining in a three year investigation period, including seasonal variations and other influencing factors.

Data generated in this programme are presented. Particular attention is paid to the determination of the relation between radioactivity bound to particles and radioactivity in solution, and the assessment of the variability of both radiological parameters (activity concentration in different compartments) and non-radiological parameters (flow rate, sediment composition, grain size).

We discuss briefly the consequences of these investigations for routine monitoring, and the interpretation of the data in an assessment of the dose from the surface water pathway, taking into account the variation of parameters.

The samples of water, suspended matter and sediment for this recent programme have been collected on a monthly basis at different locations of the Austrian part of the Danube and in some affluents. As an example of the results, the activity concentrations of $^{137}$Cs and $^{210}$Pb in sediment and suspended matter in the course of time and the respective mean values of the hydrological parameters (flow rate, concentration of suspended matter) are shown in Fig. 1. An opposite trend of activity concentrations and flow rate can be seen.

For the three year investigation period the measured values of the $^{137}$Cs activity concentration of suspended matter are between 60 Bq/kg (dry weight) and 800 Bq/kg (Fig. 1). The $^{137}$Cs activity of water samples free from solids (part of $^{137}$Cs in solution) is between 1 and 6 mBq/L. This amount of contamination of the Danube water and solids with artificial radionuclides is mainly a consequence of the
FIG. 1. Radioactivity concentrations in suspended matter and sediment samples in the Danube (km 2094.5).
The Chernobyl accident. In situ assessment of $K_d$ values gives median values for $^{137}$Cs of about 120 000 L/kg and for $^{90}$Sr of about 600 L/kg [4]. The high $K_d$ values for $^{137}$Cs suggest a contamination of solids in the catchment area. As a result of this investigation, some very significant relations between radioactivity and hydrological parameters have been obtained.

As a consequence of the weathering processes in the alpine catchment area of the Danube, the fine grained clay and mica particle fraction in suspended solids varies from 5% to 50%. As an example of the significant influence of ecological parameters on radioactivity, the correlation between the $^{137}$Cs activity and the mass fraction of grains of $<2 \mu m$ of sediment samples is shown in Fig. 2.

This important correlation between the $^{137}$Cs activity concentration and the fraction of fine grained particles in sediment, as well as the relatively high amount of fine grained solids lead to an annual mean ratio of particle-bound $^{137}$Cs activity to $^{137}$Cs activity in solution in the Austrian Danube of 9:1 [5]. This means that, on average, about 90% of the $^{137}$Cs activity transported in the Danube is bound to suspended matter.

We have also examined the relationships between the activity concentrations of other artificial and natural radionuclides (e.g. $^{90}$Sr, $^{210}$Pb, $^{226}$Ra), as well as hydrological parameters (e.g. flow rate, concentration of suspended matter) and mineralogical parameters (e.g. grain size parameters, concentration of organic matter).
CONCEPT OF A FLEXIBLE AUTOMATED SYSTEM OF RADIOECOLOGICAL MONITORING

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Analysing the radioecological monitoring systems developed in western Europe, we came to the conclusion that the structure of these systems was not suitable for Russia. The reason lies not only in the specific geographic factors, such as the size of the monitoring area, the existence of road and telecommunication networks and the population density, but also in sociological factors, such as the educational level of the population, the social and national composition and farming practices.

Purely geographic conditions indicate that even if the financial aspects of setting up a single radioecological monitoring system are settled favourably, it will take decades to establish the system because it is necessary to prepare the appropriate infrastructure.

The socioecological factors, which determine the social function of a single radiation monitoring system, indicate that if a 'western' structure is adopted, the population of certain regions will not be in a position to make use of the information obtained.

The following locations can be identified for the purpose of continuous monitoring of the gamma activity level as part of an environmental monitoring system in Russia:
POSTER SESSION 1

— Sites chosen for the construction of environmentally hazardous enterprises (initial gamma activity level);
— Mining areas of radioactive ores;
— Areas of disposal and storage sites for industrial waste from nuclear power plants;
— Areas outside the health protection zones around nuclear facilities (nuclear power stations, plants, enterprises, medical centres, etc.) (gamma background level);
— Areas of nuclear weapons testing sites and areas highly contaminated as a result of nuclear tests or accidents at nuclear installations;
— Areas through which shipments pass.

The large number of locations for continuous monitoring and also the geographic and sociological characteristics of the Russian Federation induced us to develop a concept for a continuous radiation monitoring system.

In view of the foregoing, we have formulated the following requirements for a radioecological monitoring system:

— The system should be flexible, i.e. there should be provision for upgrading, replacing and increasing the number of information sources in the system without stopping the system as a whole.
— The information sources (radioactivity monitors) should ensure that a maximum of information is obtained at a minimum of cost in terms of production and maintenance.
— The system should be compatible with the currently operating telecommunication and processing facilities.
— The radioactivity monitors should operate unattended for a long period.

These requirements can be met by a decentralized structure, organized as a system of clearing houses for information coming from the monitors. The information sources should operate unattended and should be connected to the system’s clearing houses through the existing communication lines. In other words, we are proposing to organize the structure of the radioecological monitoring system like a bank network.

Purpose of the system: The system (SPIDER) is intended for continuous radiation level monitoring in the areas of storage, transportation and conversion of gamma emitting materials within health protection and surveillance zones and also along the radioactive material transport routes.

The system is to be used for monitoring radioecological contamination with a view to protecting the civil population and the environment. It can be incorporated into the State and regional automated radiation monitoring systems.

The system is based on automatic collection of data on the radiation situation, using automatic monitors installed within the zone of stable radiocommunication and
processing of the data at a data processing centre. Such a centre performs the following functions: (a) automatic processing of the monitoring results; (b) estimation of the dose burden of workers and of the population during normal operation, accidents and disasters; (c) estimation of gamma activity of isotopes, with recommendations to operators in the event of incidents, accidents and disasters; and (d) automatic transmission of the results to the State control system.

The system can be supplemented with monitors installed on moving vehicles, boats, etc., provided with automatic or semi-automatic navigation devices. The SPIDER system can serve for monitoring in other information systems such as meteorological stations.

IAEA-SM-339/182P

FIELD MEASUREMENTS OF $^{137}$Cs BEHAVIOUR IN THE LITTER LAYERS OF A PINE FOREST IN IRELAND

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The migration of $^{137}$Cs down the forest soil profile is complex and is governed by both physical and biological processes. In this forest study, two physical migration processes ($^{137}$Cs leaching and litter accumulation) have been evaluated and their contribution to the migration of the Chernobyl contamination compared.

Sampling was carried out in a 30 year old Pinus contorta forest with sparse understorey vegetation, in Co Waterford, Ireland. Monthly measurements were made of: $^{137}$Cs content and volume of rainfall, stemflow, throughfall and leachate from the AoL+AoF horizon and the AoL+AoF+AoH horizon; $^{137}$Cs content and mass of litter fall; weight loss of AoL and AoF litter bags. Other measurements: biomass and $^{137}$Cs content of trees; soil profile $^{137}$Cs distribution and bulk density.

The results are summarized in Figs 1–3.

Leachate analysis. The leachate from the AoL+AoF horizon represents about 9% of deposited Chernobyl $^{137}$Cs in the overlying pool; this is reduced to 2% when the leachate passes through the AoH horizon. However, these data are weighted by the difference in the size of the $^{137}$Cs pools. The lower rate of leaching from the AoH horizon demonstrates $^{137}$Cs accumulation in the AoH horizon which maintains the present distribution. Standardizing the leachates by expressing them as a percent-
FIG. 1. Distribution of $^{137}$Cs of Chernobyl origin and nuclear weapons origin in the forest soil profile.

The observed replacement of the AoL layer each year implies a constant AoL depth of ca. 1 cm. The AoL accumulation of each year is, in the following year, converted to AoF material, which builds up at a rate of ca. 0.3 cm per year (taking into account decomposition and increasing bulk density). The Chernobyl contaminated AoL litter of 1986 would, by this model, have been buried at a rate of about 0.3 cm per year, bringing it, in 1992, to a depth of about 2.5 cm. Further downward, migration would have occurred via decomposition and compression of the litter (both AoF and AoH) which lay below the surface litter at the time of deposition. There is a discrepancy between the predicted burial depth and the observed depth of the Chernobyl peak deposition, but there is variability in the observed profile depths and the model is based on simple observations over one year; it does not consider other biological and physical processes which contribute to vertical migration of soil elements. This analysis assumes that (a) patterns of litter accumulation and decomposition observed in the study are relevant to each year since 1986, and (b) the overall depth of the organic horizons in the soil profile has not increased significantly in the past six years.
FIG. 2. Leaching of $^{137}$Cs (per square metre) to and within the forest floor, expressed as a percentage of total Chernobyl $^{137}$Cs deposition and as a percentage of the Chernobyl $^{137}$Cs in the overlying soil and biomass.
FIG. 3. Predicted accumulation of AoL and AoF litter on top of the Chernobyl contaminated litter, over the years since deposition. The actual accumulation is also illustrated for comparison.

Conclusions. (1) About 1% of deposited $^{137}$Cs is in circulation in this forest ecosystem; (2) the dominant process involved in the migration of $^{137}$Cs down the forest soil profile is the accumulation of relatively uncontaminated litter on top of the contaminated material; and (3) the distribution pattern is maintained by the retention of $^{137}$Cs in the AoH horizon.

REFERENCE

DO CRITICAL GROUPS EAT A LOT OF FOOD ALL THE TIME, AND WHERE DOES IT COME FROM?

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The assessment of the impact of radioactive waste disposals on the dose to the public from ingestion of actually or potentially contaminated food requires a combination of data and assumptions to quantify the environmental transfers of radionuclides and the habits of the food consumer. Nationally representative data, site specific investigations and maximizing assumptions all play a part in this process but invariably contain a common weakness: chronic exposure is the objective of the assessment, but food consumption data are all derived from short duration studies.

**Consistency of consumption.** The results of a study which has followed the food consumption habits of a group of 1220 people for 52 consecutive weeks are presented. An initial cohort of 659 persons were recruited and they provided details of their daily usage of 17 foods or food types by themselves and by members of their families. Information was collected on foods which are of particular interest because of their potential for collecting and concentrating environmental radionuclides (milk, home grown fruit and vegetables, honey, offal, shellfish). Also included in the study were certain types of food (herbs, mushrooms, shellfish, game, offal) that are not widely consumed because, without them, extrapolating existing food consumption data was likely to provide misleading results.

The results confirm that not all persons who consume a particular food in a given week or month will continue to consume that food every week or month. However, some do, and we can determine the size of this ‘regular consumer’ population for each of the foods.

**Seasonal changes in popularity of foods.** By collecting data over a whole year, a picture can be drawn up of the seasonal variation in the popularity of some foods. It seems clear that, for instance, the regulation of shooting seasons and the popularity of so-called luxury foods around Christmas has a marked influence on the usage of foods such as game and shellfish through the year.
| Source of foods. It is a key element of the assessment of radionuclide ingestion that critical groups are assumed to eat food that is local to the source of contamination. A study of farmers in England has provided information on the amount that persons living and working on a farm make use of that farm’s products. Not only does this clearly demonstrate that local food is being used and therefore provides an established pathway for activity from waste discharges to reach the local population, but such local food can also represent a substantial proportion of the total amount of some food types eaten. |
Proportion of consumers (%)  

<table>
<thead>
<tr>
<th>Proportion of food from farm</th>
<th>Cows' milk</th>
<th>Lamb</th>
<th>Potatoes</th>
<th>Vegetables</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Adult</td>
<td>Child</td>
<td>Adult</td>
<td>Child</td>
</tr>
<tr>
<td>&lt;25%</td>
<td>48</td>
<td>9</td>
<td>58</td>
<td>25</td>
</tr>
<tr>
<td>25-50%</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>5</td>
</tr>
<tr>
<td>51-75%</td>
<td>0</td>
<td>4</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>76-99%</td>
<td>4</td>
<td>0</td>
<td>0</td>
<td>3</td>
</tr>
<tr>
<td>100%</td>
<td>26</td>
<td>69</td>
<td>15</td>
<td>23</td>
</tr>
<tr>
<td>Not known</td>
<td>22</td>
<td>18</td>
<td>27</td>
<td>45</td>
</tr>
</tbody>
</table>

IAEA-SM-339/190P

PLUTONIUM IMMOBILIZATION BY LICHENS AT MARALINGA, A FORMER NUCLEAR TEST SITE IN SEMI-ARID SOUTH AUSTRALIA

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In the 1950s and early 1960s, the United Kingdom conducted nuclear tests at Maralinga in the north-west of South Australia. Apart from a series of fully nuclear explosions, many smaller trials were made to investigate various developmental aspects of nuclear components. Some of these 'minor trials' resulted in the atmospheric dispersion of plutonium oxide particles from non-nuclear explosions. Several, superficial, wind dispersed plumes of Pu extended 20 km or more from the experimental sites, well beyond the area currently being considered for further rehabilitation [1].

The test area was inhabited by indigenous Maralinga-Tjarutja people and there is the possibility that some areas with low levels of Pu contamination will be returned to them. Inhalation and ingestion of finely divided Pu dust are thought to represent the greatest radiological health risks to people living traditionally in these areas, and children under 10 years of age are identified as vulnerable. Various activities around a camp are known to stir up dust, which may be inhaled or may contaminate food.
These risks have been identified by anthropologists and broadly quantified in field studies. Environmental factors that may limit the availability of Pu dust have received little attention. An obvious example of such a factor is the widely distributed surface crust of lichen which covers the sandy soil of this semi-arid area. This organism has the potential to restrict resuspension of respirable particles of PuO₂ by incorporating Pu into its tissues or within its hyphal structure or by simply acting to bind the surface soil together, preventing erosion.

This ongoing investigation aims at distinguishing between these possibilities and determining their significance, using a combination of bulk radiological and microanalytical techniques.

Three areas located in the contaminated region and two control areas outside this region were sampled in September 1994. In each of these five areas, two sites, a swale and an adjacent dune slope, were sampled. Six cores (~38 mm diameter and ~10 mm depth) of the lichen crust and the underlying sandy soil were taken at each site. A coherent core, 2–4 mm thick, of lichen crust with incorporated soil was easily lifted from each of these dry core samples and combined in one container. The loose soil from beneath this core was separately collected and combined in another container. A third container was filled with six cores of nearby surface soil, selected as lacking an apparent lichen crust. Non-quantitative samples were taken for identification and for analyses using electron microscopy, autoradiography and secondary ion mass spectrometry. The lichen crust is tentatively identified as a species of the genus *Dermatocarpon* [2]. The extent of the lichen cover was estimated by transecting between the swale and dune crest at each site. The percentage cover of lichen was visually estimated for 1 m² quadrats spaced at 25 m intervals. A lichen cover ranging from 25 to 50% was found for these transects with a median cover of 30%. Lichen clearly plays an important role in binding the surface soil, predominantly sand, in this area, regardless of any ability of the lichen to actively concentrate Pu. This points to a significant effect of this organism in limiting the wind access to plutonium oxide particles and thereby reducing the likely exposure of humans to inhalation of this nuclide.

Bulk analyses of samples from the control areas, using ²⁴¹Am as an indicator of Pu contamination, have shown that the lichen crust contains several times more Pu than is found in the adjacent surface soil or in the soil immediately below the crust itself. Within the plume region, where much higher ²⁴¹Am concentrations are found, similar levels of Pu appear to exist in both the lichen crust and the nearby surface soil. It is not possible from these data to decide whether or not Pu has been actively taken up and concentrated in the lichen tissue, although this may be the case at the control sites.

Preliminary surface microanalysis of lichen cupules, using secondary ion mass spectrometry, indicates that despite a great spatial variability, there are statistically detectable differences in ²³⁹Pu between samples taken from inside and outside of the contaminated area. Before this pattern emerged, the signal variability between
analyses and from day to day had to be reduced by taking the ratio of Pu and an isotope of Si, which showed no statistically significant variation between cupules.

REFERENCES


IAEA-SM-339/191P

STUDY OF THE EFFECTIVENESS OF DEEP PLACEMENT AND CONVENTIONAL PLOUGHING AS COUNTERMEASURES

Role of the root distribution with respect to the location of $^{85}\text{Sr}$ and $^{134}\text{Cs}$ in the soil profile

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The objective of this study is to evaluate the effectiveness of two countermeasures, deep placement and conventional ploughing, after deposition of radionuclides on the soil surface, paying special attention to the role of the root distribution and its activity with respect to the location of radionuclides in the soil profile. This study is included in the EURO-RESSAC (Rehabilitation of soils and surfaces after an accident) project.

To carry out this experiment, sunflower plants ($Helianthus annuus$, var. Tesoro) were cultivated in Terra rossa soil (calcic luvisol), which is representative of the agricultural Mediterranean area. A total soil surface of 1.6 m$^2$ (volume 29.95 dm$^3$), put through a 2 mm sieve, was contaminated by the “Polyr” device [1], the initial activity deposited being 1.46 MBq $^{85}\text{Sr}/m^2$ and 0.17 MBq $^{134}\text{Cs}/m^2$. The contaminated surface was homogenized by mixing it with the total
soil volume. The final values of soil concentration obtained were 4.58 Bq $^{134}$Cs/g and 54.08 Bq $^{85}$Sr/g. The total soil volume was distributed in equal parts in each of the 16 experimental pots. Four treatments were compared with four replicates per treatment.

Three of the treatments consisted of placing a 1 cm thick layer of contaminated soil at different depths: (1) at the soil surface (treatment S); (2) at 15 cm depth (treatment P15); (3) at 30 cm depth (treatment P30); and (4) simulation of conventional ploughing (treatment M): this treatment consisted of mixing the contaminated soil with the upper 15 cm of soil, the final concentration after mixing being 0.43 Bq $^{134}$Cs/g and 5.73 Bq $^{85}$Sr/g.

Three samplings were performed at different stages of plant growth. At each sampling time, groups of leaves were separated by growth stage: leaves in expansion and fully expanded leaves. The samples were dried during 24 h at 60°C and ground. The activities of $^{134}$Cs and $^{85}$Sr were measured by gamma spectrometry in an intrinsic Ge n-type detector. The root mass distribution at depth was also determined.

The temporal changes in radionuclide absorption were studied and for each sampling the group of last fully expanded leaves was considered. The behaviour of $^{134}$Cs and $^{85}$Sr absorption was different with regard to the plant vegetative stage and the root mass profile at depth.

Considering the final absorption values in the whole plant (Table I), $^{85}$Sr showed maximum absorption in the P15 and P30 treatments, less absorption in treatment M and minimum absorption in treatment S. For $^{134}$Cs the absorption in treatment M is lower than that in treatments S and P15, indicating that dilution has affected $^{134}$Cs absorption. For $^{85}$Sr, no dilution effect has been observed. Therefore, conventional ploughing was effective in reducing $^{134}$Cs transfer to the crop, but for $^{85}$Sr the effect was negligible.

In the P30 treatment, there was no measurable $^{134}$Cs absorption, in contrast to the high $^{85}$Sr absorption and the root mass accumulated at the 30 cm depth. This

<table>
<thead>
<tr>
<th>Treatment</th>
<th>$^{85}$Sr (Bq/g)</th>
<th>$^{134}$Cs (Bq/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Treatment S (surface contamination)</td>
<td>6.19 ± 0.31</td>
<td>0.050</td>
</tr>
<tr>
<td>Treatment M (conventional ploughing)</td>
<td>7.82 ± 0.81</td>
<td>0.035</td>
</tr>
<tr>
<td>Treatment P15 (deep placement, 15 cm)</td>
<td>8.20 ± 0.70</td>
<td>0.048</td>
</tr>
<tr>
<td>Treatment P30 (deep placement, 30 cm)</td>
<td>10.43</td>
<td>LLD$^a$</td>
</tr>
</tbody>
</table>

$^a$ Lower limit of detection.
could indicate that a factor other than $^{134}$Cs soil fixation was involved; the root uptake mechanism for $^{85}$Sr may be different from that for $^{134}$Cs.

The fact that $^{134}$Cs absorption was two orders of magnitude lower than $^{85}$Sr absorption might be explained by the greater fixation of $^{134}$Cs in the Terra rossa soil [2].

The first experiment was carried out in a greenhouse, where the climatic and growth conditions were not representative of the real Mediterranean area; therefore, the same experiment will be carried out in the field.

REFERENCES


IAEA-SM-339/196P

INDICATOR VALUE OF AQUATIC ORGANISMS IN ENVIRONMENTAL MONITORING PROGRAMMES OF FINNISH NUCLEAR POWER PLANTS

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Since the 1970s, extensive monitoring programmes have been carried out in the environs of the Finnish nuclear power plants at Loviisa and Olkiluoto, situated on the southern coast and the western coast of Finland, respectively. In terrestrial environments, the main objects of sampling are ground level air and deposition, as well as soil, moss, grass, milk, garden produce, grain, meat and some other food-stuffs. In marine environments, samples of sea water, sinking matter, bottom sediments, fish, some seaweeds and benthic animals are taken. The total number of samples and radionuclide analyses from the two power plant sites is about 1000 per year. The Research Department of the Finnish Centre for Radiation and Nuclear Safety is responsible for the implementation of the monitoring programmes.
Indicator organisms and other indicator samples are widely used in monitoring radioactive substances in the environment. The main advantage of indicators is their ability to accumulate radionuclides from the surrounding medium; this makes it easier to detect small quantities of radionuclides in indicator samples than directly in air, soil, sea water, sediments, etc. Some indicators are specific for certain radionuclides, which enhances their usefulness in monitoring programmes. With regard to radioecology, indicators are different from air and water, which represent the discharge or spread medium, as well as different from foodstuffs, which can cause radiation exposure of people.

Several factors should be taken into account when choosing indicators for monitoring programmes. An indicator organism should be common in the area to be monitored and easy to collect. It should also have a significant role in the food-web (preferably in food-chains leading to man); in addition, different trophic levels and living habits should be represented. Furthermore, the species stock should be viable and resistant to environmental changes. To judge its significance in the accumulation process as a whole, the abundance and biomass of the species in the area and its niche in the ecosystem should be known.

The water of the Baltic Sea near Loviisa and Olkiluoto is very brackish; the water salinity ranges from almost 0 to 67 ‰. This causes difficulties in choosing indicator organisms because the fauna and flora are very poor in species, and the abundance of existing species is usually very low. In many cases the organisms are also smaller than those in more marine areas, which complicates sampling. Under these circumstances the bladder-wrack, Fucus vesiculosus, plays a very important role in littoral ecosystems and is consequently the most useful indicator organism in both areas, as are also the filamentous green algae Cladophora glomerata and Enteromorpha sp. At Loviisa, where the benthos are extremely sparse, a relict crustacean, Saduria entomon, represents the invertebrates; the bivalve mussels Mytilus edulis and Macoma baltica are the benthic indicator organisms at Olkiluoto.

With regard to indicator values of the above mentioned organisms for the most important nuclides in discharges from Finnish nuclear power plants, Fucus vesiculosus has been proven to be the best indicator for 60Co and 58Co. Cladophora glomerata and Enteromorpha seem to be better indicators of these nuclides than the benthic animals. For 54Mn, the indicator values of Fucus, Cladophora and Mytilus edulis are very similar and are clearly higher than those of Saduria and Macoma. On the other hand, Saduria is the best indicator of 110Ag and compared with Macoma, Fucus and Cladophora.

In a special study carried out in the discharge area of Loviisa in 1988-1989, 110Ag, 60Co and 54Mn were detected only in the lower trophic levels of the ecosystem, i.e. phytoplankton, zooplankton, macrophytes and benthic animals, but not in vertebrates, e.g. fish, water fowl and seals, or in their inner organs. On the other hand, the Chernobyl derived isotopes 137Cs and 134Cs were the most abundant in these groups, especially in muscle tissues of predatory fish and seals. The radio-
Caesium concentrations were generally lower in the inner organs than in flesh. Compared with these groups, *Fucus* was a weaker indicator of radiocaesium, but it was better than most other seaweeds and benthic animals.

Suspended particulate matter is collected continuously in both power plant areas by sediment traps. In a broad sense, it can be considered as a non-living indicator of radionuclides in the aquatic environment. Many nuclides tend to absorb on sinking particles; the affinity of radiocaesium to clay particles is well known. The concentrations of Chernobyl derived radiocaesium are higher in suspended particulate matter than in indicator organisms, both at Loviisa and Olkiluoto. In addition, it seems that suspended particulate matter also accumulates very effectively some other radionuclides, e.g. $^{54}$Mn, $^{58}$Co, $^{60}$Co, $^{110}$Ag$^m$ and $^{125}$Sb.

IAEA-SM-339/197p

**SOIL-PLANT TRANSFER FACTORS IN FOREST ECOSYSTEMS**

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Austria

In an Austrian monitoring programme of $^{137}$Cs behaviour in forest ecosystems in 1993, about 80 soil samples and 200 plant samples were collected in the Weinsberger Wald (spruce forest ecosystem in Lower Austria). The soil contamination was determined by analysing thin-layer profiles and pooled samples. In addition to an overview of $^{137}$Cs contamination of vegetation, special topics such as seasonality and cycling phenomena were investigated.

The results of this study and data from other countries (see Refs [1, 2]) show that the $^{137}$Cs content in different plant parts is age dependent. It is found that only plant parts of well defined age classes should be taken into account in calculating transfer factor (TF) values.

