Topical Day on Site Remediation

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Characterisation of environmental contamination with radium in Sint-Jozef-Olen

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Abstract

Radium has been dispersed in the environment in Sint-Jozef-Olen as a result of the operation from 1922 until 1969 of one of the world’s largest radium extraction plants. SCK•CEN has under contract with the government (DBIS) and with Union Minière contributed to the radiological characterisation of the contaminated sites: dumping-grounds, streets and river banks. The estimation of the population exposure required complementary information with respect to the inhalation and ingestion pathways. The principal motive to restore the sites is the fact that most of the scenarios concerning the future use of the contaminated land give rise to enhanced exposures to future generations. A committee has been established to draw up a global restoration plan. In the committee there seems to be a consensus to dig off the contaminated road foundations and river banks, incorporating them into the restoration of the dumping ground.

1 Radium, the first nuclear industry

In 1915, ore with an uranium oxide content of about 50% was discovered in Zaïre, the former Belgian Congo. In 1921 the first ore arrived in Belgium and in 1922 radium production began in Olen. Within one year Belgium dominated the world market and this until the mid 1930's when comparable high grade ore was discovered in Canada. In 1938 the Belgians and Canadians divided the world market to stabilize the price. The production continued in Canada until the mid 1950's and in Belgium until the mid 1960's. The total radium production in Olen was above 1000 gram. Due to this activity radium was dispersed in the environment.

2 Radiological characterisation of the environmental contamination

2.1 Introduction

In 1989 and 1990 media coverage of some very high contaminations in the village of Sint-Jozef-Olen resulted in a detailed radiological characterisation of the contaminated areas and an evaluation of their impact on the population exposure. The federal ministry of public health and environment (DBIS) assigned the programme to the Nuclear Research Centre (SCK•CEN) and the Institute of Hygiene and Epidemiology (IHE). The final report was approved in March 1993 and was subsequently presented by the federal ministry of public health and environment to the population.

2.2 Programme

The external exposure was determined by means of a dose rate scanning of the dumping grounds, the streets and the banks of the Bankloop, a brook that received the liquid waste. Radon measurements were performed in all the dwellings within a wide
range around the former radium factory and in open air on and nearby the dumping grounds.

The intake of radium through the food chain or by resuspension was investigated. As a check of the internal contamination the radium concentration in milk teeth of children of the municipal primary school of Sint-Jozef-Olen was determined.

### 2.3 Results and dose assessment

**Radon in dwellings and in open air**

Radon measurements were performed in all of the 846 neighbouring dwellings. In 6 dwellings the average radon concentration was above 150 Bq/m$^3$ in one of the living areas. They were inspected with portable gamma detectors and only in the dwelling with the highest radon concentration an enhanced dose rate was found in a veranda over a length of 5 m and a maximum width of 2 m. The maximum value is 600 nSv/h which is 5 to 10 times the background. The average radon concentration in the veranda was 720 Bq/m$^3$, in the living room 370 Bq/m$^3$; in the cellars 120 Bq/m$^3$ and in the bedrooms 100 Bq/m$^3$. Continuous monitoring during 2 weeks showed large variations in radon concentration between 20 and 3 000 Bq/m$^3$ depending on the ventilation and the meteorological conditions. The exposure of the occupants is 11 mSv per year assuming a dose conversion factor of 50 μSv/year per Bq/m$^3$ and a residence time of 30 % in the bedroom and 50 % in the living room. According to the owner, the lowlying ground next to his dwelling was raised including debris from the company owning the former radium plant. This debris was afterwards used as foundation for the veranda.

The radon concentrations in open air on and nearby the dumping grounds, were measured during one year with alpha track detectors. The measured values are normal for outdoor air, except on and nearby the D1 dumping ground. The average value of the 7 detectors at a height of 1.5 m above the dumping ground is 180 Bq/m$^3$. At a short distance of the D1 dumping ground in the prevailing wind direction an average radon concentration of 170 Bq/m$^3$ is measured in a garden. With a comparable radon concentration at the site of the dwelling, the contribution of the dumping ground to the exposure of the occupants is about 5 mSv per year.

**Dose rate scanning of the roads**

All the roads in the neighbouring village (Sint-Jozef-Olen) and in some suspected areas were monitored with a van equipped with several NaI detectors. The distance between two registrations was between 5 and 7 m depending on the speed of the van. The results are shown in figure 1. All the places above 200 nSv/h were subjected to a detailed review by means of automatic hand-carts especially designed for this purpose. Every 5 m the dose rate was registered in the middle of the road and on a fixed distance left and right of the middle. Attaching a surface of 5 m by 2 m at a nodal point and taking the isolated points into account, results in 5 800 m$^2$ of roads above 200 nSv/h of which 1 950 m$^2$ are above 400 nSv/h.
Fig. 1: Dose rates of the roads in the vicinity of the former radium extraction plant in Olen.

**D1 dumping ground**

The dumping ground D1 has a surface of about 100,000 m². Despite the sometimes dense vegetation, a grid was established with a nodal point every 25 m. The dose rates at the nodal points and at some local maxima were measured with portable gamma detectors. 153 out of a total 197 nodal points have dose rates above 200 nSv/h. The maximum value and the mean value are 150,000 nSv/h and 2,800 nSv/h. A local maximum of 10⁶ nSv/h was found.

The median value of the nodal points is 1,000 nSv/h which is less than half the mean value. The reason for this is the presence of very contaminated places. This is apparent from the high radium concentration of some samples. The maximum value is 34,000 Bq/g. The dose rate varies often over several orders of magnitude over distances of a few meter.

**Brook the Bankloop**

The liquid effluents of the former radium extraction plant were released into the brook the Bankloop. The contamination was mapped out from the fence of the plant to the mouth into the river the Kleine Nete. The dose rates were measured every 10 m in the middle of the Bankloop, at the borders and at both banks every 2 m until the background value was attained.

The Bankloop is 1,800 m long from the fence to the mouth. The first 1,000 m are a residential area. Then the Bankloop flows through an agricultural area. Thirty years ago, as a result of soil reclamation work, the last 420 m before the mouth was displaced. The pastures on the place of the former bed were measured according to a grid with a nodal point every 10 m.
Everywhere along the historical Bankloop a distinct enhancement of the dose rate is observed. Indeed, in 84 out of the first 144 sections a dose rate of 1 000 nSv/h is registered in at least one of the nodal points. The longest distance between two sections with a nodal point of at least 1 000 nSv/h is 80 m. The contaminated strip along the Bankloop is greatly limited to 10 m. It is mainly caused by the regular dredging of the sediment. In 47 sections 2 000 nSv/h is measured and in 17 sections at least 5 000 nSv/h. The highest dose rate of a nodal point is 50 000 nSv/h, registered in the death branch caused by the new crossing of the Bankloop with the canal. Nearby, a local maximum of 100 000 nSv/h was found. The contaminated surface along the Bankloop is estimated at 7 000 m$^2$.

Dose rates above 1 000 nSv/h are not found along the new Bankloop. This indicates that the present situation is determined by activities before 1960, thus before the displacement of the bed. In the pastures on the place of the former bed and between the former and the new bed an enhanced dose rate is observed. The investigated area has a surface of 55 000 m$^2$. The distance between nodal points is 10 m.

The average value of the 552 nodal points is 300 nSv/h. In 27 points a dose rate of 1 000 nSv/h or more is measured. The maximum value is 5 500 nSv/h. The contamination is caused by the frequent floods of the Bankloop. The surface of the pastures with a dose rate above 200 nSv/h is estimated at 25 200 m$^2$ and with a dose rate above 400 nSv/h at 9 900 m$^2$.

*Ingestion pathways*

Airborne dust and the intake of surface and ground water were shown to be insignificant exposure pathways.

The $^{226}$Ra concentration of a number of biological samples from contaminated areas was measured with the intention of determining the exposure of the food chain. Milk samples were taken from 2 farms with pastures and fields partly on grounds with an enhanced radium concentration. These grounds constitute less than 20 % of the surface area of the farms. The average $^{226}$Ra concentration of the milk, assuming a milk consumption of 120 l per year, corresponds to an intake of only 1.2 Bq.

Along the Bankloop at the place with the highest dose rate (100 000 nSv/h), 2 grass samples were taken with a time difference of 4 months. They contained a $^{226}$Ra concentration of 103 and 10.6 Bq/g dry weight, respectively. The radium concentration of the top soil is 230 Bq/g confirming the soil-to-plant transfer factor for grass on sandy soil of 0.13 of the IUR database.

In two places along the Bankloop a chicken run was found partly on contaminated grounds. The radium concentration of the eggs without shell for the most contaminated chicken run was 0.02 Bq/g fresh weight, for the less contaminated chicken run 0.002 Bq/g and 0.00012 Bq/g for an uncontaminated chicken run. Assuming a consumption of 10 kg per year the exposure is respectively 48 μSv/year, 4.4 μSv/year and 0.3 μSv/year. During the field work no crops for direct human consumption were found on contaminated grounds. It is however not improbable that this changes in the future. Considering the soil-to-plant transfer factors of the IUR, an assumed annual consumption of corn of 60 kg dry weight results in an exposure of 1 140 μSv. The consumption of 5 kg legumes constitutes an exposure dose of 51 μSv, 5 kg root crops of 56 μSv, 20 kg potatoes of 44 μSv and 5 kg vegetables of 470 μSv. Especially the production of vegetables has to be avoided. In our society it is unlikely that someone lives purely on locally cultivated foodstuffs. Therefore, the dose to the critical groups will probably be below 1 000 μSv per year.
The analysis of human teeth is one of the few methods to track an internal $^{226}$Ra contamination. Milk teeth were collected with the help of the municipal primary school of Sint-Jozef-Olen which is situated along the Bankloop at about 200 m from the fence of the former radium factory. The analysis of the milk teeth confirmed the actual absence of significant ingestion pathways.

3 Growing consensus of opinion in favour of a global restoration plan

A committee has been established to draw up a global restoration plan. In this plan, the D1 dumping ground can play a central role if there is an agreement to incorporate the contaminated road foundations and the banks of the Bankloop into the restoration of the dumping ground.

SCK•CEN has contributed in several areas. First, an active soil-depressurization system has been installed in a dwelling with radium-contaminated material under a veranda. A cavity was excavated beneath the floor of the veranda and soil gas is extracted continuously via a fan to a discharge point outside the home. Preliminary results show a decrease in radon concentration by a factor 10, to less than 100 Bq/m$^3$. Secondly, under contract with Union Minière, SCK•CEN has determined the extent and the depth of the radium contamination of the D1 dumping ground. Thirdly, by request of the authorities and in collaboration with VITO, a chemical and radiological characterization of the depth profile of the Bankloop was made. The volume of the contaminated banks in the residential area is estimated at 6 250 m$^3$. The cleanup strategy of the riverbed sediment is determined by the chemical pollution. In order to comply with the acceptance criteria for dumping grounds, it could be necessary to decrease the leachability of some heavy metals by an immobilization treatment. The volume of the contaminated sediment in the residential area is about 2 200 m$^3$, bringing the total volume at 8 450 m$^3$. Finally, on request of the municipal authorities of Olen and Geel, SCK•CEN has assisted in the remediation of the Grensstraat in Olen and the Sint-Corneliusstraat in Geel. The intervention level was decided on 200 nSv/h. The contaminated road surface, between 500 and 1 000 m$^3$, was transfered to the D1 dumping ground. Finally, on behalf of Union Minière, SCK•CEN made a radiological assessment of the different options to remediate the D1 dumping ground. More details on this study are given by Lieve Sweeck in her contribution to this topical day.

The next SCK•CEN task in this dossier is probably writing a compilation, bringing together all the relevant information so that the competent authorities can decide on the necessary licences to carry out the environmental restoration in Sint-Jozef-Olen.
Measurement techniques for Radiological characterization of contaminated sites
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Abstract

Once the decision is taken to characterize a contaminated site, appropriate measurement techniques must be selected. The choice will depend on the available information, on the nature and extent of the contamination, as well as on available resources (staff and money). Some techniques will be described on the basis of examples of characterization projects (Olen e.a.)

1 Introduction

Each remediation project has a specific starting point. This can be described as the moment that the awareness of a possible problem emerges. The reason for the awareness can be public concern induced by the media reporting on "problems", or the scientific world realizing the existing of underestimated effects, or any other incentive. If a "problem" emerges, an iterative mechanism of approach starts, with cycles of decision making, characterization, assessment and back to decision making in order to come to the remediation of the problem. A conclusion can also be that there is no problem. Because expenses for remediation projects tend to be high, it is important to carefully plan and follow up planning of a remediation project. The radiological characterization tries to estimate the source term for the radiological assessment.

2 Objectives of radiological characterization

- To verify the presence of radioactive contamination.
- To determine radiation and contamination levels, and the affected areas.
- To determine the dispersion of radionuclides following run-off, resuspension, migration, plant uptake, water transport ...
- To give the necessary input for the radiological assessment, and assist in the optimization of the remediation strategy.
- To monitor the results of intermediate or final remediation or clean-up in order to verify that remediation objectives and/or regulatory requirements are met.

3 Characterization approaches

The general approach to radiological (and other) characterization includes:
- planning
- investigation
- data analysis
- reporting.
As a first step in the process, planning is essential to properly develop a characterization strategy to meet the objectives of the remediation project. Once the initial planning is conducted, it is necessary to perform office, field and laboratory investigations to collect the information to meet the objectives. The collected data are reduced into an appropriate format and integrated and stored for future use. A description of the investigation and the related results are documented in a report.

There is a tremendous variability within this approach framework, because all contaminated sites have different contaminant histories. Any strategy, developed to characterize a specific site, must be highly flexible and be phased to revise the strategy as new information becomes available.

3.1 Planning

Proper planning is essential to provide a structure for directing the design of environmental data collection operations. International experience learns that inadequate planning can result in problems such that:
• the wrong variables were measured
• some variables that were needed were not measured
• the wrong set of samples was taken
• the data are compromised by interfering factors
• too much money was wasted on unnecessarily sophisticated instrumentation and analytical techniques
• the realized accuracy and precision are inadequate
• too much or too few samples were collected
• the methods used were not approved by regulators
• characterization consumed all available money, and nothing was left for remediation.

A Quality Assurance Plan defines how the integral quality of the environmental data collection process will be assured. The objectives are to achieve technically sufficient investigation work, at the right time and at the right price. Protocols will be available to assure the quality of the measurements.

Data Management is part of this process and will provide data traceability, data access and archiving the data for future use. Forms can be defined to facilitate the field and laboratory work to comply with QA requirements.

3.2 Investigations

For a complete characterization of a site, it will be necessary to conduct investigations on a number of subjects. This may include investigations on the history of the site and on historical data, on the contaminant sources, on the radioactive contamination, on chemical co-contaminants, on the geomorphological situation, on the geology of the site, on hydrogeologic and hydrologic characteristics, on the climate, on the pedologic properties, on the flora and fauna, on the demographic situation and on land and water use at the site.
3.3 Data analysis

The data collected will be analysed to be reduced to a format that is appropriate for assessment. This includes identification and concentrations of contaminants, and vertical and lateral distribution.

3.4 Reporting

Reports must describe the findings of the investigations, and document the decisions made and actions taken at all stages of the investigations. These reports may be consulted by different people, with different backgrounds, not necessarily having detailed technical knowledge. This makes it vital that a high standard of presentation is achieved and that the results and their assessment are presented with clarity and precision.

4 Measurement techniques and procedures

4.1 Preparation

To be able to assure the quality of the measurements, it is important to define protocols for the use of instruments, the calibration of the instruments, for taking and analysing samples and for the recording of data. The teams must be trained to follow the protocols, especially for large projects where many inexperienced people may have to participate.

The exact location of each sample or measurement must be unambiguously referred to a standard grid system. There must be a close relationship between the analysing laboratories and the characterization management, in order to optimize the number of samples, the required precision and the measuring range. The protocols may change at some point in the project, it is fundamental that measurements be related to the exact protocol of application.

4.2 Site surveys

In the first phase of monitoring, the areas of concern should be delineated. Depending on the size of the suspected area, this can be done by a quick aerial survey or a mobile survey. The aerial survey by airplane or helicopter can give valuable information on the extent of the contaminated area, but calibration and pinpointing hot spots is difficult.

Mobile systems mounted on a vehicle give much better spatial resolution, but are limited to roads and accessible places. Fine tuning, also useful if access with vehicles is difficult or impossible, can be done using appropriate hand-held instruments.

Combination of these types of surveys will be used in the different phases of monitoring. The in situ survey will result in data on the surface of the contaminated area, and possibly on the radionuclides involved.
4.3 Sampling, drilling

Sampling will be used to verify the isotopic composition of the contamination. If the lateral and vertical distribution is not too different in a larger area, sampling can be used to "calibrate" the survey instruments and to convert the readings to activity measurements. Sampling and drillings will provide the necessary information on the depth profile of the contamination. This information, together with the results of the surveys, gives the opportunity to estimate the characteristics and the volume of waste that will be produced in different remediation scenarios.

4.4 Measurement equipment: the Olen example

Aerial survey with helicopter

A helicopter survey of St-Jozef-Olen was used in the 1960's to give a general overview of the contamination of the region [1]. The helicopter was equipped with a large (4" x 4") NaI detector and associated electronics. The helicopter did fly at a height of 40 m and at 40 knots. The results were recorded on paper, and correlated with visible marks on the ground (see Fig. 1).

Fig. 1: Results of a helicopter survey of Sint Jozef Olen
In 1991-1992, an automatic γ-measuring system was installed in a van to carry out a mobile survey of all the roads of St-Jozef-Olen. The van was equipped with two large 4" x 4" NaI detectors, capable of measurements down to background levels, and two 2" x 2" NaI detectors to be able to measure higher dose rates (see Fig. 2).

The determination of the position of the van was done by inputting a starting point, and measuring the distance by sensors on the wheels and the driving angle by means of a gyrocompass. The nuclear and position data were automatically stored in the computer system, located in the van.

Calibration of the results was done in terms of dose rate for $^{226}$Ra. The van travelled at 5 to 7 km/h resulting in a measurement every 5 to 7 m.

The results were treated off-line. The data of the four detectors were combined and the data of the position logging were corrected for errors of the gyroscope. A map was built, where each measured result is represented at the location of measurement in a colour depending on the measured dose rate.
Detailed survey [2,3] A detailed survey was used to pinpoint the hot spots and to have a more detailed view of the easily accessible places, with increased dose rates. These suspected areas were measured by means of carts that were designed at SCK•CEN for this purpose (see Fig. 3).

Fig. 3: A detailed survey was done by means of carts with battery operated equipment and datalogger.

These 3-wheel carts were designed to be pushed by one person, easily stockable and entirely autonomous. They were equipped with a 2" x 2" NaI detector, a battery-operated linear rate meter, and a battery-operated data logger with a simple input facility for the operator and automatic logging of the scale of the linear rate meter. The carts were used to measure the nodal points of a grid and to search for local maxima. Off-line read-out of the data loggers for data analysis and reporting was performed between the different parts of the detailed survey. Hand-held instruments were used in less accessible places such as dumping grounds, water surfaces, river banks and on grounds with much vegetation. The dose rate results were written down on forms in such a way that the exact location of each point of measurement was registered.
Sampling and drilling

Samples were taken during the detailed survey mostly on local maxima, in order to verify in the laboratory that the contamination was related to the $^{226}$Ra production and to estimate the local $^{226}$Ra concentrations.

The measurement of samples of soil, water, sediments, vegetation, foodstuffs, will give information on the migration of radioactivity in the foodchain.

Drillings were performed in the D1 dumping ground [4], because the superficial contamination was high and because it was suspected that a large part of the contamination was dumped in layers in the past.