A significant decrease in TF values from the youngest shoots to older shoots was found for both conifer trees (*Picea abies*) and shrubs (*Vaccinium myrtillus*, see Fig. 1). In each case, the TF values could be calculated by an exponential regression term.
For *Picea abies* the TF values for 1–4 year old needles are expressed by the regression term

\[ y = 24.50 e^{-2.469x} + 0.846 \quad (R^2 = 0.556) \]

For *Vaccinium myrtillus*, the slope of the regression of 1–4 year old shoots is slightly steeper than that of conifer needles (see Fig. 1). Although \( R^2 \) reaches only 0.497, the curve represents a very good fit to the median values. Thus the estimated median turns out to be useful for describing the natural interconnection between TF value and age of plant parts.

By using SPEARMAN correlation coefficients, a relation of the TF values (spruce and ferns) with the fulvic acid/humic acid ratio was found in the litter layer \( (r = 0.817; \text{see Fig. 2}) \). In mineral soil layers the TF values show a relation with the amount of exchangeable aluminium content in soil \( (r = 0.683) \) and the optical density per gram humus \( (r = -0.750) \).

In recent studies, soil organic matter turned out to be a factor which influences the binding of \(^{137}\)Cs and the plant uptake in soils of semi-natural ecosystems [3]. A high degree of humification of organic matter implicates a large number of functional groups and therefore a low cation mobility because of complex formation [4]. Thus the TF values showed a negative correlation with the degree of humification (indicated by optical density per gram humus).

Under acid conditions, polymerization of humic acids is slow and disintegration processes readily occur [5]; therefore, fulvic acids are an important humus compound in forest soils and enhance the ion mobility.
FIG. 2. Correlation of TF values for spruce needles with the fulvic acid/humic acid (FA/HA) ratio.

Large fractions of exchangeable aluminium indicate soil acidification [6], which results in higher mobility of plant nutrients such as Ca, Mg and K. This is supported by the correlation of $^{137}$Cs TF values with exchangeable aluminium ions.

REFERENCES


In future fallout situations, $^{90}\text{Sr}$ deposition can be considerable and it may be important to control the transfer of radiostrontium from milk to man. One of the most effective methods of reducing transfer via this pathway is to treat dairy cows and goats with Sr binders which can bind radiostrontium in the digestive tract, reduce radiostrontium absorption and consequently decrease radiostrontium levels in milk. Although a number of compounds have been tested for Sr binding effects under controlled laboratory conditions, no efficient Sr binder that works in the digestive tract of ruminants has as yet been identified. A useful binder must be effective when competing ions such as Ca$^{2+}$ and Mg$^{2+}$ are present at concentrations 150–500 times higher than Sr; it must be stable and effective at pH values between 2 and 8; it must bind Sr effectively within the time of passage through the digestive tract; and it must be resistant to digestion by rumen-microbes. The compound must also be non-toxic, without effects on the availability of other essential minerals, and its use must be cost-effective. We have tested several compounds which meet some of these requirements in experiments with goats.

**STRONTIUM BINDING COMPOUNDS**

*Zeolites and mordenites* are a family of crystalline aluminosilicates associated with lattice-embedded alkali metal cations such as the zeolite A(Na) with the formula \( \text{Na}_{12}(\text{AlO}_2)_{12}(\text{SiO}_2)_{12} \cdot 27\text{H}_2\text{O} \). The relative content of aluminium oxide, silica and the lattice cation determines which cations may be exchanged with the surrounding solution. There is a wide range of both natural and synthetic zeolites with different characteristics. We have concentrated on the synthetic zeolites on account of their well defined composition. All zeolites and mordenites assessed in this study were obtained from Degussa, Germany.

*Potassium and sodium rhodizonate* (\( \text{C}_6\text{O}_6\text{K}_2 \) and \( \text{C}_6\text{O}_6\text{Na}_2 \)) have been used to chemically isolate (chelate) Sr and Ba.

*Antimonpentaoxide* (\( \text{Sb}_2\text{O}_5 \)) and *humalite*, a composite of clay minerals and organic substances, have shown a Sr binding effect under laboratory conditions.
TABLE I. REDUCTION IN TRANSFER OF $^{85}$Sr TO GOAT MILK BY USE OF VARIOUS Sr BINDERS

<table>
<thead>
<tr>
<th>Compound</th>
<th>Sr binder (g/d)</th>
<th>Reduction (%)</th>
<th>Compound</th>
<th>Sr binder (g/d)</th>
<th>Reduction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Humalite</td>
<td>1.0</td>
<td>ns</td>
<td>Zeolite A(Na)</td>
<td>1.0</td>
<td>24</td>
</tr>
<tr>
<td>Humalite</td>
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<td>ns</td>
<td>Zeolite A(Na)</td>
<td>2.0</td>
<td>24</td>
</tr>
<tr>
<td>Humalite</td>
<td>4.0</td>
<td>ns</td>
<td>Zeolite A(Na)</td>
<td>4.0</td>
<td>24</td>
</tr>
<tr>
<td>Sb$_2$O$_5$</td>
<td>0.25</td>
<td>ns</td>
<td>Zeolite A(Na)</td>
<td>10.0</td>
<td>38</td>
</tr>
<tr>
<td>Sb$_2$O$_5$</td>
<td>1.0</td>
<td>ns</td>
<td>Zeolite A(Na)</td>
<td>30.0</td>
<td>37.5</td>
</tr>
<tr>
<td>Na-rhodizonate</td>
<td>1.7</td>
<td>ns</td>
<td>Zeolite P(Na)</td>
<td>2.0</td>
<td>ns</td>
</tr>
<tr>
<td>K-rhodizonate</td>
<td>1.7</td>
<td>ns</td>
<td>Zeolite P(Na)</td>
<td>10.0</td>
<td>ns</td>
</tr>
<tr>
<td>Zeolite A(K)</td>
<td>2.0</td>
<td>ns</td>
<td>Zeolite Y(Na)</td>
<td>2.0</td>
<td>30</td>
</tr>
<tr>
<td>Zeolite A(Mg)</td>
<td>2.0</td>
<td>25</td>
<td>Mordenite</td>
<td>2.0</td>
<td>ns</td>
</tr>
<tr>
<td>Zeolite A(Ca)</td>
<td>2.0</td>
<td>ns</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

a Two or four goats; ns = not significant.

ANIMAL EXPERIMENTS

Eleven compounds (Table I) were tested for Sr binding activity in lactating goats. The level of $^{85}$Sr in milk from goats given only $^{85}$Sr (as an aqueous solution of $^{85}$SrCl$_2$) was compared with the level of $^{85}$Sr in milk from goats that were given both $^{85}$Sr and the potential Sr binder. In this way, each animal acted as its own control; during a 14 d test period, the animals were given daily doses of $^{85}$Sr and Sr binder; this was followed by a 14 d control period, during which animals were given only $^{85}$Sr. Milk samples were taken twice daily (morning and evening) during the period of equilibrium of the control period and the test period. The effects of the Sr binders were assessed by comparing the transfer of $^{85}$Sr to milk during those two periods (Fig. 1). This experiment was designed to screen for the most promising Sr binders, and two or four animals were used in each test (Table I).

The average transfer coefficient for $^{85}$Sr to goat milk during the control period was 0.015 ± 0.004 d/L, in accordance with reported values. Only the zeolite A(Na) at 10 or 30 g/d was of practical interest in reducing the transfer of $^{85}$Sr to milk. While this Sr binder is non-toxic at the doses given and is readily available in large quantities at low cost, it is also known to bind the essential minerals Ca, Cu
FIG. 1. Content of $^{85}$Sr in goats' milk from animals given both $^{85}$Sr (57 000 Bq/d) and Sr binder from day 1 to day 14, and from animals given only $^{85}$Sr from day 15 to day 28. The effect of the binder was calculated from the difference in the $^{85}$Sr level in milk on the plateaus on days 7-14 and days 21-27.

and Zn. Further studies are therefore necessary before zeolite A(Na) can be recommended for use over longer periods. We have identified zeolite A(Na) as a useful Sr binder in ruminants at a daily dose of 0.5 g/kg body weight.
MODEL STUDIES, IMPACT ASSESSMENT AND REMEDIATION

(Poster Session 2)
The term sensitivity analysis is used for techniques developed to evaluate the variations of a model output as a consequence of perturbations of model parameters. Sensitivity analysis of a model is a preliminary task in support of uncertainty analysis. Indeed, model parameters showing non-negligible uncertainty and to which the model output is very sensitive may contribute significantly to the overall model uncertainty. Thus, sensitivity and uncertainty analyses may provide useful information on the parameters that deserve better experimental evaluation to improve the predictive power of a model. The most common definition of the ‘sensitivity matrix’ $S_{ij}$ is the following:

$$S_{ij} = \frac{\mu_i \frac{\partial X_j}{X_j} \partial \mu_i}{\partial \mu_i}$$

(1)
where \( X_j \) is the jth modelled quantity and \( \mu_i \) is the ith parameter. \( S_{ij} \) is the ratio between the percentage variation of an output variable (\( X_j \)) and the percentage variation of a model parameter (\( \mu_i \)).

According to the above definition, an extensive sensitivity analysis of a model output is based on the evaluation of a large number of time functions. Unfortunately, analytical calculation of the elements of the sensitivity matrix is not always possible. It may be useful to evaluate the approximate ranges of the ratios of 'variation of the output variable/variation of the parameter' using the model results obtained by perturbing the parameters around their average values. Results of the sensitivity analysis of \(^{137}\)Cs migration models submitted to the IAEA/CEC co-ordinated research programme VAMP (Validation of Environmental Model Predictions) are summarized here.

The parameters used in models may be subdivided into two different categories: parameters for which reliable experimental site specific values are generally available (lake volume, lake area, lake depth, etc.) and parameters for which site specific data are seldom available and are difficult to measure. Some of the most important parameters belonging to the latter category are the rate constants controlling the diffusion of \(^{137}\)Cs in bottom sediments. In general, models may be very sensitive to the first parameter category, but, since the values of such parameters are accurately measured, the influence on the uncertainty of the output is negligible. On the contrary, though the model output may be less sensitive to the parameters of the second category, these can contribute significantly to the overall uncertainty of the model as a consequence of their uncertainty. As an example, some results of the sensitivity analysis of the model ENEA, developed to assess the \(^{137}\)Cs migration in water [1], are reported in Table I. The approximate ranges of the ratio 'variation of the output variable/variation of the parameter' are indicated by a capital letter (\(1/100 < L < 5/100\); \(5/100 < M < 10/100\); \(10/100 < H < 20/100\)). The ratios are evaluated at two different times: time = 30, corresponding to 30 days after the

### Table I. Results of the Sensitivity Analysis Applied to the Model ENEA

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Concentration in water (time = 30 d)</th>
<th>Concentration in water (time = 1000 d)</th>
<th>Deposit (time = 1000 d)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( k_d )</td>
<td>H</td>
<td>L</td>
<td>M</td>
</tr>
<tr>
<td>( K_{ult} )</td>
<td>L</td>
<td>L</td>
<td>L</td>
</tr>
<tr>
<td>( K_{bai} )</td>
<td>L</td>
<td>H</td>
<td>L</td>
</tr>
<tr>
<td>( K_{sd} )</td>
<td>L</td>
<td>M</td>
<td>L</td>
</tr>
</tbody>
</table>

\(1/100 < L < 5/100\); \(5/100 < M < 10/100\); \(10/100 < H < 20/100\).
Table II. Sensitivity Analysis of Some Tested Models

<table>
<thead>
<tr>
<th>Model</th>
<th>Institution</th>
<th>Method of sensitivity analysis</th>
<th>Parameters to which the model output is most sensitive</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>DETRA</td>
<td>VTT Energy</td>
<td>Monte Carlo</td>
<td>Sedimentation rate, $k_d$, resuspension from sediment</td>
<td>[2]</td>
</tr>
<tr>
<td>LAKECO</td>
<td>KEMA</td>
<td>Monte Carlo</td>
<td>$k_d$, sediment reworking rate</td>
<td>[3]</td>
</tr>
<tr>
<td>VAMP</td>
<td>Univ. Uppsala</td>
<td>Parameter perturbation</td>
<td>$k_d$</td>
<td>[4]</td>
</tr>
</tbody>
</table>

a: Output: radionuclide concentration in water.

deposition pulse; and time = 1000, corresponding to 1000 days after the deposition pulse. Of course, the sensitivity of the model output to a specific parameter may vary with time. The sensitivity analysis was carried out for the following parameters: $k_d$ (partition coefficient water/sediment), $K_{alb}$ (migration rate for sediment interface layer — bottom sediment), $K_{bal}$ (migration rate for bottom sediment — sediment interface layer), $K_{sd}$ (migration rate for bottom sediment — deep sediment).

Table II summarizes the results of the sensitivity analysis of some models tested for the VAMP project (site specific geomorphological and hydrological parameters were not considered in this analysis). The radionuclide concentration in water is very sensitive to parameters related to the phenomena of $^{137}$Cs interaction with sediment.

The sensitivity analysis relevant to the amount of radionuclides deposited on the lake surface following a single-pulse accident deserves detailed discussion. As the output depends linearly on the deposition, the model is very sensitive to this quantity. Unfortunately, deposition is often evaluated with large uncertainty and therefore may represent one of the most important factors of model uncertainty.

References

Since 1986, a great effort has been made by Italian radioecology laboratories to monitor the levels of radionuclide concentrations in the various components of freshwater systems. The Dipartimento Ambiente of ENEA is carrying out a study to develop and validate models for assessing the behaviour of radionuclides in lakes and the radionuclide transfer from the catchment to water bodies. The research was partially financed by the European Commission (Contract No. FI3P-CT93-0073).

One of the goals of the study is the analysis of the seasonal effects controlling radionuclide turnover in deep lakes. The development of a thermocline during summer influences the vertical distribution of radionuclides deposited on waters of deep Italian lakes. Indeed, radionuclides deposited on the lake surface accumulate in the upper water layer (epilimnion), since the presence of a temperature gradient (thermocline) prevents diffusion of radionuclides to deeper layers. During the period of stratification (from spring to summer) the contribution of water from the drainage basin to lakes whose drainage areas are located in the Alpine region (northern Italy) is conspicuous because of the melting of ice and snow. This effect may imply a mean water residence time in the epilimnion that is significantly lower than the yearly mean water residence time in the whole lake. As a consequence, during water stratification, the total amount of radionuclides in the lake may dramatically decrease because of the outflow through the outlet from the lake that removes a conspicuous part of the contaminated water in the epilimnion. Of course, another important phenomenon of radionuclide removal from the lake water is migration of radionuclides to the bottom sediment. The deposition of $^{137}$Cs of Chernobyl origin in
northern Italy was significantly larger than the deposition in central Italy. For instance, around lake Bracciano (central Italy) the deposition was estimated as approximately 1000 Bq/m$^2$, whereas around lake Como (northern Italy) the deposition was higher than 20 000 Bq/m$^2$. This difference in the deposition of $^{137}$Cs reflects the initial concentration of radionuclides in lake water (90 Bq/m$^3$ in lake Bracciano, $>2000$ Bq/m$^3$ in lake Como, May 1986). On the contrary, several years after the Chernobyl accident, the concentrations in lakes of northern Italy (1 Bq/m$^3$ in water of lake Como) were very low compared with the concentrations in lakes of central Italy (lake Vico $\approx$ 70 Bq/m$^3$, lake Bracciano $\approx$ 10 Bq/m$^3$, lake Bolsena $\approx$ 25 Bq/m$^3$). This difference cannot be explained exclusively by the mean water retention times of the different lakes (lake Como 4 years, lake Vico 17 years, lake Bracciano 137 years, lake Bolsena $\approx$ 100 years) and the different mean depths of the lakes (Vico 20 m, Bracciano 89 m, Como 153 m, Bolsena $\approx$ 80 m), which are some of the most important factors influencing radionuclide dilution in water. The processes that may explain the different time behaviour of $^{137}$Cs concentration in waters of the above mentioned lakes are probably the initial removal of radionuclides from the epilimnion (in the volcanic lakes of central Italy this process was negligible, as stratification occurs during a dry season in this region), the interaction with sediments (very high values of $k_d$, the water–sediment radionuclide partition coefficient, were measured in lakes and rivers of northern Italy, whereas the values of $k_d$ measured in volcanic lakes of central Italy are significantly lower) and the removal of radionuclides from water due to sedimentation. In order to account for the above described phenomena, a model has been developed to predict the migration of radionuclides in lake systems; this model may explain the different behaviour that $^{137}$Cs of Chernobyl origin shows in deep lakes of northern Italy and in deep volcanic lakes of central Italy. The model may predict that this different behaviour is a consequence of water stratification, the seasonal dependence of water contribution from the drainage area and the characteristics of sediments.
The accident at the Chernobyl nuclear power plant in April 1986, in which large amounts of radioactive material were released into the environment, was the most serious accident that occurred in connection with the use of nuclear energy for electricity generation. The radiation levels from released radionuclides were highest in the immediate vicinity of the reactor, in the western part of the former Soviet Union and in the European countries. In other parts of the world, radionuclide contamination was due not only to external radiation but also to ingestion of contaminated food, mainly milk products.

After the accident, emergency measures were taken at the Chernobyl nuclear power plant and countermeasures were taken in many European countries. In countries such as Thailand, direct radiation exposure was not significant, but imported food products might have accounted for the collective dose equivalent.

![Graph](image_url)

**FIG. 1.** Average $^{137}$Cs activity in imported milk samples (numbers of samples are given in brackets).
TABLE I. SUMMARY OF THE RESULTS OF ENVIRONMENTAL RADIOACTIVITY MONITORING AFTER THE CHERNOBYL ACCIDENT (30 April 1986 to 30 June 1987)

<table>
<thead>
<tr>
<th>Type of sample</th>
<th>Radionuclides analysed</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{131}$I</td>
</tr>
<tr>
<td>Air</td>
<td>—</td>
</tr>
<tr>
<td>Rain water</td>
<td>—</td>
</tr>
<tr>
<td>Fresh water</td>
<td>—</td>
</tr>
<tr>
<td>Fallout</td>
<td>—</td>
</tr>
<tr>
<td>Soil</td>
<td>—</td>
</tr>
<tr>
<td>Grass</td>
<td>—</td>
</tr>
<tr>
<td>Beef</td>
<td>—</td>
</tr>
<tr>
<td>Snake head fish</td>
<td>—</td>
</tr>
<tr>
<td>Kale</td>
<td>—</td>
</tr>
<tr>
<td>Chinese swamp cabbage</td>
<td>—</td>
</tr>
<tr>
<td>Radish</td>
<td>—</td>
</tr>
<tr>
<td>Onion</td>
<td>—</td>
</tr>
<tr>
<td>Sea fish</td>
<td>—</td>
</tr>
<tr>
<td>Milk</td>
<td>—</td>
</tr>
<tr>
<td>Imported milk powder and baby food</td>
<td>—</td>
</tr>
</tbody>
</table>

By means of gamma spectrometry, radionuclides such as $^{131}$I, $^{134}$Cs and $^{137}$Cs have been analysed in environmental samples, foodstuffs, milk products and imported foods (see Table I and Fig. 1). The radionuclide contamination of imported milk powder collected from the commercial sector for the survey was generally found to be below the values set for guidance of the international trade.

Since no radioactivity ($^{137}$Cs) in milk products and in local foodstuffs was found in Thailand before the Chernobyl accident, the Ministry of Public Health of Thailand announced the following maximum permissible levels for $^{137}$Cs: fresh milk 7 Bq/L, milk products 21 Bq/kg, and cereals and other foodstuffs 6 Bq/kg. Thus, it is not permitted to import milk and milk products contaminated with $^{137}$Cs.
A set of computer models has been developed for estimation of the potential consequences to man in terms of radiation doses from the dumping of radioactive waste in the Kara Sea and the Barents Sea by the former Soviet Union. The dispersion of radionuclides has been simulated with a compartmental model, taking into account the hydrodynamic transport of water from the Arctic Ocean to the North Atlantic, including the European coastal waters.

The present dispersion model is a combination of two models: (1) a new version of a model [1] based on updated information of a regional box model used for radiological assessments in the north-west European coastal areas [2], and (2) a box model covering the Arctic Ocean and the North Atlantic [3]. The latter box model has been derived from a 3-D World Ocean general circulation model the results of which have been used for the design of a box structure in the Arctic Ocean and the surrounding waters. The resulting structure and water fluxes have been selected taking into account expert information based on experimental data from the Barents Sea [4]. The model includes the transfer of radionuclides to sea-bed sediments.

The basic input data for the model have been assumed to be identical with those used in the Marina study of the Commission of the European Communities [5]. This study dealt with an assessment of the radiological exposure of the population of the European Community from radioactivity in north European marine waters. Generic values from these data have been used where no other information was available.

Collective doses to the world population have been calculated from ingestion of radionuclides in contaminated seafood (fish, crustaceans and molluscs). The levels of contamination in seafood have been estimated from the levels in filtered sea water predicted by the model, and data on the human intake of seafood are based on official fishery statistics. The ingestion pathways covered here are believed to account for the main part of the collective dose.

The reliability of the predictions produced by the present model has not been tested quantitatively. However, it has been verified that the results (total collective
Collective doses have been calculated from single releases of 1 TBq of the radionuclides $^3$H, $^{60}$Co, $^{63}$Ni, $^{90}$Sr, $^{129}$I, $^{137}$Cs, $^{239}$Pu and $^{241}$Am into either the western Kara Sea or the Barents Sea. These results show that the doses are clearly higher for releases into the Barents Sea than for releases into the Kara Sea. They also show that doses from the radionuclides $^3$H, $^{60}$Co, $^{90}$Sr and $^{137}$Cs are derived mainly from seafood from the Barents Sea (fish and molluscs), while doses from the radionuclides $^{239}$Pu and $^{241}$Am are delivered through marine produce from areas far from the Arctic Ocean.

Furthermore, collective doses have been calculated for two release scenarios, both of which are based on information on the dumping of radioactive waste into the Barents Sea and the Kara Sea by the former Soviet Union [6]. A worst-case scenario was selected according to which it was assumed that all radionuclides were available for dispersion in the marine environment at the time of dumping, thus leaving no time for decay of short lived radionuclides prior to release. For the second scenario it was assumed that radionuclides in solid waste were released over a period of 100 years from the time of dumping. All liquid waste was assumed to be available for dispersion at the time of discharge in both scenarios.

The collective doses (calculated to 1000 years) for the worst-case scenario are about 200 man-Sv and for the second scenario about 100-man-Sv. In both cases, $^{137}$Cs is predicted to dominate the peak collective dose rates, which are about 2 man-Sv per year for scenario 1 and about 0.3 man-Sv per year for scenario 2.

A comparison with the results from the Marina study shows that the radiological sensitivity (collective dose per unit discharge, e.g. man-Sv per PBq) of discharges of $^{137}$Cs into the Barents Sea and the Kara Sea is about one order of magnitude lower (4 man-Sv/PBq) than that of discharges of $^{137}$Cs into the Irish Sea (50 man-Sv/PBq). The main reason for this difference is the low production of seafood for human consumption in the Polar waters compared with that in the European coastal waters.

REFERENCES

VALIDATION OF THE MODELLING OF $^{129}$I TRANSFER THROUGH THE AIR-GRASS-MILK PATHWAY

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Moor Row, Cumbria, United Kingdom

At the site of British Nuclear Fuels plc in Sellafield, Cumbria, reprocessing of nuclear fuel results in the emission of $^{129}$I to the atmosphere. Doses to the critical group of people living close to the site and consuming locally produced food are assessed, using simple compartment equilibrium models, and compared with recommended dose limits for members of the public. Measured emissions to the atmosphere, simple Gaussian dispersion with effective emission heights to allow for building wake effects [1], generalized dry and wet deposition parameters [2], and equilibrium transfer to food products [3] are used in the model to predict the concentrations in food and the doses to the critical group. For the reprocessing plant, $^{129}$I is one of the most important contributors to the dose to the critical group, mainly through the grass-milk pathway.

The simple equilibrium approach for $^{129}$I is subject to a number of uncertainties, in the measurement of the emissions from the stacks, in the dispersion model as applied to a site with building wakes, and in the choice of deposition and transfer parameters. Recent developments in the measurement of low concentrations of $^{129}$I in environmental materials by a neutron activation method, with a detection limit below 10 mBq/kg for milk and vegetables, have allowed some of these uncertainties to be addressed [4].

In a research project at the Westlakes Research Institute the concentrations of particulate, inorganic and organic forms of $^{129}$I in air sampled at ground level have
Table I. Measured Deposition Velocities for Various Iodine Species [6]

<table>
<thead>
<tr>
<th>Iodine species</th>
<th>Air to vegetation deposition velocity $V_g$ (ms$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particulate</td>
<td>0.002</td>
</tr>
<tr>
<td>Elemental (I$_2$)</td>
<td>0.01</td>
</tr>
<tr>
<td>Hypoiodous acid (HOI)</td>
<td>$\leq 0.001$</td>
</tr>
<tr>
<td>Organic iodides (CH$_3$I and others)</td>
<td>0.000001-0.00005</td>
</tr>
</tbody>
</table>

been measured, using a three-component sampling system and extraction onto ion exchange resin prior to neutron activation at the Imperial College Research Reactor at Silwood Park [5]. These measurements indicate that, for current discharges from the plant, the dominant species of atmospheric $^{129}$I is organic and is believed to be methyl iodide. From values of the dry deposition velocity, $V_g$, available in the literature [6], this would lead to a $V_g$ value lower by at least a factor of 100 than the value of $1 \times 10^{-2}$ ms$^{-1}$ that is used in the model (see Table I). As part of the project, the deposition velocity was estimated from the ratio of the deposition rate on grass to the mean air concentration, for grass exposed to rain and for grass covered to exclude wet deposition.