An instrument to log the depth profile of the dose rate was developed. A watertight 2" x 2" NaI detector was used again, together with a battery-operated ratemeter and data logger. The detector was led down in the bore-hole by means of a battery-operated motor.

The depth position of the detector and the dose rate were measured by the data logger (see Fig.4).

Fig. 4: Instrument to log the depth profile in a borehole at grounds with difficult access
Retrieval of the data was done off-line for further data analysis and reporting (see Fig. 5).

![Drilling](image)

**Fig 5 Example of a depth profile**

### 4.5 Other tools for characterization

**High resolution γ-spectrometry**

High resolution γ-spectrometry in the field can be used if a mixture of isotopes is expected, or if near-background measurements have to be done. The system uses a repeatable fixed geometry, with the detector at 1 to 1.5 m above ground. Because it takes 15 minutes to 1 hour to make the acquisition for one point, the distance of the nodal points in a grid is usually large (km).

**α- and β-measurements**

In situ measurement of α and β emitters is very difficult and often only qualitative. Only few techniques exist.

For the assessment of plutonium concentration levels, the FIDLER detector has been reported [8]. This detector uses the 60 keV X-ray of $^{241}$Am as a tracer for plutonium. There are also reports on the measurement of $^{90}$Sr-$^{90}$Y [9].

Other systems are being developed (DOE-US), including laser-induced fluorescence [10] for the gross detection of uranium and a long range α detector [11], based on the detection of secondary ionization of air by the decay of α-emitters.
Resonant Sonic Penetration Technology

Resonant Sonic Penetration is becoming available as an alternative to classic drilling and heavy weight penetrometers. It has been demonstrated as a tool to access the subsurface for installation of monitoring and/or remediation wells. The technique does not use drilling fluids, minimizes generation of waste and worker exposure. This cone penetrometer can be configured with a variety of sensors, to collect groundwater, soil, gas or sediment samples, to measure in-situ contaminant concentrations using spectrometric methods, to make geophysical measurements, as well as to measure traditional geotechnical parameters to determine the lithologic properties of the subsurface.

Air/gas measurements

SCK•CEN has a long time experience with sampling stations for collection of airborne dust and our laboratories are capable of sophisticated analysis of radionuclide content to very low levels. Rn sampling and measurements were performed in great detail in the Olen region.

Mobile laboratories

Sample preparation and analysis is usually done in a laboratory building with experienced staff and sophisticated measuring equipment. The services of such laboratories may not be available in some cases, for example, if it is not permitted to transport samples off-site or move them across local boundaries. In this case, the laboratory staff will work in a mobile laboratory on site.

A whole range of configurations exists. A simple solution with high mobility could include a van with sampling equipment, limited sample preparation facility and a gamma-spectrometry system. A very sophisticated "transportable" solution could include a series of interconnected laboratory equipment, comparable to the ones used in fixed laboratories, meteorologic instrumentation, a ventilation system with high efficiency filters, air conditioning and a decontamination facility. Anything in between these two options is possible. The choice will depend on the characteristics of the site and on the type and size of the contamination, on the duration of the project, on available resources (staff and money), on the possibility to replace some sampling and laboratory measurements by in-situ measurements and on the possibility of access to fixed laboratories.

Direct communication between the mobile laboratory and the headquarters of the characterization/remediation project is essential. This can be done by radio, cellular telephone (GSM) or standard telephone and fax if a connection is available.

Mobile laboratories exist in many countries in the frame of nuclear emergency preparedness. These can be very useful in a characterization project, but they may need some adaptation for the specific problems of the site under investigation (special sampling and measuring equipment).

Global Positioning System (GPS)

The United States Department of Defense operates a satellite based system of absolute positioning which allows a low-cost hand-held device to give locations anywhere in the world to an absolute accuracy of about 100 m. The signals from the satellite are usually deliberately perturbed to limit the accuracy. By obtaining correcting signals from one or more base stations at known locations, positioning over a large area, such as a city or even continent, to an accuracy approaching 1 m is possible. Such a system, called Differential GPS (DGPS) can be operated entirely by the user, either in real-time or by post-facto corrections. Alternatively, correction signals are provided by commercial organizations against payment. The corrections can
be broadcast over a local radio network, multiplexed with other signals or transmitted over satellite links. The overall result is a portable system that can be carried by a person or fitted to a vehicle and can provide accurate locations. GPS or DGPS require, however, a clear view of the sky and cannot be used inside buildings or amongst closely spaced trees or buildings.

5 Conclusion

SCK•CEN is capable to perform the radiological characterization of contaminated sites. There is no specific research programme on the development of new techniques but the experienced staff is specialized in adapting existing techniques for the special requirements of each remediation project.

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Assessment of the radiological impact of contaminated discharges
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Abstract

In our department, we are using a biosphere model to calculate the release of radionuclides from contaminated soils and their dose impact on critical individuals in the environment. Normal evolution and accidental scenarios are considered. The goal of the model is to provide an indication of the radiological risk rather than a real prediction of the future impact.

1 Introduction

Two kinds of sources are considered. Near-surface dumping sites which contain radioactive contaminated waste produced by the non-nuclear industry (e.g. phosphate industry) and sites contaminated by former nuclear practices. The releases of radionuclides from these primary sources may lead to the exposure of humans via different pathways. To assess the dose impact on individuals of the critical group (the group is representative of those individuals expected to receive the highest dose equivalents from source of radiation under consideration [1]), a biosphere model was developed. This model describes the transport of the radionuclides released into the biosphere, their transfer through the foodchain and the subsequent exposure to humans. There are two release types to be taken into account: the normal evolution release and the accidental release. In the next sections, the different pathways of the two release types will be discussed.

It should be emphasized that the biosphere model and the transfer parameters herein are generalized and can only be used to give an indication of the radiological risk.

2 The normal evolution scenario

Fig. 1: Exposure pathways of the normal evolution scenario (in case of a $^{226}$Ra contamination)
This scenario calculates the dose impact due to the normal evolution of the primary source. The biospheric compartments and exposure pathways are given in figure 1. We may assume that during the first few hundred years after dumping (control period), the primary source is guarded and not accessible for the public. Radionuclides are leached out (with percolating water) and migrate to the ground water. The contaminated water flows through the underground and reaches the biosphere by discharges into surface water (e.g. river) or a well sunk in the aquifer.

As radionuclides move with the water flow, they may be retarded by physico-chemical interactions such as complexation, adsorption, precipitation, etc. In the present work, these processes will collectively be termed sorption. The overall effect of sorption/desorption reactions is the retardation of the radionuclides' migration relative to the water in which they were dissolved. This effect is represented by a retardation factor. Among the parameters involved, the distribution factor is the most important parameter affecting the transport. This parameter, which is a characteristic of each nuclide, is defined as the concentration sorbed on the solid phase to the concentration in the water phase. It is a very sensitive factor that varies with three orders of magnitude for some elements (e.g. $^{137}$Cs), depending on the soil type (sand, loam, clay) and the ionic status of the soil.

To model the transport of the radionuclides in the biosphere, several assumptions are made:
- the radionuclides are released at a constant rate
- the water flows at a constant velocity through a homogeneous underground
- the transport of radionuclides through the primary source and the aquifer is advective
- the migrating nuclides are in sorption equilibrium with the soil and this relationship is linear.

The surface water and well are secondary sources. They are contaminated by radionuclides migrated from the primary source. While the radionuclide concentration of the primary source will decrease progressively due to radioactive decay and physical releases, the radionuclide concentration of the secondary source will first increase until a certain maximum, due to accumulation of the radionuclides, released by the primary source.

The water of the secondary sources can be used for different purposes and may lead to irradiation of man by three main routes: inhalation of resuspended activity, external irradiation and ingestion of contaminated food. For Ra-226, a fourth route has to be considered, namely inhalation of exhaled radon decay products.

A closed, self-sustaining agricultural system is used to calculate the radiological risk of critical exposed individuals. In such system the exposures of individuals will be maximized by preventing any significant dilution with uncontaminated, external material as well as preventing losses from the system, except those identified in the transport sub-models.

It is not our intention to give a exhaustive list of equations used to calculate the dose impact on man. For such lists, we refer to [2] and [3]. In the following section, the relevant processes and characteristics will be briefly discussed.
2.1 External exposure

As seen in figure 1, two types of external exposure are considered.

**Soil**
Due to irrigation with contaminated water from a well or a river, the soil becomes contaminated. Irrigation flux and irrigation period determine the input of radionuclides in the root zone. The losses are brought about by leaching and radioactive decay. Men working on this soil (e.g. farmers) will be exposed to external irradiation.

**Sediment**
For the activity that concentrates in the sediment, the main exposure route is by external irradiation to people who spend time on those sediments deposited on the margins of rivers.

2.2 Internal exposure

We consider four types of internal exposure pathways: ingestion of drinking water, ingestion of contaminated foodstuffs and inhalation of resuspended particles and radon decay products.

**Drinking water**
Drinking water may be prepared from ground water (well) or river water. Preparation of drinking water from ground water is assumed not to involve any treatment, affecting the radionuclide concentration in the drinking water. Preparation of drinking water from river water on the other hand is assumed to involve the removal of solids from the water. If the concentration of the suspended particles and the corresponding distribution coefficient are known, it is possible to calculate the radionuclide concentration in the filtrated river water.

**Foodstuffs**
Food crops and pasture are considered to become contaminated through irrigation. The radionuclides are transferred to the plant by direct foliar absorption and by root uptake from the soil. For most radionuclides, the root uptake is negligible compared to the foliar absorption uptake. The values of the soil-to-plant concentration factor depend on the radionuclide, soil type (sand, loam, clay) and plant species and may vary with three orders of magnitude for some elements.

Contamination of animal products (milk and meat from cow) is possible via intake of contaminated animal feed, pasture and soil (while grazing) and via intake of contaminated water. The radionuclide concentration in milk, meat and fish are very sensitive to their corresponding milk/meat/fish transfer factors because all the contaminated routes are affected by these factors.

**Inhalation of resuspended particles**
The concentration of radionuclides (sorbed on resuspended particles) in air is assumed to be proportional with the dust loading. This may be an overestimation. Especially in the case of small contaminated surface areas, the dust loading also contains uncontaminated dust. Mostly however, the dose due to inhalation of soil particles is very small.

**Inhalation of radon decay products**
Radon gas ($^{222}$Rn) is a daughter product of $^{226}$Ra, which becomes available through emanation. Inhalation of radon decay products may enhance the dose significantly.
3 The accidental scenarios

There are various accidental scenarios: involuntary accidents like earthquakes, flooding, etc. and voluntary intrusions (caused by humans) through constructions (roads, buildings and residential houses) and cultivation of the land on the site, etc. The effect of accidents mostly consists in enhancing the entrance of radionuclides in the environment, thereby increasing exposure to humans. In the case of accidents, however, not only the dose consequences are to be considered but also the probability of occurrence of the accident, as risk is determined by both.

The human intrusion scenario is one of the most important accidental scenarios. In the frame of this paper, two types of human intrusion scenarios are considered: the construction scenario and the residential scenario and they are assumed to occur after the control period.

3.1 Construction scenario

The critical group are workers exposed during the construction of buildings, routes, etc. on the site and the re-use of contaminated material for construction purposes off-site. Important exposure pathways are: inhalation of (mechanical) resuspended radionuclides and external irradiation directly from the primary source. In the case of $^{226}$Ra contaminated sites, the radon exhalation from the primary source will be increased.

3.2 Residential scenario

The critical group are the people living in houses on the contaminated site and eating vegetables grown in their contaminated gardens. Relevant exposure pathways are: external irradiation from the primary source, inhalation due to wind resuspension of contaminated soil during gardening, ingestion of food crops grown on the contaminated soil and in case of a $^{226}$Ra contaminated site also radon inhalation.

4 Example: performance assessment of a contaminated site

Two examples are considered: a dumping site contaminated with the highly sorptive radionuclide $^{226}$Ra and a site contaminated with the very mobile radionuclide $^{129}$I by former nuclear practices. For these arbitrary examples, we calculate individual doses (critical group) per unit contamination (1 Bq/g).

4.1 The normal evolution scenario

It is assumed that the aquifer discharges in a well localized 100 m from the site or in a river localized 1500 m from the site. Important parameters for the radiological assessments are given in table 1. In the absence of site-specific data, the parameter values are default values taken from basic documents [1], [4]. The transport of the radionuclides through the aquifer is based on a simple one-dimensional advection model considering retardation due to sorption.
Table 1: Parameters relevant for the normal evolution of the arbitrary sources (dw = dry weight; fw = fresh weight)

<table>
<thead>
<tr>
<th>Site dimensions:</th>
<th>length (m)</th>
<th>width (m)</th>
<th>depth (m)</th>
<th>infiltrating rate ground water (m/y)</th>
<th>porosity</th>
<th>density (kg/m³)</th>
<th>distribution coefficient (m³/kg) for ¹²⁹I; 5 (²²⁶Ra)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Site</td>
<td>200</td>
<td>100</td>
<td>0.5</td>
<td>0.1</td>
<td>0.33</td>
<td>1300</td>
<td>0.001</td>
</tr>
<tr>
<td>Aquifer</td>
<td>Darcy velocity (m/y)</td>
<td>3.5</td>
<td>distribution coefficient (m³/kg) for ¹²⁹I</td>
<td>0.0005</td>
<td>distribution coefficient (m³/kg) for ²²⁶Ra</td>
<td>0.5</td>
<td>depth (m)</td>
</tr>
<tr>
<td>Soil</td>
<td>infiltrating water velocity (m/y)</td>
<td>0.1</td>
<td>porosity</td>
<td>0.33</td>
<td>density (kg/m³)</td>
<td>1300</td>
<td></td>
</tr>
<tr>
<td>Well</td>
<td>flux (m³/y)</td>
<td>10000</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>River</td>
<td>flux (m³/h)</td>
<td>9000</td>
<td>concentration of suspended particles (mg/l)</td>
<td>30</td>
<td>distribution coefficient (m³/kg) for ¹²⁹I; 2.5 (²²⁶Ra)</td>
<td>0.0005</td>
<td>concentration factor fish (m³/kg)</td>
</tr>
<tr>
<td>Irrigation</td>
<td>irrigation flux (l/d)</td>
<td>1</td>
<td>irrigation period (d/y)</td>
<td>100</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Crops</td>
<td>depth root soil layer (m)</td>
<td>0.3 (food crops); 0.15 (pasture)</td>
<td>mass interception factor (m²/kg fw; (m²/kg,dw) fw)</td>
<td>0.15 (food crops); 2 (pasture)</td>
<td>exposure time (d)</td>
<td>60 (food crops);30 (pasture)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>half-life, due to weathering (days)</td>
<td>15 (¹²⁹I); 30 (²²⁶Ra) for foodcrops</td>
<td>soil-plant concentration factor (dw/fw)</td>
<td>8 (¹²⁹I); 15 (²²⁶Ra) for pasture</td>
<td>soil-plant concentration factor (dw/dw)</td>
<td>0.023 (¹²⁹I); 0.005 (²²⁶Ra) for food crops</td>
<td></td>
</tr>
<tr>
<td></td>
<td>translocation factor</td>
<td>1.0 (¹²⁹I); 0.12 (²²⁶Ra) for pasture</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Meat/milk</td>
<td>transfer factor cow milk (d/l)</td>
<td>5 × 10⁻³ (¹²⁹I); 4.0 × 10⁻³ (²²⁶Ra)</td>
<td>transfer factor beef (d/kg)</td>
<td>1.1 × 10⁻² (¹²⁹I); 5.0 × 10⁻⁵ (²²⁶Ra)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>daily uptake of pasture by cow (kg dw/d)</td>
<td>14</td>
<td>daily uptake of water by cow (m³/d)</td>
<td>0.06</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>fractional uptake of soil by cow</td>
<td>0.04</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Air</td>
<td>concentration of dust particles (kg/m³)</td>
<td>1.5 × 10⁻⁴</td>
<td>breathing rate of man (m³/h)</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Consumption by man</td>
<td>food crops (kg fw/y)</td>
<td>170</td>
<td>drinking water (m³/y)</td>
<td>0.4</td>
<td>milk (l/y)</td>
<td>150</td>
<td>beef (kg/y)</td>
</tr>
<tr>
<td>External exposure time</td>
<td>field (h/y)</td>
<td>1000</td>
<td>sediment (h/y)</td>
<td>250</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dose factors</td>
<td>inhalation (Sv/Bq)</td>
<td>4 × 10⁻⁴ (¹²⁹I); 2.3 × 10⁻⁶ (²²⁶Ra)</td>
<td>ingestion (Sv/Bq)</td>
<td>6.4 × 10⁻⁴ (¹²⁹I); 3.6 × 10⁻⁷ (²²⁶Ra)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>external irradiation (Sv,m²/Bq.h'¹)</td>
<td>1.0 × 10⁻¹⁶ (¹²⁹I); 3.8 × 10⁻¹³ (²²⁶Ra)</td>
<td>emanation (Sv,g/Bq.y'²)</td>
<td>1 × 10⁻³ (²²⁶Ra)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Results

The maximum annual doses to critical individuals through the different pathways are given in table 2. Concerning protection measures, no barriers or other means of protection were considered. It is clearly seen that due to higher sorption on the solid phase (as indicated by the higher distribution coefficients in table 2), the doses are significantly lower for $^{226}$Ra releases than for $^{129}$I releases. The doses from the $^{226}$Ra contaminated site are also lower than the exemption level (0.01 mSv/g/Bq.y), the level beneath which the radiological risk is considered to be negligible. For the well as secondary source, the exemption level will only be reached at $^{226}$Ra contamination levels of at least 1.72 kBq/g.

Whereas for the $^{129}$I contaminated site, the exemption level is already exceeded at concentrations of 1 Bq/g, as demonstrated in table 2. In this latter case, site remediation has to be taken into consideration. The decision to take protection measures, however, does not only depend on radiological factors and should therefore be optimized. This requires a multi-attribute analysis, taking into account radiological, economical and social factors.

Compared to well water, the doses obtained for river water as secondary source are much lower due to sorption and radioactive decay. Consequently, this effect is more pronounced for the highly retarded $^{226}$Ra and the concentrations at which the exemption level is reached are much higher.

Table 2: Maximum annual doses for a member of the critical group (mSv.g/Bq.y) due to releases from the secondary source.

<table>
<thead>
<tr>
<th>$^{129}$I release from well (maximum time = 70 y)</th>
<th>dr. water</th>
<th>food crops</th>
<th>milk</th>
<th>meat</th>
<th>ext. irrad.</th>
<th>inhalation</th>
<th>total</th>
</tr>
</thead>
<tbody>
<tr>
<td>$4.2 \times 10^{-1}$</td>
<td>$2.6 \times 10^{-1}$</td>
<td>$3.2 \times 10^{-1}$</td>
<td>$1.9 \times 10^{-1}$</td>
<td>$1.1 \times 10^{-6}$</td>
<td>$5.0 \times 10^{-9}$</td>
<td>1.2</td>
<td></td>
</tr>
</tbody>
</table>

| $^{129}$I release from river (maximum time = 496 y) | dr. water | food milk meat external | inhal. fish | ext.irrad | total |
|---|---|---|---|---|---|---|
| $3.4 \times 10^{-4}$ | $2.1 \times 10^{-4}$ | $2.5 \times 10^{-4}$ | $1.5 \times 10^{1}$ | $9.1 \times 10^{-10}$ | $4.0 \times 10^{-12}$ | $2.1 \times 10^{-4}$ | $1.6 \times 10^{-9}$ | 1.2 $10^{-3}$ |

<table>
<thead>
<tr>
<th>$^{226}$Ra release from well (maximum time = 22000 y)</th>
<th>dr. water</th>
<th>food milk meat external</th>
<th>inhalation</th>
<th>exhalation</th>
<th>total</th>
</tr>
</thead>
<tbody>
<tr>
<td>$2.7 \times 10^{-3}$</td>
<td>$3.2 \times 10^{-8}$</td>
<td>$4.9 \times 10^{-9}$</td>
<td>$1.6 \times 10^{-9}$</td>
<td>$1.7 \times 10^{-8}$</td>
<td>$1.2 \times 10^{-12}$</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$^{226}$Ra release from river (maximum time = 282130 y)</th>
<th>drink. water</th>
<th>food milk meat external</th>
<th>inhal. fish</th>
<th>ext.irrad</th>
<th>exhal. total</th>
<th>sed.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$2 \times 10^{-60}$</td>
<td>$2 \times 10^{-60}$</td>
<td>$4 \times 10^{-61}$</td>
<td>$1 \times 10^{-61}$</td>
<td>$1 \times 10^{-60}$</td>
<td>$1 \times 10^{-64}$</td>
<td>$1 \times 10^{-60}$</td>
</tr>
</tbody>
</table>
4.2 The intrusion scenarios

Based on the probability of occurrence and on the dose consequence, the construction of houses has been selected as being the most important construction scenario. We assume that the workers build medium-sized houses with a foundation of 3.5 m and that the contaminated soil dug out will be spread around the house. Relevant parameters for the human intrusion scenarios are given in table 3.