The average measured value of $V_g$ for grass, including the effects of wet deposition, was $8.8 \times 10^{-4}$ ms$^{-1}$. It must be remembered, however, that field values of $V_g$ are subject to experimental measurement difficulties associated with air sampling and deposition collection, and also depend on the condition of the grass and the local atmospheric boundary layer.

Transfer coefficient from grass to milk

For samples of field grass and milk from two farms close to Sellafield, measurements of $^{129}$I were made at the Institut für Physikalische Chemie der Universität Bonn, using the neutron activation method [7]. From these measurements, the transfer coefficients from grass to milk were $6.6 \times 10^{-3}$ and $7.7 \times 10^{-3}$ dL$^{-1}$, with a mean value of $7.1 \times 10^{-3}$ dL$^{-1}$. These values are in good agreement with the value used in the critical group dose model, $5 \times 10^{-3}$ dL$^{-1}$ [3]. Although these measurements support the modelling of the transfer from grass to milk, the model as a whole overpredicts the concentration in milk by factors of 33.6 and 40.7, with a mean value of 37.2. This is believed to be partly the result of the lower deposition of organic $^{129}$I compared with that assumed by the model, but overestimation of stack emissions may also be a factor.
Conclusions

The equilibrium model assumes a transfer coefficient from grass to milk of $5 \times 10^{-3}$ Bq/L per Bq/d for intake by cows. This value has now been confirmed by measurements in matched samples of grass and milk from two farms close to Sellafield. However, the model as a whole predicts concentrations in milk that are an order of magnitude higher than those measured. These findings are subject to some uncertainty in the source term measurements and in the simple dispersion modelling used. Recent data from a research project to measure $^{129}$I in air and deposition near the site have shown that the speciation of $^{129}$I in air at the site perimeter is predominantly organic, probably methyl iodide. The deposition velocity measurements show that the value of $1 \times 10^{-2}$ m$^{-1}$ s$^{-1}$ assumed in the model, which would be appropriate for inorganic $^{129}$I, is too high for these circumstances and would partly explain the low concentrations found in milk.

Current work aims at substantiating these findings and establishing a deposition velocity value appropriate to the chemical form of $^{129}$I near the Sellafield reprocessing plant. This work will assist in the development of a more realistic environmental model for $^{129}$I and lead towards a greater degree of realism in the modelling of critical group doses.

ACKNOWLEDGEMENT

Professor D.C. Aumann, of the Institut für Physikalische Chemie der Universität Bonn, made the measurements of $^{129}$I on samples of field grass and milk.

REFERENCES

IAEA-SM-339/12P

THE "REFERENCE BIOSPHERES METHODOLOGY"
IN PERFORMANCE ASSESSMENTS FOR
RADIOACTIVE WASTE REPOSITORIES

F. VAN DORP
Working Group leader,
BIOMOVS II Working Group on Reference Biospheres

Status of biosphere modelling for radioactive waste repositories

Performance criteria for waste repositories are, in general, expressed in, or based on, dose or risk limits for individual members of the population.

Problematic aspects of biosphere modelling for geological waste disposal:
— Potential release and exposure occur far in the future;
— Reliable predictions of future human behaviour and the human environment at the time of the release are impossible.

Characteristics of biosphere modelling for waste disposal:
— The biosphere is considered not to be a barrier and therefore the effort invested in modelling the biosphere is often less than that invested in modelling the near field and geosphere components of the repository system.
— The biosphere may be an important factor in the performance of the repository because this is where any input occurs and criteria are set in terms of that input.

For these and other reasons, often unexplained differences between different countries exist in the approaches to biosphere modelling and in the features, events and processes included in the models.
BIOMOVS II Working Group on Reference Biospheres

The Working Group on Reference Biospheres expects to provide:

— A recommended methodology within the assessment of radioactive waste disposal, which is consistent for different types of radioactive waste and disposal concepts. This should include the justification, arguments and documentation for all steps in the recommended methodology.

— An internationally developed and structured list of features, events and processes (FEPs), which can be used for the development of biosphere models for specific assessments.

— Example(s) of how to apply the methodology. If these examples are developed in the context of a generic assessment, they can be defined as 'Reference Biospheres'. These could be used, for example, (a) for generic site independent evaluation of disposal plans, (b) as 'stylized biospheres', (c) as benchmarks for comparisons with other assessments, and (d) as sources of detailed information on biosphere modelling for waste disposal assessments.

At present, the Working Group has achieved:

— A biosphere FEP list in an electronic database (preliminary),
— A draft of the methodology,
— A draft definition of an example of the methodology.

These studies will be finalized and, in addition, the FEP list and methodology will be tested with the example, and the example will be tested in co-operation with the Working Group on Complementary Studies.

The methodology which is being developed includes:

— Identification of FEPs which are relevant for the case to be assessed (screening of the international biosphere FEP list);
— Scenario analysis, which leads to the definition of the required conceptual models;
— Development of calculational models from the conceptual models;
— Definition of the parameters and their input data;
— Production of calculational results;
— Interpretation of these results.

Conclusions

The methodology that will be proposed by the Working Group on Reference Biospheres will, if applied, achieve:

— Harmonization of the modelling approaches in the different countries, taking into account variations in modelling and protection objectives;
— Comparability of the biosphere models and their results.

The studies in and for the Working Group have identified that the translation of regulations (dose or risk limits) into practical assessment criteria can cause considerable uncertainty. This should be discussed between regulators and biosphere modellers.

The mathematical aspects of constructing calculational models and their computer codes are not seen as significant problems. However, since the biosphere models are used to assess impacts over considerable time spans, procedures to derive effective parameter values from parameters with significant spatial and temporal (short time) variation will have to be developed.

It might be proposed to use ‘self-sufficient communities’ to avoid the problem of having to predict an expected community and its variations, and uncertainties at the time and point of release. However, at present, the knowledge of parameter values for radionuclide transfer through the exposure pathways of such a community is not yet sufficient for incorporation in biosphere models for the assessment of radioactive waste repositories.

**BIBLIOGRAPHY**


Biosphere modelling for Swiss performance assessments

Biosphere modelling is an integral part of the Swiss performance assessments for radioactive waste disposal. The Terrestrial-Aquatic Model of the Environment (TAME) has been developed recently in Switzerland and used subsequently in performance assessments for repositories of high level waste (HLW) and intermediate/low level waste (I/LLW). From the use of TAME in these assessments, conclusions can be drawn concerning the benefits of detailed models versus simple, conservative models for radioactive waste disposal assessments.

TAME [1] has been constructed with the aim of being flexible and easy to use. It consists of two main parts:

— Calculation of the transport of radionuclides in the physical compartments of the biosphere — aquifer, soil, surface water and aquatic sediments;
— Calculation of the radiological impact of the uptake and accumulation of radionuclides into the food-chain.

The flexibility of TAME derives from the approach taken in the development of the conceptual and mathematical models. The starting point was the identification of all features, events and processes (FEPs) relevant to conditions (both current and future) at the Swiss sites. Detailed mathematical representations of the FEPs were then derived on the basis of mass balance considerations for transport of both liquid and solid materials. These fluxes determine the transport of contaminants in the biosphere. The resulting model is therefore constructed on a set of physical principles that are applicable to any site for which data can be obtained, i.e. TAME is generic in scope but specific by the nature of the database used for a particular application.
This approach has evolved from earlier experiences in BIOMOVS and has played a role in the current BIOMOVS II Working Groups on Reference Biospheres [2] and Complementary Studies [3].

Swiss assessment experience

In Switzerland, studies are being undertaken for two types of repository — one for I/LLW and one for HLW. The biosphere regions that could be affected by releases from these prospective repositories are quite different. In the case of I/LLW [4] the modelled biosphere is a moderately sized valley in central Switzerland, with a broad valley floor, medium sized river and steep sides. The repository is planned to be accessed by horizontal tunnels into the Wellenberg mountain at the valley side. For the HLW [5] repository, the crystalline rocks in northern Switzerland have been selected for investigation. The biosphere here is defined by the Rhine valley.

The results from these assessments illustrate different aspects of biosphere modelling. These are related to the time-scales of the releases to the biosphere and the time-scales needed for steady state concentrations to be established in the biosphere. In the systems analysed in Switzerland, the time to steady state can be longer than the time-scale for major alteration of the surface environment.

The temporal response of the biosphere model cannot be adequately characterized without a fundamental understanding of the FEPs acting in the modelled region; it can be concluded that there is no significant difference between biosphere modelling for the different classifications of radioactive waste — both require the same level of understanding of the biosphere FEPs.

It is often assumed that direct consumption of water from a well with minimal dilution is a sufficiently robust and conservative basis for estimation of the radiological impact of the release of radionuclides from the geosphere. Calculations from the HLW assessment indicate that this is not always the case and that, for some radionuclides, the doses calculated in the full biosphere representation can exceed those calculated assuming a well with minimum dilution (see Table I). An adequate representation of the radiological consequences of the release from the geosphere, based on a single exposure mechanism, should take into account the potential for accumulation in all parts of the biosphere with which the exposed individual might come into contact. This cannot be satisfactorily achieved without a detailed FEP database and it can be concluded that the use of simple dilution biosphere models to represent the radiological impact is not reliably conservative; neither does it provide any information on the time-scale of the biosphere response.

In stochastic sensitivity analyses carried out using TAME and using the databases for the Swiss sites, it can be seen that when standard tests of parameter sensitivity are used, no single parameter or group of parameters emerge as having a dominating influence on the radiological impact of the releases to the biosphere.
TABLE I. COMPARISON OF DOSES ARISING FROM THE KRISTALLIN-I REFERENCE CASE (FULL BIOSPHERE MODELLING) WITH RESULTS USING THE REFERENCE CASE GEOSPHERE RELEASE WITH A MINIMUM DILUTION WELL FOR WHICH THE ABSTRACTION RATE IS ASSUMED TO BE THE MINIMUM ECONOMICALLY VIABLE

<table>
<thead>
<tr>
<th>Nuclide/decay chain</th>
<th>Minimum dilution well biosphere</th>
<th></th>
<th>Kristallin-I Reference Case biosphere</th>
<th></th>
<th>Ratio of well dose to full biosphere dose</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Maximum dose (mSv/a)</td>
<td>Time of maximum (years)</td>
<td>Maximum dose (mSv/a)</td>
<td>Time of maximum (years)</td>
<td>Maximum pathway</td>
</tr>
<tr>
<td>$^{99}$Tc</td>
<td>$1.4 \times 10^{-7}$</td>
<td>$1.8 \times 10^{6}$</td>
<td>$5.1 \times 10^{-7}$</td>
<td>$1.8 \times 10^{6}$</td>
<td>Milk</td>
</tr>
<tr>
<td>$^{79}$Se</td>
<td>$1.7 \times 10^{-6}$</td>
<td>$6.5 \times 10^{4}$</td>
<td>$2.3 \times 10^{-6}$</td>
<td>$7.2 \times 10^{4}$</td>
<td>Meat</td>
</tr>
<tr>
<td>4N + 3</td>
<td>$7.7 \times 10^{-6}$</td>
<td>$&gt; 10^{7}$</td>
<td>$7.1 \times 10^{-6}$</td>
<td>$&gt; 10^{7}$</td>
<td>Meat</td>
</tr>
<tr>
<td>$^{126}$Sn</td>
<td>$4.09 \times 10^{-9}$</td>
<td>$8.3 \times 10^{5}$</td>
<td>$3.6 \times 10^{-9}$</td>
<td>$8.8 \times 10^{5}$</td>
<td>Grain</td>
</tr>
<tr>
<td>$^{93}$Zr</td>
<td>$2.2 \times 10^{-8}$</td>
<td>$1.0 \times 10^{7}$</td>
<td>$1.3 \times 10^{-8}$</td>
<td>$&gt; 10^{7}$</td>
<td>Meat</td>
</tr>
<tr>
<td>4N + 1</td>
<td>$1.8 \times 10^{-6}$</td>
<td>$&gt; 10^{7}$</td>
<td>$8.9 \times 10^{-7}$</td>
<td>$&gt; 10^{7}$</td>
<td>Root vegetables</td>
</tr>
<tr>
<td>$^{135}$Cs</td>
<td>$6.1 \times 10^{-4}$</td>
<td>$2.8 \times 10^{5}$</td>
<td>$2.3 \times 10^{-4}$</td>
<td>$2.8 \times 10^{5}$</td>
<td>Meat</td>
</tr>
<tr>
<td>$^{59}$Ni</td>
<td>$3.9 \times 10^{-13}$</td>
<td>$6.9 \times 10^{5}$</td>
<td>$8.6 \times 10^{-14}$</td>
<td>$7.0 \times 10^{5}$</td>
<td>Grain</td>
</tr>
<tr>
<td>4N</td>
<td>$4.8 \times 10^{-7}$</td>
<td>$&gt; 10^{7}$</td>
<td>$8.8 \times 10^{-8}$</td>
<td>$&gt; 10^{7}$</td>
<td>Dust inhalation</td>
</tr>
<tr>
<td>$^{107}$Pd</td>
<td>$5.6 \times 10^{-13}$</td>
<td>$4.3 \times 10^{6}$</td>
<td>$5.9 \times 10^{-14}$</td>
<td>$4.3 \times 10^{6}$</td>
<td>Grain</td>
</tr>
<tr>
<td>4N + 2</td>
<td>$9.0 \times 10^{-6}$</td>
<td>$&gt; 10^{7}$</td>
<td>$4.1 \times 10^{-7}$</td>
<td>$&gt; 10^{7}$</td>
<td>Well water</td>
</tr>
</tbody>
</table>

Results are shown for all the radionuclides and decay chains assessed in Kristallin-I. The ratios show that in some cases the model for the full biosphere (with intrinsically 22.1 times more dilution than the well biosphere) can give higher doses than the well biosphere ($^{79}$Se and $^{99}$Tc). In most cases the ratio of doses is much less than the ratio of dilutions in the two model representations, indicating the effects of accumulation as modelled in the detailed biosphere. In this HLW assessment, releases from the geosphere occur at times in the far future and the accumulation time-scales in the biosphere have little effect on the time of maximum dose. For earlier peaking releases, however, this is not the case ($^{79}$Se) and the role of the biosphere processes is seen. For I/LLW disposal concepts, the time-scales in the biosphere can be much more apparent.
Such results are not uncommon [6] in biosphere modelling. Consequently, it can be concluded that, although biosphere models must necessarily be complex in structure, the precision to which data values must be known is not paramount. This supports the idea that a set of reference biosphere scenarios could be developed (e.g. inland valley, regional river, coastal biosphere) which would be sufficient for the evaluation of radiological impacts in the far future.

Conclusions

From the biosphere modelling for Swiss performance assessments for radioactive waste disposal the following conclusions can be drawn:

— It is possible and desirable to develop a generically applicable biosphere model based on a thorough review of biosphere FEPs. TAME is one such model; there are others, and the work of the BIOMOVS II Working Groups on Reference Biospheres [4] and Complementary Studies [5] should resolve differences of approach on the international scene.

— The degree of complexity required for a particular biosphere model does not depend on the classification of the waste or the location of the disposal site.

— The response of any particular biosphere can only be adequately described when the relevant FEPs are thoroughly understood.

— For long time-scales and chronic releases, transport of contaminants on solid material is of particular importance for the sorbing radionuclides.

— Simple dilution models are not reliably conservative and should only be used as a means of comparing the releases from the geosphere.

— Time dependent calculations should be carried out, particularly when the characteristic time-scales of the subject biosphere are long compared with those for major changes in the biosphere.

There are a number of unresolved issues at this stage of biosphere model development:

— In order to produce suitable sets of reference biospheres, a review of the database for alternative scenarios is required. This is also necessary because the construction of models based on FEP analysis places great emphasis on the mathematical representation of the FEPs; for example, methods for treating seasonally varying data need to be optimized. This can be problematic since the database required for such models is extensive.

— The relative insensitivity of the biosphere models to uncertainty in many of the input values of the models should be confirmed by further stochastic sensitivity analysis, with a broad range of analysis tools.
REFERENCES


IAEA-SM-339/24P

EVALUATION OF THE RADIOLOGICAL CONSEQUENCES OF RADIOACTIVE RELEASES TO THE AQUATIC ENVIRONMENT FROM THE JURAGUA NUCLEAR POWER PLANT DURING NORMAL OPERATION

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Centro de Protección e Higiene de las Radiaciones, Secretaría Ejecutiva para Asuntos Nucleares, Havana, Cuba

The individual maximum dose from different exposure pathways caused by releases of radioactive material from the Juragua nuclear power plant to the aquatic environment during normal operation (Fig. 1) has been evaluated, using the compartment model recommended in Refs [1, 2].

For estimation of different radionuclide concentrations, the model proposed in Ref. [1] does not specify how to calculate the fractional loss rate of water from a mixed volume for non-tidal seas. We have suggested an expression for calculating the water fractional loss rate that can be applied to discharge configurations similar to that of the Juragua nuclear power plant (Fig. 2).

In order to estimate the volume of the mixed water, we have calculated the depth of the mixture, taking into account the buoyancy of the effluent, its outfall and
FIG. 1. Discharge configuration of the Juragua nuclear power plant.

FIG. 2. Exposure pathways for radioactive releases to the sea from the Juragua nuclear power plant.

its moment. We have also determined the radionuclides and the critical exposure pathways using the data in Refs [1, 3] and the specific site characteristics. For the assessment of model uncertainties for critical radionuclides and exposure pathways we have employed the method of simple random sampling [4].

The maximum annual individual doses for different radionuclides and exposure pathways as well as the magnitude of uncertainties have been summarized in tables and graphics.
REFERENCES


IAEA-SM-339/25P

ASSESSMENT OF THE RADIOLOGICAL CONSEQUENCES OF RADIOACTIVE RELEASES TO THE ATMOSPHERE FROM THE JURAGUA NUCLEAR POWER PLANT

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For the Juragua nuclear power plant located in the vicinity of Cienfuegos City, we have evaluated the expected radiological consequences of radioactive releases during routine operation in the case of accidents. For the assessment of the radiological impact of routine releases we have used the computer code DIFFU, which was elaborated on the basis of the methodology recommended by the IAEA [1].

The atmospheric dispersion has been evaluated using the results of aerological and meteorological observations carried out at the siting stage and results from continuous in situ investigations.

Maximum values of the dilution factor were observed for the sectors sited in the direction of the sea: SW — $4.3 \times 10^{-8}$ s/m$^3$, WSW — $6.6 \times 10^{-8}$ s/m$^3$, and SSW — $7.1 \times 10^{-8}$ s/m$^3$. The maximum values for 'terrestrial sectors' (NNW and NNE) were obtained for distances inside the protection area ($r = 2.5$ km).
Transfer factors for $^{137}$Cs and $^{90}$Sr from soil to plants and from pasture to milk have been derived from the average concentrations of these radionuclides in different media. Average concentrations of $^{137}$Cs and $^{90}$Sr have been obtained from measurements made during several years in the surroundings of the installation by gamma spectrometry and radiochemical methods. In all cases the transfer factors showed a lognormal distribution. Most of the values for the soil to pasture and soil to crop transfer factors were higher than the default values recommended in Ref. [1].

For fission and activation radionuclides, the soil to sugar-cane transfer factor has been derived from the relationship with stable elements determined experimentally under controlled agrochemical conditions. The samples have been analysed by atomic absorption spectrophotometry.

We have also performed an analysis of the sensitivity of the employed migration model to parameter variation. Uncertainties have been evaluated using the simple random sampling method [2]. For the assessment of the statistical distribution of parameters, different criteria from experts have been taken into account.

The obtained values of the effective equivalent doses for the critical groups are two orders of magnitude lower than the limits for members of the public recommended by the International Commission on Radiological Protection (ICRP). The risks of stochastic effects are several orders of magnitude lower than the risk of naturally induced cancers in the region.

The radionuclides with the largest impact on effective equivalent doses have been found to be $^{88}$Kr, $^{133}$Xe, $^{135}$Xe, $^{131}$I and $^{137}$Cs. The total contribution of these radionuclides represents 80% of the total dose.

We have also assessed the radiological consequences of likely accidental releases from the installation, using the computer code SECAN, which incorporates the models described in ICRP publication 29 [3]. We have used site specific values for the model parameters or, if such values were not available, we have used values reported in the relevant literature.

For different source terms and meteorological conditions, the dimensions of the affected areas, the zones of higher radiological risk and the doses for different pathways have been determined. For severe accidents, with PWR4 and SST1 source terms, deterministic effects are expected only for small inhabited areas close to the site ($r < 7$ km). It is recommended to take into account early evacuation of those areas in emergency planning.

The main contributions to the collective doses in the event of a severe accident are external irradiation and ingestion of contaminated food, in approximately equal parts. For the region studied, a relatively important contribution from the sugar-cane food-chain is expected.

Finally, we have identified the priorities for radioecological investigations on the site and defined effective means and suitable places for carrying out environmental radiological monitoring.
USE OF THE LAGRANGIAN-EULERIAN MODEL OF AEROSOL TRANSFER IN THE INTERPRETATION OF THE SPOT PATTERN OF RADIOACTIVE CONTAMINATION DUE TO THE CHERNOBYL ACCIDENT

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For an estimate of the impact of serious accidents at nuclear power plants such as that at Chernobyl on the flora and fauna of polluted territories it is very important to understand the meteorological conditions resulting in the formation of a complicated, inhomogeneous structure of the primary radioactive contamination. We have performed numerical experiments to clarify some aspects of the formation of a spot pattern of ground pollution, applying the regional diffusional model LEDI (Lagrangian–Eulerian Diffusional model). The model makes it possible to calculate the transport of pollutants released from a time dependent source in inhomogeneous and time dependent meteorological conditions [1]. The Lagrangian part of the model describes the transport of air portions along different wind directions, taking into account the curvature of the surface of the Earth. For the processes taking place inside every air portion, we have applied conventional time dependent two-dimensional equations of turbulent diffusion, the coefficients of which were determined using a semi-empirical procedure and the actual state of the planetary boundary layer (Eulerian part of LEDI). Vertical and horizontal turbulent transfer, decay of radionuclides and wet and dry deposition have been taken into account.
FIG. 1. Results of modelling (north-eastern part of the contaminated area).

TABLE I. MEASURED AND MODELLED GROUND DEPOSITION OF $^{137}$Cs (Ci/km$^2$) FOR EACH DAY OF EMISSION$^a$

<table>
<thead>
<tr>
<th>Date</th>
<th>Kiev measured</th>
<th>Kiev modelled</th>
<th>Baryshevka measured</th>
<th>Baryshevka modelled</th>
</tr>
</thead>
<tbody>
<tr>
<td>1986-04-30</td>
<td>0.0035</td>
<td>0.31</td>
<td>0.0020</td>
<td>0.01</td>
</tr>
<tr>
<td>1986-05-01</td>
<td>0.096</td>
<td>0.12</td>
<td>0.20</td>
<td>0.19</td>
</tr>
<tr>
<td>1986-05-02</td>
<td>0.32</td>
<td>0.21</td>
<td>0.071</td>
<td>0.05</td>
</tr>
<tr>
<td>1986-05-03</td>
<td>0.092</td>
<td>0.08</td>
<td>0.0063</td>
<td>0</td>
</tr>
<tr>
<td>1986-05-04</td>
<td>0.011</td>
<td>0</td>
<td>0.0035</td>
<td>0</td>
</tr>
<tr>
<td>1986-05-05</td>
<td>0.004</td>
<td>0.01</td>
<td>0.0026</td>
<td>0</td>
</tr>
</tbody>
</table>

$^a$ 1 Ci = 37 GBq.
The two parts of the model have been connected by using in the Lagrangian part the wind directions obtained from an average over the concentration profile from the Eulerian part.

To model transportation and deposition of radioactivity during the fire at Unit 4 of the Chernobyl NPP, data from radiosonde measurements for stations in Homel, Kiev and Kursk have been used to obtain parameters of the planetary boundary layer and the turbulence and trajectory of the radioactive jet; the modelled time dependence of the rate at which radioactive material enters the atmosphere has been used. The model of a two-dimensional hot, turbulent jet with Stommel’s entrainment has been used for estimating the initial height of the radioactive cloud. Furthermore, the diurnal change of the parameters of the planetary boundary layer due to changes of temperature stratification has been taken into account. At large distances from the source, formation of isolated spots of radioactive contamination with low or high deposition is possible. Some results of modelling are presented in Fig. 1 (northeastern part of the contaminated area) and in Table I (southern part of the area). The results obtained show that it is possible to obtain a realistic pattern of radioactive pollution.

REFERENCE

IMPACT OF ATMOSPHERIC NUCLEAR WEAPONS TESTS
AND OF THE CHERNOBYL ACCIDENT
ESTIMATED THROUGH THE $^{137}$Cs BODY BURDEN
OF PEOPLE IN THE CZECH REPUBLIC

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National Institute of Public Health,
Prague, Czech Republic

Data on environmental contamination from different sources are used for reconstruction of the doses from nuclear weapons tests in the atmosphere and from other releases of radionuclides into the environment.

After the Chernobyl accident, extensive systematic measurements of the amount of radionuclides in various environmental components, aimed at assessment of the doses to people, have been carried out by laboratories of the Radiation Monitoring Network of the Czech Republic [1]. There are extensive databases on the contamination of the environment, on food-chains and internal contamination of people, which can be used for model prediction exercises [2], re-evaluation of doses, etc. Models developed for the purpose of assessment of doses from internal contamination are quite complex and require many parameters for the transport processes from contaminated air or from fallout to man. Most such models have the tendency to overpredict the body burdens and the resulting doses to humans [2]. To determine doses in the most direct way possible with minimum assumptions, the dose due to internal contamination by artificial radionuclides was estimated from whole body counting. Whole body measurements were performed with the whole body counter of the Centre of Radiation Hygiene of the National Institute of Public Health in Prague. The measurements were carried out with semiconductor HPGe detectors. The aim was to exclude systematic errors in whole body counting when switching from the old detection system employing four NaI(Tl) scintillation detectors to HPGe detectors.

People were measured with the semiconductor detector in the sitting position [3]. The detector was placed in a room with a shielding made of old steel (originating before 1945). Five different HPGe detectors were used in the long term study of the retention of $^{137}$Cs in a reference group of about 30 persons living in Prague. Calibration of detectors for whole body counting was carried out, if possible, with phantoms or volunteers, but, because of unexpected failures of some detectors during the study, a simplified way of calibration by means of transfer of calibration data from one HPGe detector to another, which is suitable for emergency
Survey by means of measurements of $^{137}\text{Cs}$ in urine

- Nation wide
- In four regions

**FIG. 1.** Time course of the retention of $^{137}\text{Cs}$ in the Czech population.

In order to increase the number of persons involved in the study and to gain data for people who are not easily available for whole body counting, an indirect method of assessment of internal contamination through measurement of the daily excretion rate of caesium in urine was used [4]. Nationwide surveys have been made once a year, starting in 1987; the results are included in Fig. 1.

Data on the environmental contamination of the territory of the Czech Republic after atmospheric nuclear weapons tests have been obtained mainly by gross beta methods. Extensive studies concerning the contents of $^{90}\text{Sr}$ in milk and in bones of people have also been performed. However, the most comprehensive information about $^{137}\text{Cs}$ contamination can be derived from whole body counting of people, which has been performed routinely in the Czech Republic since 1965. The measurements of people in the 1960s were aimed especially at a group of people contaminated by dial paints. Other cases of internal contamination were also studied. Since for the analysis of scintillation spectra the so-called stripping method was used, it was necessary to determine $^{137}\text{Cs}$ and $^{40}\text{K}$. For the evaluation of yearly averages of $^{137}\text{Cs}$, a cumulative log-normal distribution was used, which allowed inclusion of values below the detection limit. Recently, a retrospective study was performed in which yearly averages of $^{137}\text{Cs}$ for the period 1965–1993 were evaluated. As a control of the quality of the measurement, the results for $^{40}\text{K}$ were used. The time course (yearly averages) of $^{137}\text{Cs}$ retention is shown in Fig. 2.
Minimum and maximum values found in UNSCEAR reports in given period in the temperate zone of the northern hemisphere

FIG. 2. Time course of the retention of $^{137}$Cs in the Czech population. Minimum and maximum values (from UNSCEAR reports) for the temperate zone of the Northern hemisphere.

Calculation of effective dose equivalents after the Chernobyl accident is strictly based on data from measurements on the Czech territory. Up to the end of 1993, the dose from internal contamination by $^{137}$Cs and $^{134}$Cs was about 107 $\mu$Sv. For the estimation of doses due to nuclear weapons tests, data from the literature were used in addition to experimental data.

REFERENCES

[1] Report on the Radiation Situation in ČSSR after the Chernobyl Accident, Rep. IHE-CHZ, Prague (1986); presented to UNSCEAR.


Since the early 1980s the National Radiological Protection Board (NRPB) has carried out a variety of assessments of the radiological impact of radioactive discharges, both routine and accidental, from the Sellafield site of British Nuclear Fuels plc (BNFL) in west Cumbria. Many of these assessments were of individual doses and/or risks, and these are discussed here. The individual dose assessments may be split into those which calculate the critical group dose and those which calculate the dose or risk to average members of the population.

In 1992, BNFL applied to the United Kingdom's authorizing departments, Her Majesty's Inspectorate of Pollution and the Ministry of Agriculture, Fisheries and Food, for revised authorizations to discharge radionuclides from its Sellafield site. This application took into account the proposed startup of the new Thermal Oxide Reprocessing Plant (THORP) and the introduction of various new effluent treatment plants such as the Enhanced Actinide Removal Plant (EARP). Because of requests for advice, the NRPB undertook an independent assessment of the potential doses to critical groups [1] arising from discharges made at the proposed limits. Since the object of the assessment was to predict future doses, mathematical models were used. The estimated annual effective dose to the critical group for marine discharges (following the startup of EARP) was 140 μSv, of which about 10% was due to atmospheric discharges. The annual effective dose to the critical group for atmospheric discharges was 240 μSv, of which 25% was due to direct irradiation from the Calder Hall reactors and about 10% to marine discharges.

The possibility of additivity across all feasible exposure pathways was addressed in more detail in another study of critical group doses in the UK [2]. This work included Sellafield, and the main aim was to assess annual doses representative of those in the early 1990s, taking into account a variety of pathways additively. It was a retrospective study in which environmental measurements were used wherever possible. This meant that doses could be due to multiple sources and from historical as well as current discharges from a site. This was indeed the case for the critical group for marine discharges at Sellafield where almost half of the annual dose of 250 μSv was due to discharges of natural radionuclides from a nearby phosphoric acid plant. If this source of exposure is excluded, the highest dose due entirely to activities at Sellafield was to individuals who live close to the Calder Hall power station.
The doses and risks to average individuals have been the subject of several dose reconstruction projects. The first of these was carried out at the request of an inquiry headed by Sir Douglas Black which sought to investigate whether radiation, and in particular that due to radioactive discharges from Sellafield (formerly Windscale), was the cause of a raised incidence of childhood leukaemia found in the nearby town of Seascale [3]. All the principal sources of radiation exposure for the period since operations began at Sellafield were considered, such as routine discharges and various accidental releases. Radiation from weapons fallout and from natural and medical sources was also considered. Measured data were taken wherever possible; otherwise, use was made of predictive models. The risks of leukaemia and other fatal cancers were calculated for seven cohorts of 175 children, assumed to have been born at five-yearly intervals from 1945 to 1975. The assessment indicated that in the study group of 1225 children followed to 1980, 0.1 radiation induced leukaemias would be expected in children under the age of 20. Natural radiation accounted for about 70% of this risk and the contribution from Sellafield operations was about 11%.

Additional data on Sellafield discharges became available after the completion of the first study and so the risks were reassessed [4]. The main difference in the input data was an increase in the estimate of the mass of irradiated uranium fuel particles released from the Windscale piles in the mid 1950s. This resulted in an increase in the estimated risk of radiation induced leukaemia resulting from Sellafield's operations from 0.011 to 0.016. The estimated risk was 250 times lower than that which would account for the observed number of four leukaemias in the study group. Work has recently been done on a second reassessment in response to a request from the Committee on the Medical Aspects of Radiation in the Environment (COMARE) in the UK which is further investigating the continuing cancer cluster. This work, which placed greater emphasis on sensitivity and uncertainty analysis, extended the assessment to 1992 and incorporated further revisions to discharge data, dosimetric models and environmental models.

An assessment has been made of the annual doses that could be received by people who make use of the Cumbrian coastal area. Current doses were estimated for residents and visitors to different parts of the area, with the emphasis on average exposures. The development of doses over the next 200 years was also assessed on the basis of assumptions about changes in waste management practices at Sellafield and in the geography of the coastal area. As expected, the highest doses would be received by those who live closest to the site. The general future trend was for doses to decline [5].
REFERENCES


IAEA-SM-339/33P

MODELS OF RISK ASSESSMENT OF WATER USE IN RUSSIAN REGIONS EXPOSED TO RADIOACTIVE CONTAMINATION

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1. INTRODUCTION

A number of water bodies in the Russian territory has been exposed to radioactive contamination. Models have been developed for radiation risk assessment of water use in regions where nuclear power stations are located. The models have been used for analysis of the radiation situation in cooling ponds of nuclear power plants under normal operating conditions and in the case of a radiation accident, as well as in the lakes of the Bryansk region in the territory affected by the Chernobyl accident, in the Techa river basin — the area affected by the industrial complex "Mayak", and in the Yenisei river basin — the area affected by the Krasnoyarsk mining and chemical industrial complex.
2. MODEL DESCRIPTION

The model being developed comprises the following main blocks:

(a) Modelling of ecological processes: the dynamics equations for species biomasses and ecological groups of species, with consideration of temperature, biogenic elements, illumination and other factors.

(b) Modelling of radionuclide migration in the ecosystem: the dynamics equations for radionuclide concentration in organisms, water, bottom sediments, floodplain soil, etc.

(c) Dose and radiation risk assessments.

A distinguishing feature of the model is the possibility of a complex description of ecological and radioecological processes, radioactive contamination and non-radiation factors. The model has been applied to the diagnostics and the prediction of non-equilibrium processes of radioactive contamination under normal operating conditions of nuclear reactors and in the case of accidental contamination of water bodies. The model has also been used to describe the joint impact on ecosystems of a combination of technogenic factors, such as thermal contamination, biogenic load (eutrophication) and ionizing radiation.

3. MAIN RESULTS

Table I presents the results of assessments of the radiation risk from water use for various water bodies in Russia affected by nuclear power engineering.

<table>
<thead>
<tr>
<th>Aquatic object</th>
<th>Annual risk</th>
<th>Main sources of risk (radioisotopes)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cooling pond of the Leningrad NPP (normal operating conditions, 1973-1985)</td>
<td>$10^{-8}$ to $10^{-7}$</td>
<td>Consumption of fish ($^{60}$Co, $^{65}$Zn and others)</td>
</tr>
<tr>
<td>Yenisei river, in the area affected by the Krasnoyarsk mining and chemical industrial complex (1991)</td>
<td>$10^{-5}$ to $10^{-4}$</td>
<td>Consumption of fish ($^{32}$P, $^{24}$Na)</td>
</tr>
<tr>
<td>Lakes in the Bryansk region, in the territory affected by the Chernobyl accident (1991-1993)</td>
<td>$10^{-4}$ to $10^{-3}$</td>
<td>Consumption of fish ($^{137}$Cs)</td>
</tr>
<tr>
<td>Techa river, in the area affected by waste disposal from the &quot;Mayak&quot; industrial complex (1990-1993)</td>
<td>$10^{-4}$ to $10^{-3}$</td>
<td>Floodplain contamination, consumption of fish</td>
</tr>
</tbody>
</table>
In our model calculations we have taken into account multiple pathways and sources of radiation risk: consumption of fish, crustacea, waterfowl and drinking water, as well as the agricultural use of floodplains and water, external exposure, etc. The radiation risk of carcinogenic effects has been calculated using the recommendations of Publication 60 of the International Commission on Radiological Protection. For most water bodies the main source of radiation exposure was consumption of fish. For the river Techa the possibility of using the contaminated floodplain for economic purposes must also be taken into account. For verification of the model parameters we have used data on radioactive contamination of water bodies.

4. CONCLUSIONS

The model for radiation risk assessment of water use takes into account multiple pathways and sources of radioactive contamination of aquatic ecosystems. Under normal operating conditions of nuclear power plants the levels of radiation risk from water use have been shown to be considerably lower than those from natural background radiation. The background level of radiation can be exceeded when water is used from water bodies contaminated as a result of radiation accidents or considerable chronic disposal of radionuclides.

IAEA-SM-339/36P

ASSESSMENT OF AIRBORNE SHORT TERM DISCHARGES FROM LICENSED NUCLEAR SITES

Ministry of Agriculture, Fisheries and Food, London, United Kingdom

1. INTRODUCTION

The Ministry of Agriculture, Fisheries and Food (MAFF) together with Her Majesty's Inspectorate of Pollution (HMIP) is responsible for regulating discharges of radioactive waste from nuclear sites in the United Kingdom. The role of MAFF in this area is to ensure that radiation doses to the public are kept within acceptable bounds, in particular doses arising through the food-chain [1].
Licensed nuclear sites in the UK are not permitted to discharge radioactive waste without an authorization granted by MAFF and HMIP. Authorizations impose limits and conditions on gaseous and liquid effluents. When considering an application for an authorization to discharge radioactive effluents, a detailed assessment is carried out to ensure that potential doses to the public meet nationally and internationally agreed dose criteria.

Limits are normally imposed on the amount of effluent discharged annually and, where appropriate, may also be set to control the amount of material which may be discharged over a much shorter period of time (short term limits).

The methodology used for the assessment of the food-chain implications of short, intermittent releases of radioactivity to the atmosphere is outlined. MAFF scientists perform such assessments when setting short term discharge limits for licensed nuclear sites such as gas cooled reactors.

2. REGULATORY CRITERIA

Short term limits enhance the control regime. They are particularly important for batch processes, such as isotope production, and also for gas cooled nuclear power plants which, in the course of normal operation, may discharge radionuclides, such as $^{14}$C, $^{35}$S and $^3$H, at levels which fluctuate considerably with time. The release of coolant gas on purging and blowdown of gas cooled reactors is a good example of such short, intermittent releases.

The release of radioactive material to the atmosphere over a short period has the potential (depending on the circumstances and timing of the release) to result in higher levels of radionuclides in food than the release of the same amount of material over a much longer time span. The impact of short term 'spike' discharges is assessed against two criteria: (i) the dose to the critical group, which is necessary for comparison with dose constraints [2, 3], and (ii) the peak concentration of radionuclides in foodstuffs. For some radionuclides there are limits set by the European Commission [4] and, although these are only strictly applicable to radioactivity in food after an accident, MAFF does not permit these limits to be exceeded during routine operation.

3. ASSESSMENT OF SHORT TERM DISCHARGES

The philosophy adopted when assessing the implication of short term discharges is that modelling assumptions should be cautious, to avoid underestimation, but realistic, to avoid gross overestimation. For example, for $^{14}$C discharges from gas cooled nuclear power reactors a 12 hour duration for the total release is considered to be sufficiently conservative for assessment purposes.
Weather conditions play an important role in dispersal, and a Gaussian plume dispersion model [5] (R91-STAR) is used to calculate deposition in food producing areas, such as dairy farms, orchards and gardens. The wind direction is assumed to be constant and is taken as the one which would lead to the highest dose to the critical group. A conceivable 'worst case' scenario is 12 hours of Pasquill category D weather.

The uptake and movement of radionuclides through the environment and into the food-chain can be predicted using computer codes. One example is STAR-C14 (short term atmospheric release, $^{14}$C), which has been developed for MAFF by Intera Information Technologies. Carbon accumulation in plant and animal systems is modelled mathematically using sets of discrete compartments. Migration between compartments is controlled by transfer coefficients which represent carbon transfer processes such as photosynthesis, respiration, inhalation and excretion. These transfer parameters are based on studies of the carbon cycle in plants and animals.

Food produced in the locality is assumed to be collected when the concentration in the foodstuff is at a maximum. In the calculation of the dose to the critical group it is assumed that one year’s supply of the two food categories which give rise to the highest dose are harvested and stored and then consumed over a one-year period. However, this is not appropriate for some foods for which one year’s supply is not stored but is continually collected (e.g. eggs and milk). These are assessed as being collected at intervals throughout the year, and a dose is arrived at by allowing for loss of activity by metabolic processes and radioactive decay, as appropriate. Dietary habits are based on national survey data, and food consumption by the critical group is taken at the 97.5th percentile for the two food categories.

4. CONCLUSIONS

A methodology for assessing short term discharges from nuclear sites has been described; care has been taken to choose cautious but realistic modelling assumptions. In practice, evidence shows that the actual doses to the public are much lower than the calculated doses [6, 7]. The Ministry funds a comprehensive R&D programme; the results of this programme are used to improve its modelling of atmospheric dispersion, deposition and subsequent transfer of radioactive material through the terrestrial food-chain to man.

REFERENCES


Accumulations of radioactivity at high concentrations and with associated elevated dose rates can be found in various locations of the Ribble Estuary on the west coast of England. These result from direct discharges of radioactive liquid effluents by British Nuclear Fuels plc (BNFL) from its Springfields Works to the tidal waters of the Estuary and from marine transport of radioactivity originating from discharges of liquid effluents by BNFL from its Sellafield Works in Cumbria to the Irish Sea.

A long term study of the levels and behaviour of radioactivity in the Estuary and their radiological significance has been carried out by the University College of North Wales on behalf of Her Majesty’s Inspectorate of Pollution [1].

Discharges from BNFL’s Springfields Works arise mainly from the large scale purification of uranium from ore concentrates and its subsequent manufacture into nuclear fuel. The principal radioactive components of the aqueous wastes are the
naturally occurring alpha emitting radionuclides $^{230}$Th and $^{232}$Th (and its decay products) and the beta emitting radionuclides $^{234}$Pa$^m$ and $^{234}$Th, the short lived decay products of $^{238}$U. Protactinium-234m and $^{234}$Th account for almost all of the beta activity discharged by Springfields Works.

Beta dose rates, gamma dose rates and concentrations of radioactivity in sediments were measured at 236 locations in the Estuary over an 18 month period.

Mobile fine grained sediments in the upper reaches of the Estuary were found to have high concentrations of short lived beta emitting radionuclides, originating from discharges of liquid effluents from Springfields Works. The high concentrations were transient over short time periods, being influenced by tidal movements, river flow scouring of sediments and variations in discharges. Generally, the contaminated sediments were found not to clear the Estuary. Elevated gamma radiation dose rates were found over thick deposits of immobile mud on the outer regions of the Estuary where long lived gamma emitting radionuclides ($^{137}$Cs, Pu and Am), originating from discharges of liquid effluents from BNFL’s Sellafield Works, have accumulated.

The study also involved an investigation into the agricultural, commercial and leisure uses of the Estuary. Face to face interviews were conducted at nine locations within the Ribble Estuary in order to ascertain the recreational use made of the Estuary; 196 people were questioned. In addition, 100 clubs were sent a postal questionnaire to determine their use of the Estuary. From this information, groups with associated occupancy times were considered in the radiological assessment. This information was combined with the detailed measurements to assess the radiological significance of radioactivity in the Estuary.

The most exposed groups were found to be people occupying houseboats in muddy creeks in the outer regions of the Estuary. The annual radiation exposure there was estimated at 220 μSv. Other groups, including bird watchers, beach combers, dog walkers, wildfowlers and anglers, were estimated to receive exposures in the range of 30–75 μSv/a.

The principal pathway was found to be external exposure to gamma radiation associated with accumulations of radionuclides originating from Sellafield. Radiation exposure to short lived beta emitting radionuclides originating from Springfields was estimated to be low (less than 10 μSv/a).

REFERENCE

DEVELOPMENT OF AN ACCIDENT CONSEQUENCE ANALYSIS PROGRAM BASED ON THE OBJECT ORIENTED PROGRAMMING TECHNIQUE

Integrated Nuclear Safety Assessment,
Korea Atomic Energy Research Institute,
Taejon, Republic of Korea

The KAERI accident consequence analysis program KACAP is being developed on the basis of reusable objects in PPAM (platform for the development of plant analysis and management codes). Development of PPAM is being conducted at the Korea Atomic Energy Research Institute (KAERI) in order to be able to provide portability and reusability of computer codes, and consistent user interface in developing software with the use of object oriented programming (OOP) [1] under a Microsoft Windows environment. By constructing the platform, software development can benefit from a shorter development cycle and an easier validation and verification process.

The KACAP code can be used to analyze the transport of radioactive material through the environment. Dispersion and deposition of radionuclides released from the containment of a reactor building to the atmosphere have been considered with a straight-line Gaussian plume model in KACAP. Because of the mountainous regions around nuclear power plants in Korea, the code will be improved to take into account effects of the terrain.

The initial screen of KACAP (Fig. 1) consists of three windows: MapView, Parameters and SourceView. If a nuclear power plant is chosen simply by clicking the predesignated name on the Parameters window, the entire map of South Korea with the exact location of the plant is displayed on the MapView window. The parameters necessary for calculating the radionuclide concentration, such as wind direction, wind speed and atmospheric stability, can be selected or typed into the dialogue boxes on the Parameters window. To evaluate the radionuclide concentration, one clicks on the "Accept Input" box. Then the distribution of the radionuclide concentration is displayed on the MapView window, as shown in Fig. 2. At present, the concentration is shown on the screen for radii of 12 km and 100 km from the plant, which can be chosen on the Parameters window. The SourceView window shows the default values of the release rate for the 60 radionuclides which are important for accident consequence analyses. These values can be modified according to the specific source term data.
FIG. 1. Initial screen of the KACAP.

FIG. 2. Distribution of radioactivity around the Ulchin nuclear power plant.
A module for the calculation of radiation doses to on-site and off-site groups of people will be added to the KACAP code. Exposure pathways will include direct radiation from the passing plume and from radioactive material deposited on the ground, inhalation of airborne radionuclides and resuspended ground contamination, and ingestion of contaminated foodstuffs. Short term and long term emergency response actions will also be accounted for in the evaluation of potential radiation doses. Also, modules will be added for estimating the consequences of accidents, such as early and latent health effects, and economic impacts. By incorporating all of these modules and the appropriate statistical model into the KACAP code, it can be used as a tool for level 3 probabilistic safety analysis.

REFERENCE

On the Semipalatinsk test site, more than 50 nuclear weapons tests have been conducted. Some of them caused considerable radiation exposure of the population in the adjoining territories. From the available data it is seen that part of the population of the Altai region suffered seriously from these tests. For some settlements the radiation doses exceeded 0.5 Sv [1].

Calculated data on the radiological consequences of the tests for the population of the Altai region have been compared with data on the mortality from malignant neoplasms in different years.

A special computer risk assessment module (RAM) has been developed for assessing the radiological consequences of the tests, in the framework of a research programme [2] and using the experience made when the computer module “risk assessment” was worked out as part of the more general, integrated computer decision aiding system to be used after a nuclear accident [3].

RAM includes a database which contains arrays of medical and geographical data (MGD) on the age specific death rate from different background risk factors and for different cohorts of the population, and a complex of calculation and service codes. The calculation codes have been prepared on the basis of a special variant of the risk assessment methodology (MRA), which has been developed for the present application as well as for other applications.
The development of MRA is connected mainly with specific features of RAM applications for which it is necessary to use risk indices in their detailed dependence on local conditions, time, age, exposure modes and risk factors, including background factors, etc. In this development, relevant international and national publications have been used (publications of the International Commission on Radiological Protection, and reports of the United Nations Scientific Committee on the Effects of Atomic Radiation and of the Committee on the Biological Effects of Ionizing Radiation, e.g. BEIR V). MRA and RAM can be applied to the assessment of radiological and non-radiological risks.

The following indices are used for risk assessment:

- Individual annual or lifetime risk and detriment (man-years lost),
- Number of deaths from malignant neoplasmas due to nuclear weapons tests for different years.

REFERENCES

The fate of radionuclides in the environment is influenced by a variety of non-radiological factors, including climate, ecosystem type, land cover and local management practices. Recently, the geographical distribution and interaction of these factors has been investigated, often using systems such as Geographical Information Systems (GIS). The overall flux of radionuclides in the environment and the extent to which they are transferred to various food products depend not only on rates of transfer but also on the comparative importance of different sorts of agricultural production and the agricultural practices used. For instance, the harvesting dates of different vegetables and the months during which animals graze outdoors are important factors in determining the potential extent of radionuclide contamination of food products. Spatial variation across these variables needs to be quantified in a manner which can be used in assessment models. One approach which is being developed to integrate such information into predictive models in the United Kingdom is described.

Current models which calculate ingestion doses tend to make some generic assumptions about crops grown in particular areas. A research programme is now being undertaken with the aim of identifying and assembling relevant agricultural and land cover data to express the range of variation in foodstuff characteristics throughout the UK. Geographical information concerning land use has been combined with information on averaged feedstuff and animal feed production, so that the land area surrounding each nuclear power plant (NPP) can be described in terms of
land cover and agricultural production. A model has been developed which brings together the spatial characteristics of any specified area adjacent to an NPP with databases on agricultural feedstuffs.

At the core of the model is the Land Classification developed by the Institute of Terrestrial Ecology (ITE); this classification has used a wide range of environmental variables (such as climate, geology and topography) to allocate each of the 240,000 1 km squares in the UK to one of the 32 'environmental' land classes. The classes have been described in terms of the underlying environmental variables (for example, one class may be typical of warmer, wetter, low lying land in the southwest; another class will describe colder, drier uplands in the north-east) and also characterized according to ecological data collected in the field.

The model also makes use of the Countryside Information System (developed by ITE under contract with the Department of the Environment). This is a computer based system which describes, for user specified areas of the UK, any census or sample data which can be referenced to the 1 km grid of the UK. Using the new model, spatial areas around each NPP can be specified. In the simplest case, this might be an area which lies within a given radius of the site. The model will be developed to describe ellipses and other shapes which might mimic the 'plume' of an emission. These areas can then be described in terms of the ITE Land Class composition and thence their land cover characteristics.

The second main element of the model is to derive, for each ITE Land Class, the average parameters for a range of foodstuff variables. These variables have been chosen to represent a range of food types which may be important to the population adjacent to an NPP. The model gives, for example, estimated earliest, latest and mean sowing dates, together with the range, for a variety of crops. For animal products, additional information is available, for instance on yield and feedstuff characteristics at different times of the year.

These variables have been derived from a wide range of existing data sources and have been fitted to the ITE Land Classes according to the environmental and land cover characteristics of the class. Thus, for a crop such as wheat, a class in the southeast of the UK will have a much earlier average planting date than that for a different land class in the north. The data are held in a matrix with 32 land class values for each selected foodstuff parameter.