Table 3: Parameters relevant for the intrusion scenarios

<table>
<thead>
<tr>
<th>Construction scenario</th>
<th>Residential scenario</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total construction time (h)</td>
<td>Exposure time (h/y)</td>
</tr>
<tr>
<td></td>
<td>in the house 6960</td>
</tr>
<tr>
<td>Concentration of dust particles (kg/m$^3$)</td>
<td>outside 1800</td>
</tr>
<tr>
<td>Breathing rate of man (m$^3$/h)</td>
<td>Concentration of dust particles (kg/m$^3$)</td>
</tr>
<tr>
<td></td>
<td>in the house 1.5 $10^8$</td>
</tr>
<tr>
<td></td>
<td>outside 3.0 $10^8$</td>
</tr>
<tr>
<td></td>
<td>Breathing rate of man (m$^3$/h)</td>
</tr>
<tr>
<td></td>
<td>in the house 0.75</td>
</tr>
<tr>
<td></td>
<td>outside 1.2</td>
</tr>
<tr>
<td></td>
<td>Dose factor emanation (Sv.g/Bq.y$^{-1}$)</td>
</tr>
<tr>
<td></td>
<td>in the house 16.5 $10^{-3}$</td>
</tr>
<tr>
<td></td>
<td>outside 1 $10^{-3}$</td>
</tr>
<tr>
<td></td>
<td>Correction factor (geometry) for external</td>
</tr>
<tr>
<td></td>
<td>in the house 0.25</td>
</tr>
<tr>
<td></td>
<td>outside 0.75</td>
</tr>
</tbody>
</table>

Results: The maximum individual doses are given in table 4 for the construction scenario and table 5 for the residential scenario. These doses are reached the year after the control period, which is assumed to be 200 years in this example. The probability of occurrence was not taken into account, therefore no comparisons can be made with the exemption level.

For the construction scenario, it is seen that the external irradiation has the greatest dose impact. The depth of the construction, the construction time and the radionuclide content of the site are the main characteristics in the impact assessment.

For the residential scenario, the inhalation of $^{222}$Rn determines the dose impact of the $^{226}$Ra contaminated site completely. Whereas for $^{129}$I, the ingestion of plants is the main exposure pathway.
Table 4: Maximum doses for a member of the critical group (mSv.g/Bq) due to the construction of a house on the primary source.

<table>
<thead>
<tr>
<th>radionuclide</th>
<th>ext. irradiation</th>
<th>inhalation</th>
<th>exhalation</th>
<th>total</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{129}$I</td>
<td>$4.8 \times 10^{-5}$</td>
<td>$1.7 \times 10^{-5}$</td>
<td>/</td>
<td>$6.5 \times 10^{-5}$</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td>$1.6 \times 10^{-1}$</td>
<td>$3.4 \times 10^{-3}$</td>
<td>$4.9 \times 10^{-2}$</td>
<td>$2.1 \times 10^{-1}$</td>
</tr>
</tbody>
</table>

Table 5: Maximum annual doses for a member of the critical group (mSv.g/Bq.y) for the residential scenario.

<table>
<thead>
<tr>
<th>radionuclide</th>
<th>ext. irrad.</th>
<th>inhalation</th>
<th>ingestion</th>
<th>exhalation</th>
<th>total</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>soil</td>
<td>plants</td>
<td>indoors</td>
<td>outdoors</td>
</tr>
<tr>
<td>$^{129}$I</td>
<td>$5.4 \times 10^{-3}$</td>
<td>$7.4 \times 10^{-7}$</td>
<td>$1.7 \times 10^{-4}$</td>
<td>$2.9 \times 10^{-2}$</td>
<td>/</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td>$1.9 \times 10^{-1}$</td>
<td>$1.6 \times 10^{-4}$</td>
<td>$6.2 \times 10^{-3}$</td>
<td>$1.5 \times 10^{-1}$</td>
<td>$9.8$</td>
</tr>
</tbody>
</table>

5 References


Decision Aiding Techniques for Site Remediation
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Abstract

Decision making problems in the nuclear domain are renowned for their complexity since they usually involve a wide range of technical, social and political considerations. Site restoration is a typical example of a complex nuclear decision problem, and more and more decision makers realise that they need new tools to assist in the decision making process. This paper shortly reports on multi-criteria decision analysis, a powerful tool for handling complex decisions involving multiple criteria. The motivation to use multi-criteria decision analysis in the domain of site restoration is illustrated, and we conclude by touching upon new developments and challenges of the field.

Keywords: multi-criteria, decision analysis, nuclear decision problems.

1 Introduction

The development of multi-criteria decision analysis is to be situated in the area of Operations Research, a body of various decision techniques originally designed to support the enormous operational needs of the allied armies at the end of the Second World War and later directed to a variety of operational business problems. The reader is without doubt familiar with the well-known decision techniques in such application domains as inventory control or queueing.

The main characteristic and goal of multi-criteria decision analysis is to allow the decision maker to take into his or her consideration multiple criteria at the same time. Its main attractiveness is twofold:

- foremost its capacity to provide a structured analysis tool for real-life decision problems, and secondly its suitability to model
- so-called intangible problems, for instance public anxiety or psychological stress.

Two distinct schools contribute to the further development of multi-criteria decision analysis: the Anglo-Saxon school and the French school.

The theoretical foundations of the Anglo-Saxon school are to be situated in utility theory, the main difference being that (probabilistic) uncertainty is not considered. To clarify this difference, the theoretical foundations of the Anglo-Saxon school are referred to as Multi-Attribute Value Theory or MAVT in short.

MAVT requires from the decision maker a well-balanced decision structure, often referred to as a decision tree, where the criteria deemed relevant to the decision by the decision maker are hierarchically structured. Consequently, the possible alternative actions are rated or scored against all criteria. The final outcome of this procedure is an ordered set of alternatives, presented in a linear ranking from worst to best.
The French school holds a totally different point of view. Rather than asking the decision maker to provide a global hierarchical structure, alternatives are only compared pairwise. In this comparison, the decision maker expresses his or her preference (or lack of preference, being indicated as either indifference or incomparability) with respect to each couple of alternatives. The acceptance of incomparability of alternatives — a situation that may occur when insufficient or conflicting information on two alternatives is available — is a main characteristic of the French school. The final outcome of this procedure is a set of alternatives which is not necessarily ordered in a linear way. It may well be so that two (or more) incomparable alternatives represent the best decision at the present state of the decision process, requiring from the decision maker further contemplation and research before he or she may decide on the ultimate course of action.

In the following section, we illustrate shortly the motivation to use of multi-criteria decision analysis in decisions pertaining to site restoration, and we conclude in Section 3 by pointing at some major challenges for future research and applications.

2 Multi-criteria analysis in site restoration: the motivation

Decision problems with respect to site restoration almost naturally require the integration of a variety of radiological, social and economical criteria. In a recent publication on the cleaning up of contaminated sites drafted by a consultants meeting at IAEA, this multi-criteria reality is fully recognized and illustrated throughout the document [IAEA95].

The decision to implement the clean-up of a contaminated area has to meet two requirements:
• justification and
• optimization.

For both requirements, the multi-criteria nature of the decision is constantly illustrated. We for instance cite:

"... the optimum level of intervention should be established by balancing the value of the avertable individual doses to the population by the clean-up against the clean-up costs, taking into account any other relevant factors."

Furthermore, it is stated that:

"Justification decisions in the context of clean-up will often be very complex, and could involve factors such as non-radiological risks and environmental effects, economic costs and benefits, and a wide range of social and political factors, as well as the radiological risks."

Although the multi-criteria nature of the problem is obvious, the consultants have failed to point at the technique of multi-criteria decision analysis.
3 Nuclear multi-criteria decisions: the challenges

Nuclear decision problems offer unique opportunities to the application and development of multi-criteria decision analysis techniques [Wal94c]. Some of the major challenges are the following issues.

The advanced use of computers

User-friendly graphical and interactive computer-interfaces have been widely recognized as an important tool in structuring and analysing the decision problem. Well-known examples are the VISA (Visual Interactive Sensitivity Analysis) Software from Belton, and the Promethee Software from Brans, from the Anglo-Saxon and French school, respectively. Much work however remains to be done in the representation and manipulation of data, including for instance automatic decision report generation and the use of the latest network technology or video-conferencing facilities.

Uncertainty modelling

In the field of clean-up actions or site remediation, decisions relate to the reduction of dose in the region of stochastic health effects. Existing models (Bayesian models for instance, or utility theory) still lack application in this domain. The recent development of fuzzy set theory also calls for our attention, taking into account its suitability to model decisions hampered with imprecision. Currently, decision analysis techniques are being developed at SCK•CEN in collaboration with Gent University, using more or less advanced elements of fuzzy set theory [Wal94a,Wal94b,Wal95].