Using simple matrix algebra, the model then uses the foodstuff data matrix, together with the land class composition of the chosen area, to calculate average foodstuff values for that area. Examples are given in Table I. These outputs are carried forward into further models which have already been developed by Nuclear Electric plc to calculate ingestion doses following, for example, airborne releases of radioactivity.
<table>
<thead>
<tr>
<th>Nuclear power plant</th>
<th>Mean planting date for potatoes (days from 1 Jan.)</th>
<th>Mean density (t/ha) for spring cabbages on day 365</th>
<th>Mean harvesting date for winter wheat (days from 1 Jan.)</th>
<th>Mean fruit setting date for Cox apples (days from 1 Jan.)</th>
<th>Mean yield of milk (L·ha⁻¹·a⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Berkeley (south-west England)</td>
<td>103</td>
<td>8.2</td>
<td>229</td>
<td>109</td>
<td>8949</td>
</tr>
<tr>
<td>Hunterston (mid Scotland)</td>
<td>109</td>
<td>5.9</td>
<td>260</td>
<td>—</td>
<td>8201</td>
</tr>
</tbody>
</table>
There are five sites in India where nuclear power plants (NPPs) are operating. Two of them (Tarapur and Kalpakkam) are situated on the coast and the other three (Rawatbhata, Narora and Kakrapar) are situated inland. Except for the first power station at Tarapur, which has two BWR units, all other stations have PHWR type reactors. The Tarapur reactor units (TAPS 1&2) are the oldest ones; they have been operating for 25 years (from 1969) with a good operating record. The Rawatbhata (Rajasthan) units (RAPS 1&2) have been operating from 1973 and from 1981, the Kalpakkam units (MAPS 1&2) from 1983 and from 1986, the Narora units (NAPS 1&2) from 1989 and from 1992, and one unit at Kakrapar from 1992. At present, the rated capacity is 160 MW(e) each for TAPS 1&2, 100 MW(e) for RAPS 1, 200 MW(e) for RAPS 2 and 220 MW(e) each for the other units. Thus the operation period of power reactors in India is long enough to make an assessment of the radiological impact of NPPs on the environment.

Under normal operation, the low level radioactive wastes produced in the plants are diluted and dispersed in the environment after monitoring and treatment, if necessary. Gaseous wastes are discharged through a high stack and liquid wastes are diluted with condenser cooling water or coolant blowdown water (using cooling towers) and discharged to a water body (sea or reservoir). The radionuclides released through the stack can cause direct radiation exposure of the public in the downwind direction and can also deposit on the ground and vegetation and cause exposure through different pathways. The radionuclides dispersed in the aquatic environment can be deposited on the silt bed or taken up by the aquatic flora and fauna. The radiological conditions for the first ten years of operation of Tarapur are reported in Ref. [1]. The increase in the radiation exposure of the public in the environment of TAPS and RAPS up to the year 1982 has been discussed at the 1984 congress of the International Radiation Protection Association [2]. The annual increase in exposure at the fence post (1.6 km from the NPP) for the period 1980–1982 was 0.4 mSv at TAPS and 0.18 mSv at RAPS. The environmental impact of PHWR type power stations in India up to 1990 has been reviewed in a paper presented at an international seminar held in India in 1991 [3]. The annual exposure of people living in the vicinity of RAPS and MAPS has been reported as 47 μSv and 74 μSv, respectively.
Data on radioactive waste releases to the environment are included in the performance reports of TAPS, MAPS, NAPS and KAPS, which are published annually. The results of environmental monitoring at all NPP sites, carried out by the Health Physics Division of the Bhabha Atomic Research Centre, are published annually. At TAPS, the maximum activity of the annual release of liquid waste was measured in 1974, namely 31.8 TBq (gross beta); this decreased gradually to 1.7 TBq by 1984. This decrease was due to augmentation of the liquid waste treatment; the annual release of liquid waste remained at almost the same level from 1984 to 1993. Caesium-137 was the major (50%) radionuclide in TAPS discharges dispersed into coastal waters. In coastal sediments at the TAPS site boundary, $^{137}\text{Cs}$ decreased from 7.8 Bq/g in 1975 to 0.4 Bq/g in 1985 and the following years. Caesium-137 in marine organisms collected at the site boundary also decreased from 44 Bq/kg in 1975 to less than 10 Bq/kg in 1984 and the following years. Thus the improvement in radwaste management was directly reflected in the radionuclide levels in environmental samples. The other radionuclides detected in the 2-5 km region outside the station are $^{60}\text{Co}$ and $^{131}\text{I}$. With improved fuel performance and after derating of the operating power level of the station, the stack release from TAPS decreased significantly (from 290 TBq/d in 1976 to 55 TBq/d in 1986 and to 20 TBq/d in 1987 and the following years). This resulted in a decrease in the dose to the public from fission product noble gases in the stack release. The total annual effective dose equivalent evaluated for people living in the area 5 km from the TAPS site is at present about 25 $\mu$Sv.

In the environment of PHWR type stations, $^3\text{H}$ and $^{41}\text{Ar}$ are the main radionuclides detected. The doses from the early PHWRs (RAPS and MAPS) have been evaluated. For later models of PHWRs (NAPs and KAPS) with design modifications, there is very little release of $^{41}\text{Ar}$ and almost no population exposure due to this. Unlike the BWR at TAPS, the PHWRs do not have any significant release of fission products in the liquid waste.

The increase in the radiation dose due to power station operation has been found to be negligible at all five sites, indicating that the impact on the radiological condition of the environment is insignificant.

REFERENCES


Many artificial radionuclides can contribute to the dose from external radiation. Among the fallout radionuclides, $^{137}$Cs is the most important contributor to the external radiation dose.

The purpose of the present work was to obtain results of $^{137}$Cs measurements of marine samples and to assess the doses to the Brazilian population from $^{137}$Cs radioactivity in marine food.

As part of our monitoring programme, marine samples (sea water and fish) from eight stations on the Brazilian coast, from Rio Grande do Sul to Pará, have been collected. About 100 L of sea water and 4 kg of fish from species most commonly consumed by the local population have been analysed monthly, as described in Ref. [1].

The levels of $^{137}$Cs in seawater samples ranged from 0.5 to 2.2 Bq/m$^3$, and those in fish varied from 0.01 to 0.39 Bq/kg (see Table I). The error of the analysis was 50% and 40% for sea water and fish, respectively.

The data given in Table I have been used to calculate the dose received by the population from the consumption of fish. The doses have been calculated using the formula (see Ref. [2])

$$D_{Cs} (fish) = C_p \ DCF \ I_r \ g_f$$
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Rio Grande do Sul</td>
<td>32°11' S</td>
<td>52°02' W</td>
<td>0.11-0.19</td>
<td>0.014-0.028</td>
<td>0.7-1.4</td>
<td>0.6-1.1</td>
</tr>
<tr>
<td>Paraná</td>
<td>25°37' S</td>
<td>48°16' W</td>
<td>0.10-0.18</td>
<td>0.035-0.048</td>
<td>0.9-1.7</td>
<td>0.9-1.6</td>
</tr>
<tr>
<td>São Paulo</td>
<td>23°50' S</td>
<td>45°25' W</td>
<td>0.10-0.30</td>
<td>—</td>
<td>0.9-1.5</td>
<td>—</td>
</tr>
<tr>
<td>Rio de Janeiro</td>
<td>22°57' S</td>
<td>43°55' W</td>
<td>0.15</td>
<td>0.022-0.053</td>
<td>0.7-1.3</td>
<td>1.1-1.4</td>
</tr>
<tr>
<td>Bahia</td>
<td>12°57' S</td>
<td>38°32' W</td>
<td>0.12</td>
<td>—</td>
<td>0.7-1.9</td>
<td>1.3-1.9</td>
</tr>
<tr>
<td>Pernambuco</td>
<td>08°02' S</td>
<td>34°52' W</td>
<td>0.22-0.39</td>
<td>0.063-0.22</td>
<td>0.7-2.0</td>
<td>1.2-1.7</td>
</tr>
<tr>
<td>Ceará</td>
<td>03°42' S</td>
<td>38°29' W</td>
<td>0.10</td>
<td>—</td>
<td>1.1-2.2</td>
<td>1.2-1.6</td>
</tr>
<tr>
<td>Pará</td>
<td>00°26' S</td>
<td>47°49' W</td>
<td>0.01-0.21</td>
<td>0.021-0.076</td>
<td>0.5-1.5</td>
<td>0.8-1.4</td>
</tr>
</tbody>
</table>
where $D_{Cs}$ is the annual committed effective dose equivalent from $^{137}\text{Cs}$ due to consumption of fish (Sv/a), $C_p$ is the concentration of $^{137}\text{Cs}$ in the fish sample (Bq/kg), DCF is the dose conversion factor for exposure to $^{137}\text{Cs}$ from ingestion (Sv/Bq), $I_f$ is the consumption rate of fish and $g_f$ is the fraction of the consumed fish arising from a contaminated source. The fish catch in Brazilian areas has been calculated, using FAO statistics, as 5.8 kg/a. On the basis of the radioactivity levels of $^{137}\text{Cs}$ presented in Table I and taking into consideration the DCF recommended by the International Commission on Radiological Protection (ICRP) [3], $1.4 \times 10^{-8}$ Sv/Bq, and a default value of $g_f = 1$, it has been possible to estimate the value of $D_{Cs}$. The results obtained are presented in Table II. The data vary from 0.8 to 31.7 nSv/a. These values are very low compared to the value recommended by the ICRP for the limit of the annual dose to members of the public (1 mSv/a).

Our results show that the Brazilian coast is not free of artificial radioactivity; this contamination is due to fallout, and the radionuclides released from different radioactive sources in the Northern hemisphere can reach the stratosphere and the troposphere, and is distributed over the whole globe.

It has been estimated that the global mean individual dose from $^{137}\text{Cs}$ in seafood is 0.03 μSv/a and the doses from $^{137}\text{Cs}$ show a significant geographical variation [4]. The highest doses (one order of magnitude higher than the global mean dose) have been received by populations eating fish from the north-east Atlantic Ocean, which has received most of the $^{137}\text{Cs}$ from Sellafield and Chernobyl. Our results are in agreement with this value, considering that the radioactivity in Brazil is due to fallout deposition.

### Table II. Estimated Doses from Consumption of Fish (1991-1993)

<table>
<thead>
<tr>
<th>Sampling site</th>
<th>Latitude</th>
<th>Longitude</th>
<th>1991</th>
<th>1993</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rio Grande do Sul</td>
<td>32°11' S</td>
<td>52°02' W</td>
<td>8.9-15.4</td>
<td>1.1-2.3</td>
</tr>
<tr>
<td>Paraná</td>
<td>25°37' S</td>
<td>48°16' W</td>
<td>8.1-14.6</td>
<td>2.8-3.9</td>
</tr>
<tr>
<td>São Paulo</td>
<td>23°50' S</td>
<td>45°25' W</td>
<td>8.1-24.4</td>
<td>—</td>
</tr>
<tr>
<td>Rio de Janeiro</td>
<td>22°57' S</td>
<td>43°55' W</td>
<td>12.2</td>
<td>1.8-4.3</td>
</tr>
<tr>
<td>Bahia</td>
<td>12°57' S</td>
<td>38°32' W</td>
<td>9.7</td>
<td>—</td>
</tr>
<tr>
<td>Pernambuco</td>
<td>08°02' S</td>
<td>34°52' W</td>
<td>17.8-31.7</td>
<td>5.1-17.8</td>
</tr>
<tr>
<td>Ceará</td>
<td>03°42' S</td>
<td>38°29' W</td>
<td>8.12</td>
<td>—</td>
</tr>
<tr>
<td>Pará</td>
<td>00°26' S</td>
<td>47°49' W</td>
<td>0.8-17.0</td>
<td>6.2</td>
</tr>
</tbody>
</table>
This work is part of a research programme which aims at studying radionuclide transfer through ecosystems and developing radioecological assessment models.

REFERENCES


The amount of plutonium excreted with urine has been measured for twenty Lithuanian clean-up workers in Chernobyl in order to calculate the unknown, past, internal plutonium contamination. The Lithuanian civilians were forced in 1986 or 1987 to do clean-up work in Chernobyl for 25 to 180 days. According to the model of the International Commission on Radiological Protection [1], the plutonium residence time in human bones is approximately 50 years, which makes it a useful isotope for retrospective assessments. All urine samples showed low plutonium concentration levels after alpha spectrometry, and the more sensitive detection method fission track analysis (FTA) was used for evaluation. FTA includes neutron activation of $^{239}$Pu in the sample, which was previously thoroughly decontaminated to remove uranium. The fission fragments created will form tracks in the quartz glass slide on which the sample has been deposited. After etching, the tracks are counted, manually or automatically. Figure 1 shows the characteristic features of a fission track in glass. Through optimization of FTA, a lower detection limit, equal to 2 μBq $^{239}$Pu, has been achieved [2].

Cytogenetic analyses, performed at the Chernobyl Medical Centre in Vilnius, have indicated increased levels of chromosome aberrations, which are generally considered to be indicators of radiation overexposure [3]. These observations, together
FIG. 1. Cone shaped tracks from fission fragments (for example, from neutron activation of $^{239}$Pu) in glass. The size of the tracks is approximately 1 μm in diameter.

with the large number of lung cancer cases among these clean-up workers, may indicate a high, local exposure of the lungs to plutonium or hot particles of beta or gamma emitters. The results of the chromosome aberration study of peripheral lymphocyte cells show 2.5–7.5% of aberrant cells.

In addition, every urine sample was analysed for $^{90}$Sr and $^{137}$Cs. Strontium remains in the body for many years, while caesium is excreted within several hundred days.

REFERENCES


In a large scale investigation of Polish forest litter contamination performed in recent years [1-3] we observed the variety of Chernobyl fallout in Poland. Besides large differences in the radiocaesium contamination levels, we found a non-uniform isotopic composition of the fallout; in the north-eastern part of the country we found enhanced (relative to caesium) amounts of non-volatile radioisotopes. We were still able to detect $^{144}$Ce, $^{154}$Eu and $^{155}$Eu [2] as well as plutonium isotopes of Chernobyl origin [3]. Since they exhibited activity ratios close to those of the Chernobyl reactor core inventory [4, 5] and seemed to be distributed evenly throughout the samples, our investigations support a model of precipitation of small fuel-like hot particles from the Chernobyl cloud in north-eastern Poland. All of the observed long lived non-volatile radioisotopes were accompanied by other non-volatile radioisotopes with shorter half-lives. Assuming core inventory activity ratios, we were able to reconstruct the initial fallout activity of more than ten non-volatile isotopes with half-lives of more than one week.

The obtained data on fallout were used as input into a simple model that can be used to calculate the inhalation dose from fallout. The resulting external effective dose equivalent (EDE) was also calculated for this case. A similar attempt was made for another case — that of the region with the highest radiocaesium contamination, which is situated in south-western Poland. The external dose in the first year was higher in the region with precipitation of fuel-like hot particles than in south-western Poland. The obtained EDE values were compared with the fifty-year Chernobyl EDE estimated for an average Pole (Ref. [6], based on observations in Warsaw of the isotopic composition of the Chernobyl cloud and an average caesium level) and with the EDE due to mushroom consumption in Poland [7].

Our reconstruction of the inhalation EDE gives an additional value (28-100 μSv), which is comparable with the average inhalation EDE in Poland (45 μSv). However, the value obtained by us for the external fifty-year EDE for people living in the region with the highest contamination is up to ten times higher than the upper limit of the Chernobyl EDE in Poland estimated previously (ca. 1 mSv [6]). The consumption of mushrooms can also significantly increase the EDE value. The number of people who might be affected by maximum EDE values is very limited.
REFERENCES


IAEA-SM-339/75P

APPLICATION OF THE CLRP MODEL TO THE ASSESSMENT OF THE THYROID CONTENT AND THE COMMITTED DOSE EQUIVALENT OF THE POLISH PEOPLE FROM RELEASE OF $^{131}$I TO THE ENVIRONMENT

Verification of model predictions on the basis of Chernobyl data

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The CLRP model was applied to the assessment of the $^{131}$I content in the thyroid for various age and sex groups of the Polish population, considering protective measures that had been introduced after the Chernobyl accident [1-3]. The CLRP model permits simulation of many different radiological situations (chronic or acute releases) and measures to reduce the internal dose, including restrictions on
the human diet, change of the feeding regime for dairy cows and, later on, administration of stable iodine. The CLRP model has been designed as a set of Excel worksheets that simulate the transport of radionuclides through agricultural ecosystems to humans. All dynamic processes are described by exponential formulas and are solved numerically.

The CLRP model is deterministic and yields single estimates of specified variables, but it is also possible to run the model with Monte Carlo subroutine, which enables an uncertainty analysis to be performed. Radioiodine concentrations in particular components of the terrestrial ecosystem, e.g. soil, vegetation, animal tissues and animal products, are calculated as a function of the time after deposition from the atmosphere. From these data, the radionuclide content in a specific organ as a function of time is calculated and the committed dose equivalent from internal contamination is estimated as an integral of the resultant dose rate over a sufficient period of time. For radioiodine, the five-compartment model of the metabolism of iodine, developed by Johnson [4], has been used. In addition, the model allows to estimate the inhalation dose from the time integrated air concentration, and the external dose from total deposition, using reduction factors for different types of shielding conditions. Dynamic processes considered in the model include foliar interception, weathering, plant growth and root uptake, leaching and radioactive decay. The model takes into account seasonal changes in the vegetation biomass and in animal diets, as well as specific ploughing and crop harvest dates. Human dietary data are included to permit calculation of time dependent radionuclide ingestion rates for adults and for 10 year, 5 year and 1 year old children.

The justification of the model performance for $^{131}$I has been done on the basis of the post-Chernobyl data for Poland. These data included: the air concentration of $^{131}$I measured in Warsaw by air sampling stations (also determination of the physico-chemical forms of iodine), the concentration of $^{131}$I in milk for the whole region, and about 1500 measurements of the $^{131}$I contents in the thyroid for representative groups of the Polish population. Since prophylactic doses of stable iodine had been distributed in 1986 to infants, 1–10 year old children and up to 16 year old teenagers, the reduction in thyroid uptake had to be taken into account by model calculations. Additionally, diet restrictions and limitations on grazing of cows were introduced in the model calculations. On the basis of model predictions, the effectiveness of the administration of stable iodine regarding the reduction of the $^{131}$I thyroid doses was assessed. The obtained $^{131}$I thyroid contents were compared with results of measurements of $^{131}$I in the thyroids of Polish people.

Figure 1 shows a comparison of the $^{131}$I thyroid contents predicted by the CLRP model with measurement data for different variants of thyroid blocking measures for people living in Warsaw. Analysis of the measurement data showed that the effect of a single dose of stable iodine was insignificant. It should be noted that the high concentration of $^{131}$I in air was relatively short compared with the concentration of $^{131}$I in milk and other food products. The uptake of radioiodine by the
FIG. 1. Comparison of predicted and measured data for adults who lived in a moderately contaminated area, (a) for persons who received no stable iodine and (b) for persons who received 60 mg of stable iodine on 29 April 1986.
thyroid was effectively blocked only during a short period of time and the thyroid dose due to further intake of $^{131}$I was not reduced. It is concluded that in the case of prolonged and widespread contamination with $^{131}$I, dietary restrictions seem to be more effective than administration of a single dose of stable iodine.

REFERENCES


Historical background. In 1956, a licence for the discharge of liquid radioactive waste was issued to the Nuclear Energy Research Centre in Mol. The liquid waste comes from the radioactive waste treatment installations and is discharged through an underground pipeline to the river Molse Nete. The licence was transferred to BELGOPROCESS in 1989.

This licence, which was revised in 1986, contains a limit for the total activity. This total activity is determined by using a weighting formula (as recommended by the Benelux Committee on Public Health [1]) which is based on the maximum allowable concentrations in drinking water:

$$7.5 \, ^{90}\text{Sr} + 0.001 \, ^{3}\text{He} + 3 \, ^{131}\text{I} + \text{total } \beta + 300 \, ^{226}\text{Ra} + 5 \, \text{total } \alpha$$

The total weighted activity discharged is limited to 166 GBq/month; the radioactivity concentration is limited to 37 MBq/m$^3$.

Given a water flow of 1 m$^3$/s at the point of discharge, a maximum discharge of 166 GBq/month would lead to an activity concentration of 0.064 MBq/m$^3$ in the river water, to be compared with a maximum allowable concentration in drinking water of 1.11 MBq/m$^3$ for occupationally exposed workers and ten times lower for the general public.
It is pointed out that in the determination of these discharge limits, only the drinking water pathway was taken into consideration. Other exposure pathways, e.g. via sedimentation and external exposure, were not considered.

In the framework of an IAEA co-ordinated research programme on the role of sediments in transport and accumulation of radionuclides in waterways, a study was carried out in the early 1980s to model the radionuclide distribution between the aqueous and particulate phases of the river water compartment and the transfer of radionuclides from the water compartment to the bed sediment [2]. External irradiation by sediment deposits appeared to be the most important exposure pathway.

In 1991, a more detailed assessment was carried out by the National Radiological Protection Board (NRPB, United Kingdom) [3] on behalf of BELGOPROCESS, confirming the earlier findings. Given the results of this assessment and the publication of the 1990 Recommendations of the International Commission on Radiological Protection (ICRP) [4], the Radiological Protection Office of the Ministry of Public Health and the Environment requested a review of the discharge limits and the underlying weighting formula.

**Dose criteria.** The starting point is the dose limit for members of the public recommended by the ICRP in Publication 60 [4]. To take account of the fact that individuals may be exposed to more than one source, a dose constraint of 0.33 mSv per year has been adopted for the derivation of the discharge limit.

**Mathematical model.** To derive the discharge limits, the mathematical model from the impact assessment by the NRPB [3] has been used. The following assumptions have been made:

- The source term is constant (Bq/a); the radiological impact (mSv/a) reaches, after a number of years, an equilibrium which depends on the exposure pathway; this equilibrium value is used in the derivation.
- The radionuclide concentration on river banks equals the concentration in the river bed sediment, i.e. the same accumulation and the same maximum value are assumed to occur at the same distance from the discharge point.
- Irrigation with unfiltered water, dredging and evacuation of aquatic plants has no effect on the radionuclide inventory in the river compartment.
- Every five years, the sediments are spread out over pastures (1.5 mm) and agricultural land (3 mm); the model converts these quantities into yearly quantities.
- Every five years, aquatic plants are removed from the river and used as fertilizer; the model also converts this quantity into a yearly quantity.

Calculations of doses to adults and children, through 52 different exposure pathways, have been performed for the different compartments of the river; the highest values appear to be reached in the compartment situated 4.2-6.8 km from the discharge point. These values have been used to derive the discharge limits. It also appears that the doses to adults are always higher than the doses to children.
Critical groups. Four critical groups have been identified, considering actual habits and land use:

- Group No. 1 consists of fishermen staying, or people walking, on the river bank for 250 h per year and eating 5 kg of fish per year.
- Group No. 2 consists of people who have the same habits and who, in addition, obtain 10% of their foodstuffs locally, i.e. they are potentially contaminated (directly, or indirectly from animal products) through the use of river water for irrigation and the use of riverbed sediment and aquatic plants as fertilizer.
- Group No. 3 consists of individuals staying, for 300 h per year, on grounds on which river-bed sediment has been deposited and obtaining 10% of their foodstuffs from such grounds.
- Group No. 4 consists of individuals with the same habits as those in group No. 3, but who obtain 10% of their foodstuffs locally, i.e. they are potentially contaminated through all possible pathways.

Doses to the critical groups. Doses have been calculated for the relevant radionuclides, taking the discharges in 1989 as reference for the isotopic composition. Individuals belonging to group No. 2 appear to receive the highest annual dose. The contribution from the food-chain is very small in comparison with the dose due to external exposure (essentially from $^{60}$Co, $^{134}$Cs and $^{137}$Cs).

Analysis of the calculated doses allows to determine the relative importance, per unit of activity released, of the contribution from each radionuclide with respect to the contribution from $^{60}$Co. Hence, a weighting factor for calculating a weighted activity can also be determined. The radionuclides are then grouped and a weighting factor is assigned to each group of radionuclides, as indicated in the following list.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Relative importance (%)</th>
<th>Weighting factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>H-3</td>
<td>0.0024</td>
<td>0.00002</td>
</tr>
<tr>
<td>Cr-51</td>
<td>0.13</td>
<td>0.1</td>
</tr>
<tr>
<td>Mn-54</td>
<td>17</td>
<td>0.1</td>
</tr>
<tr>
<td>Co-58</td>
<td>2.6</td>
<td>0.1</td>
</tr>
<tr>
<td>Co-60</td>
<td>100</td>
<td>1</td>
</tr>
<tr>
<td>Sr-90</td>
<td>42</td>
<td>0.4</td>
</tr>
<tr>
<td>Ru-106</td>
<td>1.4</td>
<td>0.1</td>
</tr>
<tr>
<td>Sb-124</td>
<td>1.1</td>
<td>0.1</td>
</tr>
<tr>
<td>Sb-125</td>
<td>5.2</td>
<td>0.1</td>
</tr>
<tr>
<td>I-131</td>
<td>4.4</td>
<td>0.1</td>
</tr>
<tr>
<td>Cs-134</td>
<td>180</td>
<td>1.5</td>
</tr>
<tr>
<td>Cs-137</td>
<td>160</td>
<td>1.5</td>
</tr>
</tbody>
</table>
This analysis has led to the following revised weighting formula, to be used in the discharge licence:

$$0.00002 \, ^3\text{He} + ^{60}\text{Co} + 1.5 \, ^{137}\text{Cs} + 0.4 \, ^{90}\text{Sr} + 0.1 \, \beta + 2.5 \, \text{total} \alpha$$

where $\beta = \text{total} \, \beta - ^{137}\text{Cs} - ^{60}\text{Co} - ^{90}\text{Sr} - ^3\text{H}$. The use of this formula in calculating the doses received leads to values which are slightly higher than the doses calculated with the exact 'dose factors'. However, the use of the weighting formula makes it easier to control compliance with the discharge limits.