Group decision making

The decision making authority in nuclear decision problems rarely is one single person. Instead, a number of people often having opposite or conflicting views on the decision may contribute to the process. Research opportunities in this respect are plenty: the use of networked computers, the application of the theory of social choice, or the development of group decision procedures. Currently, software is being prototyped at SCK•CEN to meet the above challenges. The software has been tested in a number of small examples outside the nuclear domain (personnel selection). The software makes use of the research center's computer network, has an advanced graphical user interface, and incorporates a new uncertainty modelling technique based on fuzzy set theory.

It allows up to ten decision makers in the process, and has some basic built-in communication facilities.

4 Conclusion

This paper puts the technique of multi-criteria decision analysis into perspective, without going into technical details or without giving demonstration. Its use for decision problems in the site restoration domain is motivated, and some of the nowadays challenges of the field are described.
5 Bibliography


Effect of AFCF on the soil-plant transfer of $^{134}$Cs

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Abstract

The possible use of AFCF (Ammonium-Ferric-hexaCyano-Ferrate) as a countermeasure for radiocaesium soil-to-plant transfer is evaluated. On a sandy agricultural soil, AFCF application doses of 10 and 100 g AFCF m$^{-2}$ reduced the radiocaesium transfer to ryegrass with a factor of 25 and 225, respectively, without affecting plant growth. Even additions of 1 g AFCF m$^{-2}$ resulted in a fourfold reduction of the radiocaesium transfer factor. Additions of less than 1 g AFCF m$^{-2}$ are hardly effective in reducing radiocaesium transfer to ryegrass. Depending on the soil radiocaesium contamination level, the cost of AFCF application can be out-levelled by the benefits due to decreased averted dose. The decrease in averted dose when people are consuming less contaminated food due to AFCF application, was assessed using a simple cost-benefit model. For a loamy soil with an intrinsically higher radiocaesium fixation capacity than a sandy soil, AFCF additions of 10 g m$^{-2}$ only reduced transfer with a factor of 3, which is on this soil type only as effective as ploughing in reducing transfer to ryegrass. AFCF application should thus not be recommended as countermeasure on loamy soil.

1 Introduction

When looking for substances to reduce the uptake of radiocaesium by domestic animals and to enhance its elimination from the body, AFCF [(Ammonium-ferric-hexacyano-ferrate(II), NH$_4$Fe(III)Fe(II)(CN)$_6$] came out as being very effective [1, 2, 3]. When given at a rate of 2 g per day, AFCF reduced the radiocaesium content in milk and meat of cow with 80-90 % [4].

The use of AFCF as a countermeasure for radiocaesium uptake by domestic animals, leaves us with at least four major topics to investigate: 1) What is the long-term availability of radiocaesium to the plants when contaminated faeces, containing AFCF is used as manure? 2) Will AFCF (present in the manure) affect plant growth? 3) Is AFCF also effective in reducing the soil-to-plant radiocaesium transfer? 4) In case AFCF addition reduces transfer, is its application also cost-effective? This article will deal with the last three questions.

2 Materials and Methods

Cs-134 and AFCF (Giese Salt, containing 60-65% AFCF and 35-40% NH$_4$Cl) were used throughout the experiments.

For the transfer studies in greenhouse an Orthic Podzol and an Orthic Luvisol (so called sandy and loamy soil) were used. For the experiment on sandy soil, moist (Field capacity, FC) soil was transferred to rectangular shaped darkened containers. Italian ryegrass (Lolium multiflorum) seed was spread evenly over the soil and was covered with a 0.4 cm moist soil layer, homogeneously contaminated with 1 MBq $^{134}$Cs. Seven different concentrations of AFCF (0, 0.03, 0.1, 0.3, 1, 10 and 100 g m$^{-2}$) were evenly sprayed over the soil surface.
All treatments were set up with 4 replicates. Ryegrass was harvested every two to three weeks, depending on growth. Dry weight was determined and activity measured by γ-counting (minaxi, auto-gamma 5000 series-gamma counter). The transfer factors are defined as (Bq g⁻¹ dry plant material)/(Bq g⁻¹ dry soil). Over a period of 1 year, 19 cuts were harvested. For the transfer experiments on loamy soil, moist (FC) soil was transferred to 1 L containers. Ryegrass was sown and covered with a 0.4 cm ¹³⁴Cs contaminated layer. Four concentrations of AFCF were applied on top of the contaminated soil (0, 1, 3, 10 g m⁻²). To simulate the effect of ploughing on transfer, ¹³⁴Cs was homogeneously mixed through the profile and 1 g AFCF m⁻² was applied superficially, in a fifth treatment. All seven treatments were set up with 4 replicates. Plants were treated and data were analysed as in the experiments on sandy soil. 14 cuts were harvested.

3 Results

3.1 Experiment on sandy soil

AFCF did not affect ryegrass growth (results not shown). The TF for the control (1.869±0.946) (Figure 1) is in agreement with literature data [5,6]. Levels of 0.1 g m⁻¹ AFCF and higher did significantly reduce the soil-to-plant transfer of ¹³⁴Cs: by a factor of 4 when 1 g AFCF m⁻² was applied and by a factor of 25 and 225, with 10 and 100 g AFCF m⁻², respectively, resulting in transfer factors of 0.069±0.036 and 0.008±0.007.

![Graph showing the effect of AFCF concentrations on the transfer factor (TF) for ¹³⁴Cs in sandy soil.](image)

Fig. 1: Effect of AFCF additions on ¹³⁴Cs-transfer to ryegrass grown on sandy soil. Numbers in bars represent % TF compared to the control. TF with different letters are significantly different at the 5 % level.
Regression analysis was performed to quantify the reduction in radiocaesium transfer with AFCF dose rate. When the reduction in transfer with AFCF treatment \[(TF \text{ control} - TF \text{ treatment})/TF \text{ control}\] was regressed against AFCF application rate, no significant regression was found. Regression was significant, however, when using an equation of the form

\[
TF\text{-reduction} = (1-e^{-B \cdot AFCF})
\]  
(Eq. 1)

with 'B' being a coefficient and 'AFCF' the amount of AFCF applied in g m\(^{-2}\). When AFCF→∞ g m\(^{-2}\), TF-reduction is maximal and thus 1. 'B' is the rate with which every additional AFCF dose will decrease TF. Eq. 1 shows that AFCF reduces transfer, but the effect on TF-reduction with every additional AFCF decreases exponentially. By fitting Eq. 1 to our data, the following regression equation was obtained:

\[
TF\text{-reduction} = (1-e^{-2.27960.111(AFCF)})
\]  
(Eq. 2)

Figure 2 shows the agreement between experimental and predicted data.

\[\text{TF reduction} = (1-\exp(-2.279\cdot AFCF))\]

3.2 Experiment on loamy soil

As for the experiment on sandy soil, AFCF did not affect ryegrass yield (results not shown). When considering the first 4 treatments (\(^{134}\text{Cs} \) applied superficially), the results obtained for the loamy soil (Figure 3) differ from those obtained for the sandy soil on two major points: 1) The TF of the control is only 0.117±0.012 (Bq g\(^{-1}\) plant)/(Bq g\(^{-1}\) soil). This value is in perfect agreement with the TF recorded in a separate study for ryegrass on loamy soil under field conditions (0.114±0.080, [6]). 2) Only AFCF additions of 3 g AFCF m\(^{-2}\) and higher result in a significant TF decrease: with 3 and 10 g AFCF m\(^{-2}\), TF was decreased with 28 % (factor ±1.5) and 64 % (factor ±3, respectively. Ploughing had a similar effect on TF as AFCF additions of 10 g m\(^{-2}\). When comparing treatments 2 and 5 (for both treatments, 1 g AFCF m\(^{-2}\) applied superficially and \(^{134}\text{Cs} \) added superficially or mixed) the effect of ploughing on TF can be estimated. Under our experimental conditions, ploughing reduced TF with 62 %.
AFCF and Cs-134 added superficially
AFCF: superficially, Cs-134: mixed

Fig. 3: Effect of AFCF additions and 134Cs application mode on 134Cs-transfer to ryegrass grown on loamy soil. Numbers in bars represent % TF compared to the control. TF with different letters are significantly different at the 5 % level.

4 Conclusions

Only a few studies have been conducted to investigate the effect of AFCF addition on the radiocaesium transfer to plants. Segal [7] reports vaguely about studies done in Russia about the effect of amendments (including AFCF) on transfer to plants. There was little evidence that these materials were effective in practice in agricultural systems. Jones *et al.* [8] tested different soil amendments, including AFCF, to reduce radiocaesium plant uptake.

The radiocaesium content of the natural upland vegetation of Wales and Cumbria was only reduced to a limited extend (20 %) when the highest AFCF dose (2 g m$^{-2}$) was applied. Its effect declined and radiocaesium uptake became more variable after this first year, for unknown reasons. Hove *et al.* [9] describing experiments done in Ukraine and Belarus, reported that AFCF (unspecified quantity) in cattle manure, reduced radiocaesium levels in lupin by 63 % and in grass with 50 %, compared to the corresponding controls.

In our experiment on a sandy soil, AFCF is undoubtedly effective in reducing 134Cs transfer to ryegrass without affecting its growth. Among some of the countermeasures applied after the Chernobyl accident, ploughing and application of mineral fertilizers yielded at most a 4-fold reduction in transfer and results were often inconsistent [5, 10, 11] . This 4-fold reduction in TF was already obtained with 1 g AFCF m$^{-2}$. One hundred g AFCF m$^{-2}$ even resulted in a 225 fold reduction of the transfer factor. The relatively low cost (±2000 Bfr kg$^{-1}$), its easy application (an AFCF solution can be sprayed making use of the machinery for applying pesticides), its non-toxicity for plants and its long-term effectiveness (even after 1 year AFCF was still effective in reducing the TF) make AFCF a seemingly ideal countermeasure on soils with a low affinity for Cs.
Since the transfer reduction with AFCF dose is quantifiable in a mathematical equation, transfer reductions (and consequently dose reductions) can be estimated for every AFCF dose within the range tested. This can be a useful tool for cost-benefit studies of remedial actions.