**Discharge limits.** Using the weighting formula, a weighted 'dose factor' of $10^{-12} \, \text{mSv} \cdot \text{a}^{-1} \cdot \text{Bq}^{-1}$ can easily be derived. Hence, the dose constraint of 0.33 mSv/a corresponds to a discharge of a weighted activity of 330 GBq/a. The current discharges amount to 4–5% of this value, corresponding to an annual dose of 0.015 mSv for individuals of the critical group. This newly proposed limit corresponds to about 30% of the actual discharge limit. Also, a limit of 40 GBq/month has been proposed.

Finally, to take account of the changes in the weighting formula, a concentration limit of 20 MBq/m$^3$ has been proposed (which corresponds to the actual limit of 37 MBq/m$^3$).

**REFERENCES**

[1] BENELUX COMMITTEE ON PUBLIC HEALTH, Maatregelen ter bescherming van de gezondheid van de mens in verband met de toepassing van kernenergie voor vredzame toepassingen — Aanbevelingen van de bijzondere Beneluxcommissie voor de volksgezondheid (1961).

DOSE IMPACT FROM ROUTINE DISCHARGES OF THE SCK·CEN NUCLEAR RESEARCH CENTRE

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The SCK·CEN Nuclear Research Centre in Mol comprises several test reactors and nuclear laboratories. Gaseous radioactive effluents are being discharged into the atmosphere through stacks. Liquid radioactive effluents are being discharged into the Molse Nete, a small river with a flow rate of the order of $1\text{ m}^3/\text{s}$, through a 9 km long pipeline.

Table I gives the total activities discharged annually during the period 1986–1993 (1986–1990 for liquid discharges) and the contributions of the most important radionuclides. Concerning the noble gas activities discharged into the atmosphere, only maximum values can be given because the activities lie below the minimum measurable limits.

In both the gaseous effluents and the liquid effluents, tritium is shown to be the radioisotope with the highest discharge rate, as expected. Concerning the $\beta \gamma$ emitters, the contributions of $^{60}\text{Co}$, $^{137}\text{Cs}$, $^{134}\text{Cs}$ and $^{131}\text{I}$ are among the higher ones for both types of effluent.

On the basis of these annual discharge rates, the maximum individual effective doses after 15 years of discharges (half of the normal life of a nuclear reactor) are calculated. The dose calculations are performed by applying site specific data and exposure scenarios, where available. The site specific data are principally related to transfer parameters in the biosphere and dietary habits of the population. Biospheric parameter values are derived from experiments or measurements at the site (such as in Ref. [3]) or from experimental data reported in the literature. Experimental values obtained at sites with specified characteristics have been collected in a database [4].
TABLE I. ANNUAL DISCHARGES FROM SCK·CEN

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Annual atmospheric discharges [1] (kBq/a)</th>
<th>Annual liquid discharges [2] (kBq/a)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\beta\gamma$ total(^a)</td>
<td>$1.17 \times 10^4$</td>
<td>$8.80 \times 10^7$</td>
</tr>
<tr>
<td>Co-60</td>
<td>981</td>
<td>$2.77 \times 10^7$</td>
</tr>
<tr>
<td>Co-58</td>
<td>Not measured</td>
<td>$2.71 \times 10^7$</td>
</tr>
<tr>
<td>Cs-137</td>
<td>$1.37 \times 10^3$</td>
<td>$1.02 \times 10^7$</td>
</tr>
<tr>
<td>Cs-134</td>
<td>547</td>
<td>$1.09 \times 10^6$</td>
</tr>
<tr>
<td>Sr-90</td>
<td>Not measured</td>
<td>$4.75 \times 10^6$</td>
</tr>
<tr>
<td>I-131</td>
<td>$8.67 \times 10^3$</td>
<td>$1.24 \times 10^6$</td>
</tr>
<tr>
<td>$\alpha$ total</td>
<td>39.1</td>
<td>$2.41 \times 10^5$</td>
</tr>
<tr>
<td>Pu-239</td>
<td>Not measured</td>
<td>$1.26 \times 10^5$</td>
</tr>
<tr>
<td>Am-241</td>
<td>Not measured</td>
<td>$6.72 \times 10^4$</td>
</tr>
<tr>
<td>Tritium</td>
<td>$4.03 \times 10^9$</td>
<td>$1.28 \times 10^{10}$</td>
</tr>
<tr>
<td>Noble gases</td>
<td>$&lt;2.2 \times 10^9$</td>
<td>—</td>
</tr>
</tbody>
</table>

\(^a\) Without tritium or noble gases.

The Faculty of Agronomy of the University of Leuven has carried out a study on the characteristics of irrigation practices in the region of SCK·CEN (northern part of Belgium), the results of which have also been included in the database. Data on dietary habits have been taken from publications of the National Institute of Statistics and are also recorded in the database.

Regarding exposure scenarios, not only conventional exposure pathways, which are generally considered in most impact assessment studies, are taken into account but also specific scenarios, which have been observed in the Molse Nete (for the liquid discharge impact), are allowed for. They are related to the application of river sediments or aquatic plants onto agricultural soil.

Table II presents the maximum individual doses after 15 years of routine atmospheric discharges into the Molse Nete and into the atmosphere, and the doses due to liquid discharges for three types of exposure pathway.

As far as atmospheric discharges are concerned and not considering noble gases, tritium is by far the most critical nuclide; the critical exposure pathway is ingestion of grains, followed by inhalation of tritium from the plume.

However, the total individual doses from liquid discharges are much higher than those from atmospheric discharges; cultivators living at the Molse Nete near the discharge point of the pipeline constitute the critical group regarding the impact from
TABLE II. ANNUAL EFFECTIVE INDIVIDUAL DOSES (μSv/a)

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Atmospheric discharges</th>
<th>Liquid discharges</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Conventional pathways</td>
<td>Application of sediment</td>
</tr>
<tr>
<td>Co-60</td>
<td>2.4 x 10^{-4}</td>
<td>31.1</td>
</tr>
<tr>
<td>Co-58</td>
<td>—</td>
<td>1.9</td>
</tr>
<tr>
<td>Cs-137</td>
<td>5.5 x 10^{-4}</td>
<td>26.6</td>
</tr>
<tr>
<td>Cs-134</td>
<td>1.2 x 10^{-4}</td>
<td>4.2</td>
</tr>
<tr>
<td>Sr-90</td>
<td>—</td>
<td>2.8</td>
</tr>
<tr>
<td>I-131</td>
<td>5.8 x 10^{-4}</td>
<td>0.5</td>
</tr>
<tr>
<td>$\beta_y$ total</td>
<td>1.5 x 10^{-3}</td>
<td>68</td>
</tr>
<tr>
<td>$\alpha$ (Pu-239)</td>
<td>$1.3 \times 10^{-3}$ b</td>
<td>0.5</td>
</tr>
<tr>
<td>Tritium</td>
<td>$2.85 \times 10^{-2}$</td>
<td>3.3</td>
</tr>
<tr>
<td>Noble gases</td>
<td>$&lt;3.7 \times 10^{-2}$ c</td>
<td></td>
</tr>
<tr>
<td>Total</td>
<td>$3.1 \times 10^{-2}$ d</td>
<td>71</td>
</tr>
</tbody>
</table>

* Negligible.

b Assuming all $\alpha$ emitters = $^{239}$Pu for the atmospheric discharges.

c Corresponding to the minimum measurable activity and the typical isotopic composition.

d Without noble gases.

Discharges of SCK-CEN. The application of sediments from the Molse Nete onto agricultural land results in doses that are higher than those from the conventional exposure pathways. It should be noted that the doses from the three exposure scenarios may not be added together, since the application of sediments and that of aquatic plants exclude each other and will also influence the effective decay constants of soil contamination due to irrigation.

The critical nuclide for all three classes of exposure pathway is $^{60}$Co; external exposure from fields treated with river sediments is the critical exposure pathway.

REFERENCES

Mathematical models are used extensively for evaluating the environmental impact of releases of radionuclides from nuclear power plants, nuclear waste disposal facilities, uranium ore mining, etc. What can be studied by means of mathematical methods is not reality, but abstract and simplified ideas of the real situation. Thus, we study the behaviour of mathematical models, which only approximate real world conditions, and all predictions will be associated with various degrees of uncertainty, which in turn depend on the underlying understanding of ongoing processes.

The representation of reality in a simplified form for modelling purposes is by no means easy and the best we can do is to try to specify all relevant processes we can identify. However, the conceptual uncertainties remain, and additional uncertainty will be linked to deficiencies in (i) the mathematical description of relevant processes, (ii) the uncertainty in the model’s input parameters, and (iii) the evolution of the system under investigation.

Opportunities to test predictions against observations should be exploited whenever actual releases occur, but, because such opportunities arise infrequently, predictions from several models applied to a common scenario should be compared and reasons for discrepancies investigated and explained.
BIOMOVS-II (BIOspheric MOdel Validation Study) is an international co-operative effort to test biospheric models designed to calculate the environmental transfer and bioaccumulation of radionuclides and other trace substances. The study uses a variety of methods to determine the level of confidence that can be placed in the predictions of environmental transfer models.

The first phase of the BIOMOVS study was officially launched at a meeting in Paris in 1986 and was finished in Stockholm in 1990. At the Stockholm meeting a strong wish was expressed for a continuation of the study.

In the second phase of BIOMOVS a number of issues are investigated that have been identified in the first phase of the study and which are believed to be of special importance in environmental transfer modelling. Emphasis is being placed on scenario development and model evaluation, uncertainties in the predictions of assessment models and mathematical aspects of assessment models.

The results may be used in conjunction with findings from other model testing studies, e.g. the IAEA-CEC co-ordinated research programme VAMP (Validation of Environmental Model Predictions), to enhance the reliability of model predictions.

BIOMOVS-II is managed through a steering committee composed of members of the financing organizations: Atomic Energy Control Board (AECB) of Canada, Atomic Energy Agency of Canada Limited (AECL); Empresa Nacional de Residuos Radiactivos SA (ENRESA), Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT) and Instituto PRYMA, in Spain; and the Swedish Radiation Protection Institute (SSI).

The primary objectives of BIOMOVS are threefold, namely:

— To test the accuracy of predictions of environmental assessment models for selected contaminants and exposure scenarios;
— To explain differences in model predictions due to structural deficiencies, invalid assumptions and/or differences in selected input data; and
— To recommend priorities for future research in order to improve the accuracy of model predictions.

A secondary objective of the study is to act as a forum for the exchange of ideas, experience and information to improve the confidence with which the environmental behaviour of trace substances in the biosphere can be assessed quantitatively.

BIOMOVS utilizes two approaches to fulful these objectives, namely:

— Approach A, in which test scenarios are formulated on the basis of suitable observational data, and model predictions are compared with these independent data sets; observations are not made known to the modellers until predictions have been submitted.
— Approach B, in which model predictions and the related uncertainty estimated for specific test scenarios are compared.
Examples of work tasks being handled within phase II of BIOMOVS are the following.

*Reference biospheres:* Development of a methodology for providing reference biospheres for long term assessments, and demonstration of the application of the methodology, initially to radionuclide releases to the biosphere in the groundwater from a deep geological disposal facility.

*Complementary studies:* Extension of those modelling scenarios considered in the first phase of BIOMOVS which were relevant to long term radioactive waste disposal assessments. The intention is to allow more detailed intercomparisons of the model results for different releases from the geosphere, and for subsequent transport, accumulation and radiological exposures.

*Uranium mill tailings:* Comparison of models used to assess the long term impact of contaminant releases from uranium mill tailings, involving multiple pathways, multiple contaminants and multiple environmental receptors.

**Uncertainties and validation**

*Model complexity:* Study of how mathematical models of varying degrees of complexity describe radionuclide transport in the biosphere. The current exercise is based on modelling radionuclide transport in soil.

*Modeller interpretation:* Study of the differences that can arise in model results owing to user interpretation of models and user interpretation of the system to be modelled.

*Natural analogues:* Extension of the understanding of the processes continuously affecting the turnover of elements in the biosphere and their accurate description by radioecological models.

*Quantitative comparisons of model predictions:* Investigation of ways of comparing model predictions with other model results and with observational data.

*Lysimeter data:* Testing of models of upward migration of radionuclides in soil using lysimeter data.

**Post-Chernobyl data**

*Wash-off scenario:* Improvement of models concerned with the movement of trace contaminants from terrestrial sources to water bodies, taking into account chemical speciation and the effects that this has on the transfer of contamination from soil to water, and the geochemical and geophysical processes affecting such transfer.

*Cooling pond scenario:* Testing of models for radioactive contamination of aquatic ecosystems, using the data for the cooling pond of the Chernobyl nuclear power plant. The dynamic processes of radionuclide migration and accumulation in closed aquatic systems are studied.
Resuspension scenario: Consideration of the atmospheric resuspension of radionuclides in Ukrainian regions impacted by Chernobyl fallout. Resuspension is an important pathway, considering the long term effects of radioactive releases to the environment.

Special radionuclides

Tritium: Testing of the biotic and soil elements of tritium assessment models. This involves a short term, e.g. accidental, release of HTO to an agricultural area at the peak of the growing season.

Carbon-14: Prediction of $^{14}$C concentrations in water, sediment and biota in a lake following contaminant release directly into the lake.

BIOMOVS-II is scheduled to last for five years and to be terminated in 1996. There is one workshop per year in which all project teams come together. The project teams meet individually twice per year. For the five years of operation, reports for all individual work tasks and specialized topics will be published. A final report, reviewing the study as a whole, discussing its overall accomplishments and highlighting some recommendations for future work, will be published after the study is finished.

REFERENCES

In Publication 60 of the International Commission on Radiological Protection (ICRP), two groups of radiation criteria are considered, including the conditions of normal practice and intervention. From our point of view, rehabilitation, which has a border position, joins these two basic situations. This is why, in this case, two additional groups of criteria must be established: (1) for intervention and (2) for abolition of intervention when acceptable levels of risk are reached.

We consider here the problem from a more general viewpoint, for cases where the level of acceptability of measures is determined on the basis of an analysis of the combined expenditures for reducing contamination to an acceptable level and the benefits to be derived from returning territories to normal economic usage.

For dose regulation, we distinguish between two principally different groups of persons: (1) the personnel, i.e. the group of people professionally occupied with the rehabilitation work (normal practice); (2) the population living on the contaminated territories (intervention, normal practice). The hierarchical structure of rehabilitation objects can be elaborated either at the State level or at the regional level or at the local level. For each object we use a hierarchical classifier of rehabilitation techniques.

In general, the measures for rehabilitation of contaminated territories can be symbolically divided into three groups, depending on the duration of the rehabilitation work and the expected time required to achieve positive results: short term (e.g. decontamination), intermediate (e.g. agricultural measures) and long term (improvement of medical services, social rehabilitation, etc.).

Countermeasures can have a direct impact, e.g. relocation of people, decontamination, restrictions on the consumption of locally produced foodstuffs, recultivation and agrochemical measures, and processing of contaminated raw material.
Countermeasures can also have an indirect, compensatory effect:

(a) Regarding the radiation factor (reduction of irradiation by reasonable organization and an increase in the efficiency of X-ray and radiodiagnostic procedures, use of materials with low natural radionuclide content for construction purposes);

(b) Regarding non-radiation factors (reduction of exposure from other industrial sources of contamination);

(c) Regarding social measures (improvement of the socio-cultural sphere and better medical supplies).

For solving high level tasks (State or regional), the analytic hierarchy process (Saaty T.L.) is best suited.

Expedience of rehabilitation work is determined by the expected level of rehabilitation expenditures, the rehabilitation 'cost' and the level of existing resources (material, technical and others).

It is obvious that all of these characteristics are interconnected by a complicated dependence. The following formula can be used as an approximate model:

\[
RL = \frac{1}{\sum_{i=1}^{M} \sum_{j=1}^{N} \frac{1}{\eta_{ij}} \exp \left( \frac{K_{ij}}{X_{ij}} \right)}
\]

where \( RL \) is the rehabilitation level, \( RL = 0-1 \); \( \eta_{ij} \) is the efficiency factor for the \( i \)th technique and the \( j \)th component of the infrastructure of the affected area, \( \eta_{ij} = 0-1 \); \( K_{ij} \) is the function for the \( i \)th technique and the \( j \)th component of the infrastructure of the affected area, which is a function of the rehabilitation 'cost' (C); and \( X_{ij} \) indicates the total rehabilitation expenses.

The rehabilitation 'cost' can be characterized by the ratio of the total benefits (B) to be derived by the society to the total expenditures (E):

\[
C = \frac{B}{E}
\]

The complexity of the quantitative estimation is determined by the fact that the benefit factor in the numerator and the expenditure factor in the denominator are of a different nature. However, regarding the standardized meaning of the factors, which vary in the range 0-1, where 0 corresponds to minimum benefits and maximum expenses, the rehabilitation 'cost' can be calculated as follows:

\[
C = \frac{1}{P} \sum_{i=1}^{L} a_i F_{Bi}
\]

\[
C = \frac{1}{P} \sum_{j=1}^{P} b_j F_{Ej}
\]
where $a_i$ and $b_j$ are weighting factors for benefits and expenses, and $F_{Bi}$ and $F_{Ej}$ have the standard meaning of benefit factors and expense factors. For standardizing the factors of benefits and expenses it is advisable to use the method of comparison with an ‘alternative’ state. This method consists of the following: For each factor the ‘best’ (the most ‘desirable’) and the ‘worst’ (the most ‘undesirable’) meanings are introduced. Then the distance of these meanings from the ‘best’ ($D^+$) and the ‘worst’ ($D^-$) alternatives is determined. The standard meaning will be

$$F = \frac{D^+}{D^+ + D^-} \quad (4)$$

Using this standard, the value of the rehabilitation ‘cost’ changes in the range 0-1, where 0 corresponds to maximum benefits at minimum expenses and 1 corresponds to minimum benefits at maximum expenses. It is pointed out that, without a description of the concrete connection of the function $K$ with the rehabilitation ‘cost’ and the level of expenses, the mentioned model is, to a large degree, qualitative. The study of this model and a more detailed description of it will be the subject of further investigations.

The choice of the optimum strategy for rehabilitation of a territory is connected with the estimation of benefits and expenses for realizing rehabilitation plans. For this purpose, a hierarchy of benefits and expenses has to be drawn up. It is necessary to estimate the vectors of the alternative priorities from the viewpoint of benefits and expenses. With regard to these factors, a choice of one of the set alternatives is made. The alternative with the maximum ratio is chosen as the method for rehabilitation of a territory. Thus, a non-linear dependence between the levels of expenditures and rehabilitation can lead to a considerable increase in expenditures, with a small positive effect. If it is decided to rehabilitate a territory and if different plans are made (scenario of technical, organizational and other measures), then it is necessary to establish a hierarchy of the benefits and expenses. The optimum alternative must be chosen by calculating priority vectors. For lower hierarchical levels (local or sub-local), more simple methods of optimization can be used, for example a cost/benefit analysis. The scheme of decision making (e.g. for populated rural areas) can be as follows: (1) estimation of the individual dose value and its structure; (2) forecast of the expected doses if no countermeasures are taken; (3) estimation of damage and total risk for the case that no countermeasures are taken, and (4) estimation of the cost and the efficiency of countermeasures and of the expected risk level.

As mentioned above, individual and collective risk will be the determining indices for creating rehabilitation optimization models. It is obvious that risk includes controlled and uncontrolled components.
The North Bohemian Cretaceous area is the newest uranium ore production area in the Czech Republic. It is also the area in which exploration, production and processing of uranium ores have had the greatest influence on the environment. The main environmental effects are the extensive changes in the regime and the quality of the groundwater, which have been caused by a disadvantageous combination of the in situ leaching (ISL) technology and classical deep mining. This involved using sulphuric acid as lixiviant and using other chemical agents in the process of leaching.

The main problem is the existence of huge amounts of contaminated groundwater in the aquifers of North Bohemian Cretaceous sediments and the potential threat to the high quality groundwater sources in this area. The main causes of the problem are:

- Disregard of environmental questions by the communist regime;
- Selection of leading managers according to the criteria of communist party discipline;
- Insufficient evaluation of geological exploration works;
- Underestimation of mining and technological conditions and false strategy in the development of the area.

The development of the problem is directly proportional to the extensive development of ISL since the mid-1960s and the parallel development of classical deep mining in the Hamr deposit, in close neighbourhood of the ISL plant.

The quality and quantity of the problem are determined by the following points:

Contamination of the Cenomanian aquifer (the lower aquifer): volume approx. 150 million m$^3$, approx. 4.8 million tonnes of total dissolved salts (TDS), area approx. 24 km$^2$.

Characteristics of the contamination: the main contaminant is SO$_4^{2-}$ (approx. 80%); high content of radionuclides, ammonia, aluminium, etc.

Contaminated water flows to the south-west, following the natural state of the hydrogeological structure, and final dewatering is done in the Labe river line. During
the flow of the water to the south-west, there is a significant risk of overflow into an upper Turonian aquifer because of the artesian character of the Cenomanian aquifer.

The present contamination of the Turonian aquifer (the upper aquifer) in the area of the ISL plant is as follows: volume approx. 70-80 million m$^3$, approx. 25 000 to 30 000 tonnes of TDS with approx. 75% of SO$_4^{2-}$, area approx. 7 km$^2$.

Systematic work has been performed between 1991 and 1995 to solve the problems with regard to four basic points:

— Geological events (analysis of potential risk in the rock environment, mathematical modelling of the influence on the groundwater);
— Physicochemical processes underground in connection with ISL and the following remediation work;
— Optimal technology for decontamination of the groundwater;
— Influence on the surface area.

These problems are being solved through wide professional co-operation (in the Czech Republic and abroad) and under Government control.

The results of the problem evaluations are as follows:

— The situation in the rock environment in the direction of natural groundwater flow to the south-west is regarded as being risky because of possible contamination overflow to the surface (Turonian aquifer).
— For removal of contamination in the Cenomanian and Turonian aquifers it is suggested to use the hydrodynamic principle: contaminated water is stabilized in the hydraulic depression and pumped to the surface.
— Removal of contamination in the Cenomanian aquifer is expected to be on a relatively high level: from the present level of approx. 25 g/L TDS to less than 3 g/L TDS.
— For the Turonian aquifer it is expected that the original groundwater quality will be restored.
— The technology for Cenomanian groundwater decontamination is based on a multistep principle, the main steps being evaporation, crystallization, mother liquor processing and use of the membrane technology (in the period of lower salinity after the year 2007).
— The technology for decontamination of the Turonian groundwater is based on the membrane technology.

The present cost estimate for the complete restoration work is more than one billion US dollars.
A recently developed deterministic model has been used for predicting activity concentrations in rivers. In the framework of the IAEA–CEC co-ordinated research programme on Validation of Environmental Model Predictions (VAMP) (aquatic working group, river subgroup), scenarios for the Clinch River/Tennessee River and for the Dnieper river have been provided. The two river systems have different characteristics, and the model was tested for both applications.

The model was initially developed to describe the transport of radioactive material in the river Danube, assuming continuous discharge. It is a simple deterministic model, taking into account dilution, interaction with suspended material, decay and sedimentation. The model was used because only a few parameters are required and because they are generally available.

The basic equation for calculating the activity concentration is:

\[ C_{i,j} = C_{(i-1),j} \frac{1}{W_j} e^{-((k'+\lambda)x) (1 - E_{i,j})} \frac{1}{1 + K_d S} \]

where \( C_{i,j} \) is the activity concentration of radionuclide \( i \) at site \( j \) (Bq/m\(^3\)), \( C_{(i-1),j} \) is the activity concentration of radionuclide \( i \) at site \( j - 1 \) (Bq/m\(^3\)), \( W_j \) is the mean annual flow at site \( j \) (m\(^3\)/s), \( W_{(j-1)} \) is the mean annual flow at site \( j - 1 \) (m\(^3\)/s), \( k' \) is the depletion factor for sedimentation (m\(^{-1}\)), \( \lambda \) is the decay constant of radionuclide \( i \) (s\(^{-1}\)), \( x \) is the distance (m), \( v \) is the flow velocity (m/s), \( K_d \) is the distribution factor (L/kg) and \( S \) is the concentration of suspended material (kg/L).

The model has been used for both scenarios, for which calculations as well as sensitivity analyses have been carried out.

Figures 1 and 2 show results for the Clinch River and the Dnieper river, respectively.

In conclusion, it is pointed out that the model gives reasonable results for simple river systems; it is, however, difficult to apply the model to complex systems with reservoirs.
FIG. 1. Experimental and computed data for $^{90}$Sr in the Clinch River (Centiers Ferry, CRM 14.5).

FIG. 2. Experimental and computed data for $^{137}$Cs in the Kiev reservoir (Dnieper).
REGULATORY IMPLICATIONS ASSOCIATED WITH THE IMPLEMENTATION OF NEW RADIOACTIVE RELEASE CONCENTRATIONS FOR RADIOISOTOPE USERS IN CANADA

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The Atomic Energy Control Board (AECB) of Canada has developed maximum release concentrations for licensed radioisotope users in hospitals, industries and research institutions, for releasing radioactive material to the environment under certain conditions. The regulatory implications of these new release concentrations, which replace the existing limits using scheduled quantities, are discussed.

In Canada, a wide variety of waste is generated from all sectors of the nuclear industry, including radioisotope users. Under certain conditions the radioactive waste may be released by controlled discharge to the environment, at which point there is no regulatory control of the radiological properties of the waste. Each release scheme is evaluated by the AECB on a case-by-case basis or as a common practice if it is used by several licensees. The doses resulting from releases must not exceed the regulatory dose limits and shall be as low as reasonably achievable. Most large operators, such as nuclear generating stations, are required to calculate their own site specific emission limits, using pathways analyses to ensure compliance with the above dose criteria. However, releases from radioisotope users normally represent such a low radiological hazard that the AECB currently permits direct releases to the atmosphere or to the sewer system, or releases as conventional solid waste, by specifying release limits in the licence, using fractions of scheduled quantities listed in AECB regulations.