One could attempt to do a simple cost-benefit analysis on the use of AFCF e.g. for Kopachy (Ukraine) with radiocaesium contamination levels of 1 (and >) MBq m\(^{-2}\). In the following simplified example, we consider the reduction in averted dose from the consumption of milk from cow grazing on AFCF treated pastures compared to cows grazing on non-treated pastures. If the daily consumption of milk of one person is 0.5 L, this mounts to 183 L a\(^{-1}\). If a cow produces 10 L d\(^{-1}\) (a reasonable figure for the region considered), she has to be fed in total 18 days, and with a daily need of 15 kg (dw) ryegrass a day (she is on a ryegrass diet!) she will consume 270 kg ryegrass. Since the ryegrass yield is about 2 Tons ha\(^{-1}\) a\(^{-1}\), 0.13 ha is needed. If this 0.13 ha has to be treated with AFCF (cost 2000 Bfr kg\(^{-1}\)), the cost of AFCF treatment is 2600 Bfr when 10 kg ha\(^{-1}\) (1 g m\(^{-2}\)) are applied and 26000 Bfr when 100 kg ha\(^{-1}\) (10 g m\(^{-2}\)) are applied.

From table 1 we can infer that application of AFCF under mentioned conditions of contamination level and consumption behaviour is certainly cost-effective. It can be calculated that, again for the conditions considered, the minimal contamination level should be 0.622 MBq m\(^{-2}\) for costs and benefits to break even when 10 kg AFCF ha\(^{-1}\) is applied. Few comments are in order. First, compensation cost for annually averted dose for Belgium was used in the calculations since these data are not available for the CIS-situation. Secondly, the cost-effectiveness of AFCF application is possibly larger since the effect of AFCF may extent over a longer period than a year. Furthermore, the reduced dose, due to the consumption of the meat of animals feeding on AFCF treated pastures, is not considered. On the other hand, in the experimental set-up AFCF is immediately applied after radiocaesium contamination. In the real situation there might be a time-lapse between both and AFCF might no be so effective in decreasing 'aged' radiocaesium uptake. Thirdly, the cost-effectiveness of AFCF application to pastures should be compared to the cost-effectiveness of the administration of AFCF directly to the animals (lick stones) and the practicability feasibility of both methods. Finally, apart from cost-benefit parameters, socio-political and cultural parameters should be taken into account (partly the topic of the speech by Bartel Van De Walle). Often, however, these other parameters are difficult to get access to and are not easy to incorporate in the validation exercise.
Table 1: Effect of AFCF application on adverted dose and compensation cost considering situation for Kopachy (contamination level 1 MBq m$^{-2}$=5000 Bq kg$^{-1}$)

<table>
<thead>
<tr>
<th>AFCF applied (kg ha$^{-1}$)</th>
<th>0</th>
<th>10</th>
<th>100</th>
</tr>
</thead>
<tbody>
<tr>
<td>TF for ryegrass on sandy soil (Bq g$^{-1}$ dw/Bq g$^{-1}$ soil)</td>
<td>1.8</td>
<td>0.45</td>
<td>0.07</td>
</tr>
<tr>
<td>Contamination level in ryegrass (Bq kg$^{-1}$) considering soil contamination of 5000 Bq kg$^{-1}$</td>
<td>9000</td>
<td>2243</td>
<td>343</td>
</tr>
<tr>
<td>Daily intake cow (Bq d$^{-1}$) cow consumes 15 kg dried grass</td>
<td>135000</td>
<td>33645</td>
<td>5145</td>
</tr>
<tr>
<td>Contamination level milk (Bq L$^{-1}$) with TF to milk 7*10$^{-3}$ (Bq L$^{-1}$/daily intake in Bq)</td>
<td>943</td>
<td>235</td>
<td>36</td>
</tr>
<tr>
<td>Annual intake (Bq a$^{-1}$) considering milk consumption of 0.5 L d$^{-1}$</td>
<td>172071</td>
<td>42881</td>
<td>6570</td>
</tr>
<tr>
<td>Committed dose (mSv a$^{-1}$) with committed dose for ingestion of $^{134}$Cs: 1.3*10$^{-3}$ Sv Bq$^{-1}$</td>
<td>2.231</td>
<td>0.560</td>
<td>0.085</td>
</tr>
<tr>
<td>Difference in committed dose (mSv a$^{-1}$)</td>
<td>1.671</td>
<td>2.146</td>
<td></td>
</tr>
<tr>
<td>Benefits (Bfr a$^{-1}$) with compensation of:</td>
<td>4178</td>
<td>21460</td>
<td></td>
</tr>
<tr>
<td>1000 Bfr mSv$^{-1}$ for 0-1 mSv</td>
<td>2500 Bfr mSv$^{-1}$ for 1-2 mSv</td>
<td>10000 Bfr mSv$^{-1}$ for 2-5 mSv</td>
<td>25000 Bfr mSv$^{-1}$ for 5-10 mSv</td>
</tr>
</tbody>
</table>

On a loamy soil AFCF is less effective in reducing the transfer. Undoubtedly so because the loamy soil has a high radiocaesium fixation capacity, which is related with its micaceous clay content (qualitative XRD-analysis: results not shown). For the control treatments, the $^{134}$Cs transfer factor for the loamy soil is 16 times lower than for the sandy soil. Only when 10 g AFCF m$^{-2}$ was applied to the sandy soil, was its transfer comparable to the transfer on the loamy soil without AFCF addition. Mixing $^{134}$Cs through the profile of the loamy soil resulted in a three fold reduction of the transfer. Similar reductions were found after shallow ploughing [10]. This reduction in TF was comparable with the decrease in TF when 10 g AFCF m$^{-2}$ was applied. Therefore, ploughing is a more advisable than AFCF application as a countermeasure for $^{134}$Cs uptake by plants on a loamy soil.
5 References


The use of zeolites in countermeasure strategy: predicted versus observed effects
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Abstract

Among the possible remedial actions to reduce the soil-to-plant transfer of radiocaesium and radiostrontium, the addition of clay minerals or zeolites aims at permanently increasing the radionuclide fraction adsorbed on the solid phase, thus lowering the radionuclide solution level available for root uptake. The traditional procedure to test the effectiveness of such amendments is rather empirical: amendments are applied to the soil and the effect is expressed in terms of the change of the transfer factor, defined as the ratio of Bq/kg plant and soil. This procedure yields no insight in the process(es) responsible for the observed effect and allows no predictions for other scenarios. Based on the knowledge that soil-to-plant transfer is essentially dependent on soil type, the utilisation of soil amendments was investigated by quantitatively addressing the physico-chemical parameters which govern the solid/liquid partitioning of the radionuclides in soils and soil amendments. It is shown that this approach allows to identify the potential soil/amendment combinations and to estimate the effects to be expected.

1 Introduction

Consumption of food products grown on soils contaminated with radiocaesium ($^{137}$Cs) and/or radiostrontium ($^{90}$Sr) is known to contribute significantly to the total radiation dose of the population living in or close to the affected regions [1,2]. Consequently, reducing the soil-to-plant transfer of these radionuclides is one of the most effective means of mitigating the consequences of such a contamination. One of the possible remedial actions is the application of soil amendments such as zeolites. Such amendments aim permanently at enhancing the radionuclide fraction adsorbed on the solid phase, thus reducing the soil solution activity concentrations of the radionuclides available to the plant roots. Both positive and negative or no effects have been reported in literature [2-8]. There may be several reasons to explain these diverging results, such as the difficulties encountered with mixing the contaminated soil and soil amendment, the influence of the soil amendment on the ionic composition of soil and soil solution, ... [3-5]. However, the main reason is undoubtedly the lack of sufficient understanding of the different processes which govern the soil-to-plant transfer of a radionuclide, and which are acting both at the soil physico-chemical and at the plant physiological level.
The traditional procedure to test the effectiveness of soil amendments is based on the measurement of the combined effect of both types of processes: soil amendments are applied to the soil and the effect is quantified by measuring the change of the radionuclide soil-to-plant transfer factor (TF), defined as the ratio of $\text{Bq kg}^{-1} \text{dry plant material}$ and $\text{Bq kg}^{-1} \text{dry soil}$ or $\text{Bq m}^{-2} \text{contaminated soil surface}$.

In this paper, a methodology is presented which allows to predict the effect of zeolite amendments on the soil-to-plant transfer of radiocaesium and radiostrontium. It is based on a quantitative understanding of the different factors which govern the solid/liquid distribution behaviour of these radionuclides in soils and zeolites [7-14].

## 2 Theoretical aspects

The most important conclusion of this quantitative understanding regarding this discussion is that the potential of an adsorbent (e.g. soil or zeolite) to absorb a radionuclide is governed by the product of the capacity of the ion exchange sites involved in the adsorption process times the selectivity coefficient of the radionuclide with relation to the most important competitive cation on these ion exchange sites. In the case of radiocaesium and radiostrontium, K and Ca are the most important competitive cations, respectively.

The capacity times selectivity product has been shown to be a fairly constant value which can be identified with the $K_d^{\text{Cs}} N_K$ or $K_d^{\text{Sr}} N_{\text{Ca}}$ product if $K_d^{\text{Cs}}$ or $K_d^{\text{Sr}}$ are measured at sufficiently high solution concentrations of K or Ca, respectively ($N_K$ or $N_{\text{Ca}}$, 0.01 equiv. dm$^{-3}$) [8,13,14]. The product has been referred to as the radionuclide (radiocaesium or radiostrontium) adsorption potential (RAP, equiv. kg$^{-1}$), and is denoted as $[\text{Cs}^{d} N_K]$ and $[\text{Sr}^{d} N_{\text{Ca}}]$, respectively. For soils, the Sr to Ca selectivity coefficient is about unity, and hence the soil's cation exchange capacity (CEC) is a good estimate of the $[\text{Sr}^{d} N_{\text{Ca}}]$ value [11]. The practical significance of the constancy of the capacity times selectivity product is that $K_d^{\text{Cs}}$ or $K_d^{\text{Sr}}$ values are inversely proportional to the solution concentration of the respective competitive cations.