In recent years, AECB staff have determined that the use of scheduled quantities is not appropriate for licensing the releases to the environment [1]. Because the scheduled quantities are based on simple intake scenarios, they do not adequately represent the possible exposure scenarios associated with these releases. Thus, the AECB has developed maximum release concentrations (MRCs) to replace the current use of scheduled quantities. The MRCs have been calculated for the most common radioisotopes released to the atmosphere, the sanitary sewer system and as solid waste, and are listed in the proposed AECB Consultative Document C-123 [2]. The MRCs are based on an individual dose criterion of 50 \( \mu \text{Sv/a} \) to a member of a critical group, using an environmental pathways analysis method similar to that used in the calculation of derived emission limits for Canadian nuclear facilities. It is recognized that more than one licensee may release radioisotopes into the same atmospheric
region, a common sewer system or landfill site. An AECB funded research study is currently investigating the levels of radioisotopes in municipal sewer and solid waste disposal systems. The results of the study will be used to determine if the doses resulting from the current releases of radioactive material by radioisotope licensees are within the above dose criteria.

The new MRCs may be incorporated directly into AECB licences in the same manner as the current limits. Licensees will be required to demonstrate that the concentrations of radioactive material released to the environment do not exceed the new MRCs specified in their licence. Licensees will need to determine the quantity of radioactive material in each solid waste package, the amount of radioactive liquid and the release rate to the sewer, and the quantity and the exhaust flow rate of gaseous radioactive material discharged to the atmosphere, and to calculate the concentrations of radioisotopes accordingly. When licensees release more than one radioisotope to the environment, they will be required to apply the summation rule. MRCs do not apply to sealed sources.

Licensees will not be permitted to deliberately dilute high specific activity materials in order to comply with the MRCs. In cases where the MRCs cannot be met, licensees may store or otherwise hold their waste for some time in order to reduce the concentrations of radioisotopes in the waste prior to discharge — a practice known as delay and decay. If licensees use radioisotopes for which no MRCs are listed in C-123, they may derive their own release limits if they can demonstrate that doses to the public will be less than 50 μSv/a. This approach is consistent with the AECB policy for exempting certain materials from further radiological control upon disposal [3]. Under this policy, the AECB uses a de minimis dose of 50 μSv/a for deciding such exemptions on a case-by-case basis.

As is currently the case with existing release limits, radioisotope licensees using the MRCs will not be required to do environmental monitoring. However, they will be required to maintain adequate records to demonstrate compliance with the MRCs.

REFERENCES

In Canada, the use of radioisotopes in hospitals, industries and research institutions is regulated through licensing by the Atomic Energy Control Board (AECB). Licensed radioisotope users are permitted to release radioisotopes to the atmosphere, the sanitary sewer system and solid waste disposal facilities, provided the concentrations of radioisotopes are less than the maximum discharge concentrations specified in their licences. These maximum values are fractions of the scheduled quantities (per unit volume or mass) of radioisotopes listed in Schedule I of the 1974 AEC Regulations, which in turn are based on the 'exempt quantities' contained in the IAEA Safety Series No. 9 [1]. In recent years, AECB staff have determined that the basis of these concentration limits is not appropriate for regulating the releases of radioactive materials to the environment. Therefore, the AECB has developed new concentration limits, called maximum release concentrations (MRCs), using an environmental pathways analysis approach. The derivation of these concentration limits is described here.

The MRCs are calculated on the basis of a dose criterion of 50 \( \mu \text{Sv/a} \) to the critical group. For each release scenario (i.e. to the atmosphere, to the sewer, to a solid waste disposal facility) the potential receptor groups are identified and the exposure pathways considered to be the most significant are described. Mathematical equations are developed for each exposure pathway, relating the release of the radioisotope from the source (as a concentration) through the environment to the receptor (as a dose). Since the MRCs will apply to a wide variety of radioisotope licensees, whose operating conditions, discharge rates, geographic locations and locations relative to populated areas are varied, it has been necessary to adopt a generic approach in terms of the models and parameters employed. Further, release concentrations rather than total release activities are calculated because of the need to develop such generic values and to facilitate implementation and verification of the MRCs. Accumulation and concentration of the radioisotopes in environmental media are taken into account, as well as dilution and dispersion in the environment. Radioactive decay is accounted for where necessary.

For releases to the atmosphere, such as via fume hoods, adults and infants living near a facility are identified as potential receptor groups. The following exposure pathways are modelled: inhalation of airborne radioactive material,
external irradiation from both immersion in a cloud and ground deposition, and ingestion of contaminated foodstuffs. Buildup in the environment is taken into account.

For releases to the sanitary sewer system, adults and infants are identified as the potential receptor groups. Exposure of sewage treatment plant workers is not modelled because a Canadian study has shown such exposures to be insignificant [2]. It is assumed that radioisotope releases to the sanitary sewer system will eventually be discharged to a receiving water body. The exposure pathways modelled are ingestion of contaminated water and fish.

Two exposure scenarios are modelled for disposal of solid waste radioisotopes. The first considers adults and infants who are exposed to atmospheric releases resulting from the incineration of solid waste, and who ingest water and fish contaminated by migration of leachate from landfill sites. The second scenario considers adults working at landfill and incinerator facilities who are exposed to ash from the incineration of solid waste, and who also ingest water and fish contaminated by migration of leachate from landfill sites. It is assumed that 1% of the waste received by disposal facilities contains radioisotopes. Further, in the first scenario it is assumed that 100% of the radioisotopes are released to the atmosphere during incineration, and in the second scenario that 100% of the radioisotopes are retained in the ash which is subsequently used for landfill.

The MRCs are described in the AECB Consultative Document C-123 [3]. This document must first be distributed for public consultation, and modified as necessary, before the values are incorporated into the AECB licensing process.

REFERENCES


RESRAD is one of ten models tested in the IAEA-CEC co-ordinated research programme on Validation of Environmental Model Predictions (VAMP), Multiple Pathways Assessment (MPA) Working Group Test Scenario S (Chernobyl data collected in southern Finland). RESRAD, the only non-dynamic model used in the VAMP–MPA Scenario S test, is a pathways analysis model designed for evaluation of radioactively contaminated soils. The RESRAD code was developed by Argonne National Laboratory and has been used by the US Department of Energy and its contractors to derive cleanup criteria for many contaminated sites.

The RESRAD code had to be modified in order to meet some of the VAMP–MPA Scenario S requirements. One major modification was the incorporation of a Monte Carlo shell to study the uncertainties of model predictions. This modification extends RESRAD from a deterministic model to a stochastic (probabilistic) model. Another improvement to the code was the addition of the ability to output media concentrations in both tabular and graphic formats. A final modification required for Scenario S was provision for the input of measured surface water concentrations for the first four years after plume arrival. This change was needed because measured $^{137}$Cs concentrations in surface water were available as part of the scenario.

Since RESRAD is a non-dynamic model, it is unable to predict the seasonal variation in media concentrations, especially for the first few years after plume arrival. However, the long term lifetime dose predicted by RESRAD compares very well with the dose calculated from observation data (see Figs 1 and 2). Radionuclide concentrations in beef have been measured at various times. The RESRAD prediction generally underestimated these concentrations for the first four years and overestimated them for the beginning of the fifth year. The lifetime ingestion dose calculated by various models and the estimated values based on observed concentrations are compared in Fig. 2. The total ingestion dose predicted by RESRAD matches the estimated dose very well.

From this exercise, we have been able to predict (with uncertainties) media concentrations, body burdens and doses for Scenario S. Our experience indicates
FIG. 1. Observed and RESRAD predicted radionuclide concentrations in beef (— RESRAD prediction, ---- uncertainties).

FIG. 2. Estimated and predicted lifetime ingestion dose.
that, although RESRAD is not specifically designed for this type of application, with proper selection of input parameters it can be used to predict media concentrations and doses, especially for the later years after arrival of the plume. We have also identified improvements of RESRAD needed to provide for better modelling under dynamic conditions.

**REFERENCE**


IAEA-SM-339/137P

**CHARACTERIZING SETTLEMENTS:**
**A CLUSTER-REGRESSION MODEL FOR IMPROVED ESTIMATES OF INGESTION DOSE IN BELARUS**

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Following the accident at Chernobyl, a range of countermeasures has been implemented with the aim of reducing ingestion doses. Byelorussian policy is that each countermeasure should only be implemented if the average individual ingestion dose within a settlement is expected to exceed a specified level. In order to implement this policy most effectively, it is therefore necessary to have reliable estimates of average doses. Since contamination levels and consumption rates for different foods vary greatly from settlement to settlement, the best method for estimating average individual ingestion doses is to use whole body measurement data for each settlement. This is technically not feasible, nor is the expenditure of so much resource justified. It is therefore necessary to adopt a modelling approach for estimating average doses.
TABLE I. COMPARATIVE PERFORMANCE OF THREE INGESTION DOSE MODELS

<table>
<thead>
<tr>
<th></th>
<th>Traditional model [1]</th>
<th>Empirical models</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>No prior</td>
<td>Full</td>
</tr>
<tr>
<td></td>
<td></td>
<td>characterization</td>
<td>characterization*</td>
</tr>
<tr>
<td>Correlation coefficient</td>
<td>0.36</td>
<td>0.41</td>
<td>0.81</td>
</tr>
<tr>
<td>Normalized standard error</td>
<td>2.7</td>
<td>0.95</td>
<td>0.32</td>
</tr>
</tbody>
</table>

* Use of the model described here.

Currently, a traditional modelling approach has been adopted in the commonwealth of independent States (CIS), which is based on measured activity concentrations in foods and consumption rate data [1]. However, this model is not very reliable since it uses consumption rate data averaged over a large area. Moreover, this approach also requires many measurements to be made in each settlement.

The empirical model described here has been developed in order to enable average individual ingestion doses in a settlement to be estimated on the basis of only limited direct radiological data or even without such data. Other empirical models which have been developed are inadequate (see Table I) because the dose estimates obtained demonstrate only a poor correlation with doses deduced from whole body measurements. This is because they do not take account of the key factors which influence the individual ingestion dose, such as the availability of privately produced milk. If the settlements are first classified according to these key factors, then a linear relationship is obtained between the ingestion dose and the density of ground contamination, from which substantially more reliable dose estimates can be obtained. This cluster-regression model is described below.

The working instruments of the cluster-regression model are cluster analysis techniques [2], including the optimization of the quality of intraccluster regression of the ingestion dose on the ground contamination level. The model was developed using data for 230 settlements in the Gomel region of Belarus. These were settlements for which detailed information was available on a wide range of radiological and associated parameters, such as ground contamination levels, whole body measurements and consumption rate data. To identify which factors were important for characterizing settlements, this cluster-regression model was tested by classifying the settlement according to a number of different sets of factors. This iterative process indicated that the key factors are the soil-to-milk transfer factor, the average consumption rate of privately produced milk, the proximity of the settlement to a forest and the level of ground contamination.
This model was tested against a further 122 settlements for which detailed radiological data were available. Each settlement was assigned to one of five model clusters, based on the four key factors. The model was then used to estimate the average individual ingestion dose from average ground contamination levels. These model estimates of the dose were then compared with doses based on whole body measurements for a settlement using two characteristics: (1) the correlation coefficient as a measure of a statistical connection between actual and estimated doses:

\[
\text{Correlation coefficient} = \frac{\sum_{i=1}^{n} (d_{i}^{\text{act}} - \bar{d}^{\text{act}})(d_{i}^{\text{est}} - \bar{d}^{\text{est}})}{\sqrt{\sum_{i=1}^{n} (d_{i}^{\text{act}} - \bar{d}^{\text{act}})^2} \sqrt{\sum_{i=1}^{n} (d_{i}^{\text{est}} - \bar{d}^{\text{est}})^2}}
\]

where \(d_{i}^{\text{act}}\) is the i-th actual dose, \(\bar{d}^{\text{act}}\) is the mean actual dose, and \(d_{i}^{\text{est}}\) and \(\bar{d}^{\text{est}}\) are the i-th and the mean estimated doses, respectively; and (2) the normalized standard error as a measure of a deviation. The results are shown in Table I. For comparison, the results for the traditional model [1] and also for an empirical model which does not first classify the settlements are also given. It can be seen that, whereas the correlation between the estimated and the actual average individual doses is not very good for the traditional model or the empirical model without prior characterization of the settlements, it is substantially improved when the model described here is used. The ratios of the standard error to the average measured whole body dose are also given in the table. Again, the model described here performs markedly better than the other two models.

ACKNOWLEDGEMENT

This work was partly funded through the CEC/CIS Joint Studies Project 2.

REFERENCES


We focus here on the results of field studies of the factors governing the distribution of $^{137}\text{Cs}$ and $^{90}\text{Sr}$ in the soil surface run-off system and the method of prediction of the concentrations of these radionuclides in the surface run-off.

The field studies consisted of simulation of rainfall on run-off plots of 1-600 m$^2$ located on typical soils of the 30 km zone around the Chernobyl nuclear power plant.

It has been shown that the $^{137}\text{Cs}$ and $^{90}\text{Sr}$ concentrations in run-off do not depend on the plot size and the rain intensity. This indicates that the radionuclides in the overland flow are in equilibrium with the upper soil layer and that the rate of attaining sorption-desorption equilibria is quite high.

The principal mechanism of reversible sorption of $^{137}\text{Cs}$ and $^{90}\text{Sr}$ is ion exchange. At ion exchange equilibrium, the concentrations of identically charged cations are related by the ratio:

$$ A_1 = \frac{A_{ex}}{K_B^{A}} \frac{B_1}{B_{ex}} $$

where $A_1$ and $B_1$ are the concentrations of A and B cations in the liquid phase, $A_{ex}$ and $B_{ex}$ are the concentrations of A and B cations in the solid phase, and $K_B^{A}$ is the selectivity coefficient for exchange of the A cation for the B cation.

As follows from Eq. (1), the concentration of radionuclides is directly proportional to the concentrations of macro-cations in run-off; this is supported by the simulated experiments of rainfall on run-off plots with different cation concentrations in the rain water. The composition of cations in the run-off is determined by the mineralization of the rain water and the release of cations from the soil solution into run-off.

In addition to the concentration of cations in the run-off, Eq. (1) includes the concentration of exchangeably sorbed cations and radionuclides in the upper soil layer, which can be determined by the standard technique with 1N ammonium acetate extraction. However, given the non-uniform distribution of radionuclides and macro-cations in the soil profile, the result will depend on the depth of the sampled
soil layer. One of the ways in which the problem can be solved is to use a concept of a complete mixing layer (CML) or, in other words, a surface layer in which the concentrations of dissolved compounds in soil solution are equal to the run-off concentration. In long term experiments, we have determined CML values from the time dependence of the $^{90}\text{Sr}$ concentration in run-off; they are about 5 mm or less for all plots. According to our data, exchangeably sorbed forms of $^{137}\text{Cs}$ and $^{90}\text{Sr}$ are distributed fairly uniformly in the upper soil layer, and the concentration of exchangeable forms of radionuclides and macro-cations in the 0–5 mm layer can be used as a measure for their concentration in the CML.

Selectivity coefficients for the upper soil layer run-off system needed for the calculations have been obtained from the results of plot experiments. We have used $\text{Ca}^{2+}$ and $\text{K}^{+}$ as competing ions for $^{90}\text{Sr}$ and $^{137}\text{Cs}$, respectively. The mean values of the selectivity coefficients are $K_{\text{Cs}}^{\text{Sr}} = 2.1 \pm 0.9$ and $K_{\text{K}}^{\text{Cs}} = 20 \pm 4$ for all of the studied plots, except for the plot near Korogod, where they are 3.8 and 67, respectively.

The values of the selectivity coefficients and the concentrations of exchangeable forms of radionuclides and macro-cations in the upper 0.5 cm of the soil have been used to calculate the wash-off coefficients. The results are presented in Table I.

**Table I. Predicted and Measured Normalized Wash-Off Coefficients of Radionuclides**

<table>
<thead>
<tr>
<th>Field plot location</th>
<th>Predicted $^{90}\text{Sr}$</th>
<th>Predicted $^{137}\text{Cs}$</th>
<th>Measured $^{90}\text{Sr}$</th>
<th>Measured $^{137}\text{Cs}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pripyat catchment</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chernobyl</td>
<td>22</td>
<td>1.6</td>
<td>$24 \pm 13^a$</td>
<td>$2.5 \pm 1.2^b$</td>
</tr>
<tr>
<td>Kopachi</td>
<td>12</td>
<td>—</td>
<td>$18 \pm 8^a$</td>
<td>—</td>
</tr>
<tr>
<td>Il'ya catchment</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Staraya Rudnya</td>
<td>—</td>
<td>3.4</td>
<td>$36.9 \pm 12.2^b$</td>
<td>$5.0 \pm 2.2^b$</td>
</tr>
<tr>
<td>Rudnya Il'yanetskaya</td>
<td>60</td>
<td>5.2</td>
<td>$36.9 \pm 12.2^b$</td>
<td>$5.0 \pm 2.2^b$</td>
</tr>
<tr>
<td>Kliviny</td>
<td>17</td>
<td>2.4</td>
<td>$36.9 \pm 12.2^b$</td>
<td>$5.0 \pm 2.2^b$</td>
</tr>
</tbody>
</table>

$^a$ Wash-off coefficients have been calculated using the radionuclide activity measured in natural run-off samples from plots during rainfall on 6 August 1988.

$^b$ Averaged wash-off coefficients for 1988 calculated for the Il'ya catchment by Nikitin et al. [1].
been used for estimation of the normalized wash-off coefficient for radionuclides in the rainfall run-off from the catchments of the rivers Pripyat and Il'ya. The concentration of cations in the run-off has been assumed to be identical with that in the rain water. The results of calculations are given in Table I. The estimated normalized wash-off coefficients, which are equal to the ratio of the radionuclide concentration in the run-off to the surface density of catchment contamination, are in good agreement with the experimentally measured values.

**REFERENCE**


IAEA-SM-339/152P

**ANALYSIS OF THE CONSEQUENCES OF POSTULATED ACCIDENTS FOR AN ADVANCED REACTOR IN THE NORTH-WESTERN NEGEV, ISRAEL**

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Yavne

D. ILBERG
Israel Atomic Energy Commission,
Tel Aviv
Israel

An assessment of the radiological consequences of postulated accidents has been performed for a 500 MW(e) high temperature gas-cooled reactor (HTGR), hypothetically located at Shivta in the north-western Negev, Israel.

Computations were carried out by means of the REMAND probabilistic consequence model [1]. Meteorological and demographic site specific input data were used. Emphasis was put on identifying the extreme meteorological conditions which characterize the arid Shivta site. Three types of rainfall were therefore simulated, including localized convective showers of high intensity [2]. The different release
categories described in a German study [3, 4] were adopted. Each category is defined by the source term for radionuclides released to the atmosphere, as well as its yearly frequency of occurrence. Short term protective measures (i.e. sheltering, evacuation, intake of stable iodine) were not considered, in order to establish the feasibility of relying only on the inherent safety of the assumed reactor.

It has been shown that the incidence of non-stochastic effects, including early fatalities, is not expected and that an incidence of stochastic effects is very low to negligible. Assuming extreme meteorological conditions, such as localized convective showers associated with an unstable atmosphere, did not significantly enhance the consequences. Because of the very low population density around the Shivta site, the consequences are much lower (by more than two orders of magnitude) than those expected from corresponding results obtained in the aforementioned German study in which representative siting conditions were assumed. Moreover, the consequences for the HTGR at Shivta were found to be much lower than those expected for severe accidents occurring in a pressurized water reactor of equivalent power at the same site, confirming the inherent safety of the assumed advanced reactor.

REFERENCES

SEASONAL DEPENDENCE OF RADIATION EXPOSURE AND RELEVANT PATHWAYS AFTER ACCIDENTAL RELEASE OF RADIONUCLIDES TO THE ATMOSPHERE

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Institut für Strahlenschutz, GSF-Forschungszentrum für Umwelt und Gesundheit GmbH, Neuherberg, Germany

The radiological consequences of an accidental release of radioactive material to the atmosphere depend strongly on the time of year when the release occurs. This is true especially for the ingestion pathway, since it is influenced to a high degree by the seasonality of plant growth, animal feeding habits, etc.

The dynamic radioecological model ECOSYS-87 [1] has been used to study the dependence of the expected doses on the time of the accidental release. The model parameters reflect the radioecological situation of central Europe. Ingestion doses have been estimated assuming local production of all foodstuffs. To demonstrate the effect of the physicochemical characteristics of radionuclides, the four nuclides $^{137}$Cs, $^{90}$Sr, $^{131}$I and $^{106}$Ru have been considered. The same activity concentration in air of $10^6$ Bq·s/m$^3$ has been assumed for all nuclides and for deposition times varying from the beginning of January until the end of December. Only dry deposition has been considered, since it demonstrates most pronouncedly the effect of seasonality. For each case the doses due to ingestion, inhalation and external exposure from airborne and ground-deposited activity have been calculated.

Figure 1 shows the resulting lifetime ingestion doses for all four radionuclides as a function of the time of deposition. The ratio of the highest ingestion doses (which occur after deposition in summer) to the lowest doses (for wintertime deposition) can reach more than two orders of magnitude. This depends mainly on

- the half-life of the nuclide considered,
- the root uptake of the nuclide,
- the mobility of the nuclide within the plant (determining the amount of translocation).

As a consequence, the variation of ingestion dose with time of deposition is highest for $^{137}$Cs. For $^{90}$Sr the variation is smaller owing to its lower translocation and its higher root uptake (which contributes during the 50 year period considered). For $^{131}$I there is a large difference between wintertime and summertime deposits, but a small variation during the growing period, since only milk and vegetables contribute significantly to the dose.
FIG. 1. Dependence of radionuclide ingestion doses on deposition time.

Because of the strongly varying ingestion dose, the relative importance of the different exposure pathways also shows a pronounced seasonal dependence. For the nuclides considered here, ingestion is often the dominant pathway after depositions during the growing period, whereas for accidental releases during the wintertime, inhalation or external exposure from the ground is more important.
Figure 2 shows the contributions of different groups of foodstuffs to the lifetime ingestion dose from $^{137}$Cs, again as a function of deposition time, which has been assumed to be the first day of each month. It can be seen that the main contribution to the ingestion dose is from different foodstuffs at different deposition times. Moreover, within the foodstuff groups shown here, the contribution of single products (the model considers 33 different products) varies considerably with deposition date.

These results demonstrate that it is very important to consider a variety of agricultural products and the development of the plants in detail, in order to give realistic assessments of ingestion doses and of the effect of countermeasures.

The seasonal dependence of food-chain transfer as shown here cannot be applied directly to other regions without model adjustment to the radioecological properties of these regions.

REFERENCE

In the context of the IAEA–CEC co-ordinated research programme on Validation of Environmental Model Predictions (VAMP), a generic model for lake ecosystems has been tested under a variety of environmental conditions. The main emphasis has been placed on predicting the levels of $^{137}$Cs in water and predatory fish, which are of most concern from a radiological point of view. The model is based upon solving first order differential equations where all parameter values are generated from pre-described distributions using Latin-Hypercube sampling. The model consists of compartments for drainage area, water masses, sediments, and up to four trophical levels in the food-chain. The actual number of compartments varies according to the ecosystem to be studied. The model responses are analysed by correlation and regression procedures for identifying the processes and parameters which mainly contribute to the uncertainty in the model results. The analyses are
performed as a function of time — in this case, twice a year; the peak values for predatory fish can also be predicted.

The use of the above method is of considerable importance when addressing the precision of model results, which is a major consideration when applying models for making predictions of future levels. By identifying the parameters which contribute substantially to the uncertainty, areas of improvement can be identified and efforts made to reduce this uncertainty. However, the uncertainty emanates from two major sources — variability and lack of knowledge. The lakes studied cover a wide range of environmental conditions and characteristics. Naturally, the generic model takes such factors into account by varying the parameter values according to the system to be studied. Not only the uncertainties can be addressed with this methodology but also the probability of exceeding certain recommended dose values. This is illustrated in Fig. 1, where the basic model is applied to the Swedish lake Hillesjön; the deposition was decreased by a factor of five for illustration. Using the original deposition, all concentrations of $^{137}$Cs in fish during the first five years would exceed the recommended dose of 1500 Bq/kg. The figure shows the probability of exceeding this level as a function of time. The calculated and observed means are also shown. It can be seen that this probability decreases with time. After one year, the arithmetic means are already below 1500 Bq/kg, although the probability of exceeding that value is 30%.

The major parameters contributing to the uncertainty in the concentrations of $^{137}$Cs in water and predatory fish for the lakes considered were identified. Similar ranges of parameter values were used, but known parameter values, for example sedimentation rates, were varied over lower ranges than unknown values. The main
FIG. 2. Major parameters contributing to the uncertainties in the levels of $^{137}$Cs in the water of lake Devoke.

The objective of this study was to compare results and to draw general conclusions concerning the major parameter contributing to the uncertainty of $^{137}$Cs concentration in water as a function of time. The results show that it may differ among lakes. The results for lake Devoke are shown in Fig. 2.

IAEA-SM-339/161P

ENVIRONMENTAL IMPACT OF LIQUID RADIOACTIVE RELEASES INTO THE SEA OF JAPAN IN OCTOBER 1993

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Russian Navy, Vladivostock

Russian Federation

On 17 October 1993, low level liquid waste (LLW) was discharged into the Great Peter Gulf of the Sea of Japan, under the control of high sensitivity underwater γ-ray spectrometric facilities. Detailed quantitative data on the radionuclide content, the total activity and the concentration in sea water at the time of this discharge were obtained.
TABLE I. RADIONUCLIDE CONTENT AND CONCENTRATION IN TANKS OF TNT-27

<table>
<thead>
<tr>
<th>Radionuclides</th>
<th>Concentration</th>
<th>Total activity</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ci/L</td>
<td>Bq/L</td>
</tr>
<tr>
<td>Cs-137</td>
<td>$3.25 \times 10^{-7}$</td>
<td>12 029</td>
</tr>
<tr>
<td>Sr-90</td>
<td>$8.97 \times 10^{-8}$</td>
<td>3 318</td>
</tr>
<tr>
<td>Cs-137</td>
<td>$2.24 \times 10^{-9}$</td>
<td>83</td>
</tr>
<tr>
<td>Co-60</td>
<td>$2.24 \times 10^{-9}$</td>
<td>83</td>
</tr>
</tbody>
</table>

From the special tanker TNT-27, 892 m$^3$ of LLW were discharged at a depth of about 2 m. Among the various $\beta\gamma$ emitting radionuclides, $^{137}$Cs prevailed (see Table I). The total activity discharged into the surface layer of the sea water was about 14 GBq (0.38 Ci).

The special spectrometric facilities "Fortepiano" were used for direct measurements of $\gamma$ emitting radionuclides in the surface water layer. These facilities were designed for detecting and measuring radioactive anomalies in sea water with high sensitivity — almost 1 pCi/L for a measurement time of about 10 s. The "Fortepiano" facilities, mounted on board of the small ship, were used to control this discharge, with the ship performing a zigzag manoeuvre along the path of the discharged LLW.

It was shown by direct $\gamma$ spectrometric analysis that 6–8 min after the discharge of LLW the $^{137}$Cs concentrations in the sea were lower than the permissible levels. After 15–25 h, the $^{137}$Cs concentrations had decreased to the background level. The size of the temporarily contaminated zone did not exceed 500 m in diameter.
VALIDATION OF MODELS FOR
RADIONUCLIDE MIGRATION
IN RIVERS AND RESERVOIRS

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\textsuperscript{1} Institute of Mathematical Machines and Systems, Cybernetics Centre, Ukrainian Academy of Sciences, Kiev, Ukraine

\textsuperscript{2} Atominstiut der Österichischen Universitäten, Vienna, Austria

INTRODUCTION

The IAEA–CEC co-ordinated research programme on Validation of Environmental Model Predictions (VAMP) was a good opportunity to validate assessment models on the basis of data from environmental releases of radionuclides. After the Chernobyl accident, a set of mathematical models was developed in the Cybernetics Centre, Kiev, to simulate radionuclide dispersion in the Pripyat–Dnieper river/reservoir system. These models were validated on the basis of two scenarios prepared by the VAMP River and Reservoirs Sub-group. The first scenario describes the long term release of $^{137}$Cs, $^{90}$Sr, $^{106}$Ru and $^{60}$Co into the Clinch River–Tennessee River system. The second scenario was prepared for long term prediction (from 1987 to 1991) of water and sediment contamination of the Dnieper set of reservoirs by $^{137}$Cs and $^{90}$Sr after the Chernobyl accident and for the calculation of the doses to the Ukrainian population.

METHODS

The existing precise presentation of the river systems and the radioactive releases permits implementation of different kinds of models describing the main processes, involving migration of long lived radionuclides in rivers, bottom sediments and aquatic food-chains. For obtaining the dispersion of radionuclides in water bodies, two types of model can be used: the one-dimensional model RIVTOX and the null-dimensional (box) model WATOX \cite{1}. These models can be used to simulate the propagation of pollutants in dissolved and suspended form; there are also
sub-codes for studying radionuclide sedimentation–resuspension and absorption–desorption processes. Both codes are time dependent and can also calculate the flow field and internal transport of suspended sediment. Migration of radionuclides in bottom sediments is described by the one-dimensional model BOTTOX, which uses data from the RIVTOX model for the sedimentation rate and the concentration of a toxicant in the upper bottom layer. The special code for fish contamination and dose calculation is the last one in this chain of models.

FIG. 1. Dynamics of $^{137}$Cs in water of the Kiev reservoir (Dnieper river study).

FIG. 2. Core profile of $^{137}$Cs in bottom sediments (Clinch River–Tennessee River study).
RESULTS

The simulations were done in two steps. During the first stage, results were prepared for comparison with previously unknown experimental data. The best estimate and the confidence interval were obtained with the help of uncertainty analysis. Subsequently, the model parameters were tuned in order to reach good agreement with experiment and were examined for their sensitivity so that the ones most important with regard to radionuclide migration in river systems could be defined.

For the Dnieper study, site specific model parameters were obtained, such as the distribution coefficient and time parameters for absorption and desorption processes which allow the measured data to be compared with the model results (Fig. 1). The results of the core profile simulation are shown in Fig. 2; this simulation was done in the frame of the Clinch River–Tennessee River scenario and covers the period from 1947 to 1983.

ACKNOWLEDGEMENT

One of the authors (A.M.) is indebted to the IAEA for support by fellowships.

REFERENCE

In the course of its work for the International Arctic Seas Assessment Project (IASAP) under the auspices of the IAEA, the Source Term Working Group has developed simple models to predict the patterns of radionuclide release into the Kara Sea from spent nuclear fuel (SNF) and from activated components of the seven nuclear submarines dumped by the former Soviet Union.

The models account for the progressive removal of containment barriers through corrosion and other mechanisms, breakdown of the organic fillers, and degradation and leaching from the fuels. Where reliable data are currently unavailable, such as filler degradation rates, simple ‘worst case’ assumptions are made. Release rates are predicted up to the year 4500 for pressurized water reactors (PWR) and beyond the year 10 000 for liquid metal reactors (LMR).
### TABLE I. KARA SEA DUMPING SITES AND DETAILS OF THE DUMPED SUBMARINE REACTORS

<table>
<thead>
<tr>
<th>Dumping site</th>
<th>Dumped unit</th>
<th>Submarine factory number</th>
<th>Number of reactors$^a$</th>
<th>Year of dumping</th>
<th>$^{235}$U Load (kg)</th>
<th>Enrichment (%)</th>
<th>Estimated total activity for 1994 (Bq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Abrosimov Fjord</td>
<td>Reactor compartment</td>
<td>901</td>
<td>2</td>
<td>1965</td>
<td>100</td>
<td>20</td>
<td>$7.3 \times 10^{14}$</td>
</tr>
<tr>
<td></td>
<td>Reactor compartment</td>
<td>285</td>
<td>1</td>
<td>1965</td>
<td>100$^b$</td>
<td>7.5</td>
<td>$6.6 \times 10^{14}$</td>
</tr>
<tr>
<td></td>
<td>Reactor compartment</td>
<td>254</td>
<td>2</td>
<td>1965</td>
<td>100$^c$</td>
<td>20</td>
<td>$9.5 \times 10^{12}$</td>
</tr>
<tr>
<td></td>
<td>Reactor compartment</td>
<td>260</td>
<td>2</td>
<td>1966</td>
<td>100$^c$</td>
<td>20</td>
<td>$5.1 \times 10^{12}$</td>
</tr>
<tr>
<td>Novaya Zemlya depression</td>
<td>Reactor in special container</td>
<td>421</td>
<td>1</td>
<td>1972</td>
<td>50</td>
<td>20</td>
<td>$2.9 \times 10^{14}$</td>
</tr>
<tr>
<td>Stepovogo Fjord</td>
<td>Submarine</td>
<td>601</td>
<td>2$^d$</td>
<td>1981</td>
<td>180</td>
<td>90</td>
<td>$8.4 \times 10^{14}$</td>
</tr>
<tr>
<td>Tehcheniye Ford</td>
<td>Reactors in special container</td>
<td>538</td>
<td>2</td>
<td>1988</td>
<td>100$^c$</td>
<td>20</td>
<td>$4.5 \times 10^{12}$</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td></td>
<td></td>
<td>6</td>
<td></td>
<td>7</td>
<td>730</td>
<td>$2.5 \times 10^{15}$ (6.8 $\times 10^4$ Ci)</td>
</tr>
</tbody>
</table>

$^a$ All reactors are of the PWR type unless otherwise noted.

$^b$ One reactor fuel load of 50 kg was removed before disposal.

$^c$ Both reactor fuel loads of 50 kg each were removed before disposal.

$^d$ Liquid metal reactors.
The reactors were dumped at four locations along the coast of Novaya Zemlya. Table I gives details of the objects dumped at each location, together with the date of dumping and the estimated total activity for each object in 1994.

The Source Term Working Group is continuing to refine and revise the models as more data become available from the former Soviet Union.

IAEA-SM-339/180P

ASSESSMENT OF DOSES FROM RADIOACTIVE RELEASES OF THE PAKS NUCLEAR POWER PLANT

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The nuclear power plant in Paks started operation in 1982 and is now working with four blocks of PWR type reactors made in the former Soviet Union. In addition to the plant itself, the following authorities are involved in monitoring of the environment: Ministry of Agriculture, Ministry of Environment and Regional Policy and Ministry of Welfare. The National Research Institute 'Frédéric Joliot-Curie' for Radiobiology and Radiohygiene — as a member of the authorized environmental radiation monitoring system — is responsible for the dose assessment of the population around the nuclear power plant. For routine operation the institute has to carry out yearly dose calculations and for accidental cases it is involved in assessment of doses from the food-chain.

The main radionuclides released from the plant in 1993 and their chemical forms are given in Table I.

Dose assessment from atmospheric releases

For normal operation the dose calculations are based on a Gaussian plume model for steady state solution [2]. Some minor modifications have been introduced only for deposition rates. The dose constants from external radiation (air and surface deposition) have been taken from Ref. [3]. The transport of radionuclides through the food-chain has been described by compartment models developed in the IAEA-CEC co-ordinated research programme on Validation of Environmental Model Predictions (VAMP).
TABLE I. RELEASES OF RADIONUCLIDES IN RELATION TO THE INDIVIDUAL DOSE NEAR THE PLANT [1]

<table>
<thead>
<tr>
<th>Nuclides</th>
<th>Atmospheric releases</th>
<th>Aquatic releases</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ar-41</td>
<td>37.0 TBq (noble gases)</td>
<td>—</td>
</tr>
<tr>
<td>Kr-85</td>
<td>0.16 TBq (noble gases)</td>
<td>—</td>
</tr>
<tr>
<td>Kr-88</td>
<td>0.24 TBq (noble gases)</td>
<td>—</td>
</tr>
<tr>
<td>Xe-133</td>
<td>5.2 TBq (noble gases)</td>
<td>—</td>
</tr>
<tr>
<td>Xe-135</td>
<td>4.3 TBq (noble gases)</td>
<td>—</td>
</tr>
<tr>
<td>H-3</td>
<td>3.9 TBq (HT, HTO)</td>
<td>18 TBq</td>
</tr>
<tr>
<td>C-14</td>
<td>1.0 TBq (CO₂, CH₃)</td>
<td>—</td>
</tr>
<tr>
<td>Co-60</td>
<td>25.9 MBq (aerosol)</td>
<td>563 MBq</td>
</tr>
<tr>
<td>Sr-90</td>
<td>0.063 MBq</td>
<td>2.1 MBq</td>
</tr>
<tr>
<td>Ag-110m</td>
<td>1.26 GBq (aerosol)</td>
<td>0.365 GBq</td>
</tr>
<tr>
<td>I-131</td>
<td>0.26 GBq (aerosol + gas + organic)</td>
<td>0.014 GBq</td>
</tr>
<tr>
<td>Cs-137</td>
<td>29.0 MBq</td>
<td>0.394 GBq</td>
</tr>
</tbody>
</table>

Table II gives the results of dose assessments for steady state solutions with respect to the different pathways.

**Dose assessment from aquatic releases**

The water system is represented by a compartment model and the polluted Danube water is being investigated. The most important pathways considered in the model are: external exposure from the polluted river, river side and irrigated land; and internal exposure due to drinking water, and consumption of fish, vegetables, milk and meat.

For normal operation, the steady state water concentrations of radionuclides in the Danube are calculated by a simple dilution model. The concentrations of the different foods are estimated by the concentration factor method based on Ref. [4], with the data for food consumption and environmental usage adapted to the situation in Hungary. The external and internal doses due to annual liquid effluents from the plant are given in Table III. The critical radionuclide is ³H and the critical pathway in the aquatic food-chain is drinking water.

The doses due to aquatic releases are much lower than the doses due to air-borne effluents (~10%).
TABLE II. INDIVIDUAL DOSE CONTRIBUTION OF AIRBORNE RELEASES NEAR THE PLANT

<table>
<thead>
<tr>
<th>Pathways</th>
<th>Dose (nSv)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>External radiation</strong></td>
<td></td>
</tr>
<tr>
<td>Noble gases (from air)</td>
<td></td>
</tr>
<tr>
<td>Ar-41</td>
<td>38</td>
</tr>
<tr>
<td>Kr-85</td>
<td>0.02</td>
</tr>
<tr>
<td>Kr-88</td>
<td>0.51</td>
</tr>
<tr>
<td>Xe-133</td>
<td>0.28</td>
</tr>
<tr>
<td>Xe-135</td>
<td>1.5</td>
</tr>
<tr>
<td>Ag-110m (surface deposition)</td>
<td>7.4</td>
</tr>
<tr>
<td>Others</td>
<td>1</td>
</tr>
<tr>
<td><strong>Internal radiation</strong></td>
<td></td>
</tr>
<tr>
<td>Inhalation</td>
<td>0.2</td>
</tr>
<tr>
<td>Ingestion, Ag-110m</td>
<td>5.8</td>
</tr>
<tr>
<td>Ingestion, C-14</td>
<td>30</td>
</tr>
<tr>
<td><strong>Total dose</strong></td>
<td>86</td>
</tr>
</tbody>
</table>

TABLE III. EXTERNAL AND INTERNAL DOSES DUE TO ANNUAL LIQUID EFFLUENTS (1993)

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Exposure (nSv)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>External</td>
</tr>
<tr>
<td>H-3</td>
<td>a</td>
</tr>
<tr>
<td>Mn-54</td>
<td>0.07</td>
</tr>
<tr>
<td>Co-60</td>
<td>0.46</td>
</tr>
<tr>
<td>Ag-110m</td>
<td>0.07</td>
</tr>
<tr>
<td>I-131</td>
<td>a</td>
</tr>
<tr>
<td>Cs-134</td>
<td>0.06</td>
</tr>
<tr>
<td>Cs-137</td>
<td>0.14</td>
</tr>
<tr>
<td>Others</td>
<td>0.02</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td>0.82</td>
</tr>
</tbody>
</table>

*a Dose less than 0.001 nSv.
REFERENCES


IAEA-SM-339/193P

A FOREST AND NATURAL ECOSYSTEM MODEL FOR SHORT AND LONG TERM DISTRIBUTION OF RADIONUCLIDES FROM NUCLEAR ACCIDENTS

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The Chernobyl accident in 1986 caused radionuclide contamination in most countries in eastern and western Europe. A prime example is Belarus, where 23% of the total land area received chronic levels; about $1.46 \times 10^6$ ha of forested lands were contaminated with $> 1-5$ Ci/km$^2$ ($37-185$ kBq/m$^2$), and $2.5 \times 10^4$ ha received $> 40$ Ci/km$^2$ ($1480$ kBq/m$^2$) of $^{137}$Cs and other long lived radionuclides,
such as $^{90}$Sr and $^{239,240}$Pu [1]. Since the cumulative radiological dose will occur over long time periods (decades to centuries), we need to determine what countermeasures can be taken to limit this dose so that the affected regions can, once again, safely provide habitat and natural forest products. These contaminated regions now provide a global environmental testing laboratory for the international scientific community.

To address these problems, we have attempted to formulate a mathematical model, FORESTPATH, which describes the kinetic processes and pathways of radionuclides in forest and natural ecosystems and which can be used to predict the

FIG. 1. Conceptual representation of the FORESTPATH model.
FIG. 2. Accumulation of activity for an acute deposition of $^{137}$Cs in generic forests: (a) coniferous and (b) deciduous; 1 — organic layer, 2 — tree, 3 — understory, 4 — labile soil, 5 — fixed soil. Leaching to deep soil is negligible (note the scale of the ordinate for 1 to 100% and (2-5) to 50%.

radionuclide levels in the future (see Fig. 1). The generic model calculates the time dependent radionuclide concentrations in compartments, using as input literature values of the residence times for the two forest types: coniferous and deciduous, as shown in Fig. 2 and Table I.

One objective is to assess the short and long term radiological hazards caused by the radionuclides initially released and deposited from the reactor source [2]. A
second objective is to provide process information on the forest ecosystem which can be used in land rehabilitation and countermeasures for the affected regions. A third objective is to provide early public information relative to policy decision making in the event of another such nuclear reactor accident. To test and validate the models, we require actual site specific data, which are now being collected by several co-ordinated groups in Belarus and Ukraine at sites shown in Table II.

A quality controlled field sampling programme in the Chernobyl exclusion zone was initiated in July 1994 by the International Sakharov College on Radioecology, which has both exploratory and monitoring functions and which extends a sampling programme initiated in 1992 [3]. The exploratory sampling is designed to collect information on functional relationships among different components of the contaminated ecosystem, while monitoring over several years provides information on the dynamics of radionuclide cycling in these ecosystems. Measurements of the 500 samples and data collected will serve as an initial approach to validating the predictive model. We seek answers to the following questions:

— In a given region, what is the regional deposition of the several radionuclides (kBq/m²)?
— What is the radionuclide distribution pattern among forest compartments, i.e. tree, understory, organic layer, soil?
— What is the radionuclide distribution pattern among tree components, i.e. branches, leaves, needles, bark, stem, roots?
— What is the infiltration rate of radionuclides in the soil compartments?
— Do different initial contamination levels affect the inter-compartmental distribution of radionuclides?
— What are the differences in radionuclide cycling in deciduous, coniferous and mixed forests?
— What is the influence of ‘hot particles’ on the radionuclide cycling?

REFERENCES


**TABLE I. FORESTPATH MODEL COEFFICIENTS USED AS $^{137}$Cs INPUT IN GENERIC FORESTS**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Coniferous forest</th>
<th>Deciduous forest</th>
<th>Reliability</th>
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<tr>
<td></td>
<td>Generic value</td>
<td>Range</td>
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<tr>
<td>Absorption half-time (years)</td>
<td>0.64</td>
<td>0.15–5</td>
<td>0.64</td>
</tr>
<tr>
<td>Tree biomass (%)</td>
<td>60</td>
<td>45–75</td>
<td>60</td>
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<tr>
<td>Understory biomass (%)</td>
<td>20</td>
<td>5–35</td>
<td>20</td>
</tr>
<tr>
<td>Desorption half-time (years)</td>
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<td>Interception</td>
<td>0.8</td>
<td>0.6–0.9</td>
<td>0.5</td>
</tr>
<tr>
<td>Fraction (%)</td>
<td>—</td>
<td>—</td>
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<tr>
<td>Leaching half-time (years)</td>
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<td>100–6000</td>
<td>400</td>
</tr>
<tr>
<td>Organic layer</td>
<td>8</td>
<td>1–100</td>
<td>3</td>
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<tr>
<td>Removal half-time (years)</td>
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<td>—</td>
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<td>Radiation half-time (years)</td>
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<td>Tree uptake half-time (years)</td>
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<td>1</td>
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<td>Short (&lt;1 week)</td>
<td>Intermed. (&lt;1 year)</td>
<td>Long (&gt;1 year)</td>
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**Notes:**
- GRAPH: Graphical representation available.
- NA: Not applicable.
- Poor, Moderate, Good: Qualitative assessment of performance or quality.
<table>
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<tr>
<th>Characteristics</th>
<th>Station 2 Babchin</th>
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<th>Station 5 Korenevka (Control)</th>
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</table>
WHEN DOES CHEMICAL TOXICITY OF $^{129}$I BECOME IMPORTANT?

S. SHEPPARD
AECL Research,
Atomic Energy of Canada Ltd,
Pinawa, Manitoba, Canada

The environmental impact of $^{129}$I has traditionally been assessed by its radiological effects on human health. However, protection of human health does not ensure protection of other biota, and there is increasing need to demonstrate protection of all parts of the environment. Because of the very long half-life of $^{129}$I, a relatively high molar concentration of iodine is associated with any specified level of radioactivity. The potential exists for the chemical toxicity of iodine to exceed the radiological toxicity of $^{129}$I in importance.

In comparison with radiological effects, the chemical toxicity of $^{129}$I to humans is not a concern. This is related to the regulatory objective to protect individual humans through a long life-span. Thus, long term carcinogenesis is a key impact to be avoided and only very low risks are accepted. Radiation effects dominate this risk assessment. However, for other organisms, a common objective is to only protect populations, not all individuals. Thus, regulatory targets for non-human biota may only seek to avoid acute radiation effects, and higher concentrations of $^{129}$I may be considered acceptable. These may be so high that chemical toxicity supersedes radiation effects in importance.

In laboratory bioassays, we found that levels as low as 5 mg iodine/kg soil caused detrimental effects on growth and survival of terrestrial organisms [1]. Similarly, Laverock et al. [2] found that levels as low as 0.16 mg iodine/L water caused mortality in water flea (Daphnia magna). If the iodine in these studies had been $^{129}$I, the corresponding radiological dose rates would have been less than 0.1 $\mu$Gy/h, below which consistent radiological effects are expected [3]. Thus, the effects of radiation from $^{129}$I would not have been as important as the chemical toxicity of iodine. In addition, environmental stable iodine would augment the effect of the chemical toxicity of $^{129}$I.

In comprehensive risk assessments, where effects on both human and non-human biota are considered, these findings may be significant. Clearly, if humans were assumed to utilize to any large extent the same soil and water resources as the other biota, then the $^{129}$I contamination levels considered acceptable for humans would also protect the other biota. However, in settings where humans do not have full access to the same soil and water, effects on non-human biota may become
important in the overall risk assessment. For $^{129}$I, these effects will likely involve chemical toxicity.

REFERENCES


IAEA-SM-339/201P

**THE SOIL INGESTION PATHWAY:**
**ENRICHMENT, BIOAVAILABILITY AND OVERALL IMPORTANCE**

S. SHEPPARD
AECL Research,
Atomic Energy of Canada Ltd,
Pinawa, Manitoba, Canada

Soil ingestion is an important exposure pathway for radionuclides that are not otherwise very mobile in the environment. The health of both humans and animals can be affected. Models of the pathway require estimates of the amounts of soil ingested, the concentration of radionuclides relative to the original soil (enrichment) and the bioavailability of the radionuclides ingested with soil in the gut. There are few data on enrichment [1, 2] and there is only some recent information on bioavailability [3–7].

The basic model of soil ingestion can be expressed as follows:

$$EIS = (FI \times SI_f) \times CE + RB$$  \hspace{1cm} (1)

The effective ingestion of soil, EIS, is a function of the amount of soiled food ingested per unit time, $FI$; the soil ingested per unit food, $SI_f$; the soil ingested per unit time because of other activities, $SI_a$; the concentration enrichment, $CE$; and the relative bioavailability of the radionuclides on the soil, $RB$. The units of EIS are: soil intake per unit time, comparable with the units of $SI_a$ and the product of $FI$ and $SI_f$. Both $CE$ and $RB$ are unitless ratios.
In many cases, CE will have values greater than one because the processes that lead to soil ingestion also cause a selection of the smaller soil particles. Because of their large surface area per unit mass, these small particles are enriched in radionuclides. For example, CE values above unity result from the rainsplash of soil onto plants, with subsequent erosion of the larger particles off the leaves by rain and wind. Values of CE can be estimated from information about the mechanical analysis of a soil:

$$CE = \frac{240S + 2400C}{240S + 2400C + 24(1 - S - C) S + C} \frac{1}{S + C}$$  \hspace{1cm} (2)$$

The parameters S and C refer to the mass fraction of silt sized and clay sized particles in the original soil, and the constants are the relative particle surface areas for each size class, including the residual fraction, \((1 - S - C)\), which is the fraction of sand sized particles.

Values of RB may be greater than unity because the radionuclides sorbed or otherwise associated with soil particles may not be as readily absorbed from the gut as the same radionuclides in food or water. The magnitude of RB varies among radionuclides and may be especially large for caesium. Our data suggest a value for caesium of about 4. The value is also expected to vary inversely with particle size.

REFERENCES

### CHAIRMEN OF SESSIONS

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<td>W. WHICKER</td>
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<td>2</td>
<td>F. FRY</td>
<td>United Kingdom</td>
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<tr>
<td>3</td>
<td>L. HÅKANSON</td>
<td>Sweden</td>
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<td>4</td>
<td>L. BULDAKOV</td>
<td>Russian Federation</td>
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<td>M. ZHELEZNYAK</td>
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<td>6</td>
<td>I. MALATOVA</td>
<td>Czech Republic</td>
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<td>7</td>
<td>F.O. HOFFMAN</td>
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<tr>
<td>8</td>
<td>M. BALONOV</td>
<td>Russian Federation</td>
</tr>
<tr>
<td>9</td>
<td>A. AARKROG</td>
<td>Denmark</td>
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<tr>
<td>G.S. LINSLEY</td>
<td>Scientific Secretary</td>
</tr>
<tr>
<td>M.S. BAXTER</td>
<td>Co-Scientific Secretary</td>
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<tr>
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