The addition of a sorption competitive zeolite to a soil leads to an increase of its overall radionuclide adsorption potential, resulting in (a) a decrease of the liquid phase radionuclide levels and (b) in a shift of the fraction of reversibly retained radionuclide, associated with the soil, towards the zeolite. The extent of the effect will depend on the dose and the relative values of the radionuclide adsorption potentials of soil and zeolite. The effect can be written as:

$$\text{effect} = \frac{\text{RAP}_s \cdot m_s + \text{RAP}_a \cdot m_a}{\text{RAP}_s \cdot m_s} = 1 + \frac{\text{RAP}_a \cdot m_a}{\text{RAP}_s \cdot m_s}$$  \hspace{1cm} (Eq. 1)

in which $m$ refers to the masses and the subscripts $s$ and $a$ to soil and amendment. First, it is seen that at a dose of 1 wt.% we need a two order of magnitude difference in RAP values for obtaining a two-fold increase of the RAP value of the amended soil. Depending on whether the contamination is located at the surface or ploughed within the 25 cm ploughing layer, a 1 wt.% dose corresponds with some 4 to 8 and 20 to 40 tons ha$^{-1}$, respectively (depending on the bulk density of the soil). Secondly, equation (1) shows that the effect is linear in the dose of the zeolite added.
3 Predictions of the effectiveness of zeolite amendments

3.1 RAP values for soils and zeolites

Ranges of $[K_{d}^{Cs}.N_{K}]$ and $[K_{d}^{Sr}.N_{Ca}]$ values for the major different soil types [7-11,14] and for some reference zeolites (both natural and synthetic) [12,13] are summarized in table 1. These data warrant various comments. $[K_{d}^{Cs}.N_{K}]$ values for peaty and sandy soils vary between 0.01 to 0.30 equiv. kg$^{-1}$. These soil types are characterized by high radiocaesium soil-to-plant transfer factors and are the most vulnerable regarding radiocaesium contamination [7]. $[K_{d}^{Sr}.N_{Ca}]$ values for zeolites vary in the range of 15 to 120 equiv. kg$^{-1}$, depending on the type of zeolite and on the K/Ca ratio in the solution. Especially for clinoptilolite and mordenite, which are known for their pronounced Cs selectivity, $[K_{d}^{Cs}.N_{K}]$ values exceed those of the sandy and peaty soils by some two to three orders of magnitude. These zeolites are therefore expected to be effective soil amendments for radiocaesium, even at 1 % doses. These values apply for reference materials, and lower $[K_{d}^{Cs}.N_{K}]$ values are expected, and measured, for less pure zeolites.

Table 1: Ranges of $[K_{d}^{Cs}.N_{K}]$ and $[K_{d}^{Sr}.N_{Ca}]$ values (equiv. kg$^{-1}$) for the major different soil types and some reference zeolites.

<table>
<thead>
<tr>
<th>soil or zeolite type</th>
<th>$[K_{d}^{Cs}.N_{K}]$</th>
<th>$[K_{d}^{Sr}.N_{Ca}]$</th>
</tr>
</thead>
<tbody>
<tr>
<td>sandy soils</td>
<td>0.10 - 0.30</td>
<td>0.02 - 0.15</td>
</tr>
<tr>
<td>peaty soils</td>
<td>0.01 - 0.15</td>
<td>0.25 - 1.00</td>
</tr>
<tr>
<td>loamy soils</td>
<td>1.50 - 3.50</td>
<td>0.15 - 0.25</td>
</tr>
<tr>
<td>clayey soils</td>
<td>3.50 - 5.00</td>
<td>0.25 - 0.40</td>
</tr>
<tr>
<td>clinoptilolite (Hector, California)</td>
<td>30 - 55</td>
<td>3.0 - 15.0</td>
</tr>
<tr>
<td>mordenite (Akzo Nobel)</td>
<td>60 - 120</td>
<td>1.0 - 5.0</td>
</tr>
<tr>
<td>mordenite (Karpats, Ukraine)</td>
<td>60 - 120</td>
<td>1.0 - 5.0</td>
</tr>
<tr>
<td>zeolite 5A (Union Carbide)</td>
<td>8 - 15</td>
<td>20.0 - 28.0</td>
</tr>
</tbody>
</table>

The lowest $[K_{d}^{Sr}.N_{Ca}]$ values are found for sandy soils, which are the most vulnerable soils regarding radiostrontium contamination [7]. Zeolite 5A - a synthetic zeolite in the Ca-form with an Al/Si ratio of 1 - shows a high $[K_{d}^{Sr}.N_{Ca}]$ value which is relatively independent of the K/Ca ratio in the solution. This zeolite can therefore be expected to be an effective soil amendment in sandy soils contaminated with radiostrontium. For mordenite and clinoptilolite, $[K_{d}^{Sr}.N_{Ca}]$ values are strongly dependent on the K/Ca ratio in the solution, which is of course due to their pronounced K selectivity. For K/Ca ratios occurring under soil conditions, these zeolites are not expected to be effective soil amendments for radiostrontium.
3.2 Predicted versus observed effects

The characterizations discussed in the previous sections allow predictions of the effects to be expected, according to equation (1). The question of course is whether these predictions agree with observed effects, since soil-to-plant transfer integrates the effects of processes acting at both the soil physico-chemical and the plant physiological level. To answer this question, a potted soil experiment was performed. 1 wt.% doses of natural clinoptilolite (Hector, California), synthetic mordenite (Akzo Nobel; Na-form) and zeolite 5A (Union Carbide) were thoroughly mixed with a sandy soil which had been contaminated priorily with $^{137}$Cs and $^{85}$Sr. Spinach (Spinacia oleracea L.) was grown on the different systems (5 replicates) and $^{137}$Cs and $^{85}$Sr activities were measured in the shoots of one month old spinach plants. Results are presented in table 2, together with the predictions based on equation (1). For $^{137}$Cs, TF’s are reduced by a factor of 3 to 4.6, and predicted effects coincide reasonably well with observed effects. The effect is higher for mordenite, which shows a higher RAP value compared to clinoptilolite (table 1). For $^{85}$Sr, TF’s are reduced by a factor of 25, and the predicted effect also agrees reasonably well with the observed effect.

Table 2: Soil-to-plant transfer factors for $^{137}$Cs and $^{85}$Sr (ratio of Bq kg$^{-1}$ dry material and dry soil), observed and predicted effects. Data refer to averages and standard errors (in brackets) of five replicates. $[K_{d}^{Cs}, N_{K}]$ and $[K_{d}^{Sr}, N_{Ca}]$ values for the sandy soil are 0.15 and 0.018 equiv. kg$^{-1}$, respectively.

<table>
<thead>
<tr>
<th>treatment</th>
<th>TF (kg soil/kg plant)</th>
<th>observed ratio of TF</th>
<th>predicted ratio of TF</th>
</tr>
</thead>
<tbody>
<tr>
<td>radiocaesium</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>controle</td>
<td>0.75 (0.09)</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>mordenite (1%)</td>
<td>0.16 (0.04)</td>
<td>4.6</td>
<td>9.3</td>
</tr>
<tr>
<td>clinoptilolite (1%)</td>
<td>0.25 (0.04)</td>
<td>3.0</td>
<td>4.3</td>
</tr>
<tr>
<td>radiostrontium</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>controle</td>
<td>35.1 (6.4)</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>zeolite 5A (1%)</td>
<td>1.4 (0.1)</td>
<td>25.0</td>
<td>16.3</td>
</tr>
</tbody>
</table>

The predictive value of this approach is also confirmed when considering the data of Nishita and Haug (1972) [4]. These authors measured the effect of 1 wt.% additions of Ca-treated clinoptilolite (Hector, California) on the soil-to-plant transfer of $^{137}$Cs and $^{90}$Sr in clover (Trifolium repens L.) grown on a sandy-loamy soil. They reported that the $^{90}$Sr uptake was reduced with 60 to 70 % relative to the control (i.e. a reduction with a factor 2.5 to 3.3), whilst the application of Ca-treated clinoptilolite had but a very small effect on the $^{137}$Cs content of the clover. The CEC value, which is a good estimate of $[K_{d}^{Sr}, N_{Ca}]$, was 0.076 equiv. kg$^{-1}$. Taking a $[K_{d}^{Sr}, N_{Ca}]$ value of 15 equiv. kg$^{-1}$ for Ca-treated clinoptilolite (table 1), we would predict a reduction of TF with a factor of 3.3, which corresponds very well with the observed effects. In 1972, no data were available as to the $[K_{d}^{Cs}, N_{K}]$ value of the soil. Taking the value of 2.3 equiv. kg$^{-1}$ which is reported for sandy-loamy soils from Belgium [8], and a $[K_{d}^{Cs}, N_{K}]$ value of 50 equiv. kg$^{-1}$ (table 1), we would predict a reduction of TF with a factor of 1.2. Again, this
prediction is in reasonable agreement with the reported small effect on the $^{137}\text{Cs}$ content of clover. Moreover, it shows that the successful use of zeolite amendments - at reasonable doses of 1 wt.% - is restricted to soils with low [$K_d$] values, as could already be inferred from the data presented in table 1.

4 Conclusions

It is shown that a characterization of soil and soil amendment in terms of their radionuclide adsorption potentials allows to make reliable predictions on the effect to be expected, thus avoiding the costly and time-consuming field campaigns traditionally performed to test the effectiveness of soil amendments. Though, the predictive method presented here is a strong simplification, since the processes acting at the plant physiological level and the effect of other potentially competitive cations are neglected. The disappointing results sometimes reported in literature may partly be attributed to wrong combinations of soil and soil amendment. Finally, it should be mentioned that also other aspects should be examined when considering the application of e.g. zeolites as a countermeasure, such as potential side effects (nutrient status of the soil), long term effects (degradation of zeolites) and economical aspects (cost-benefit).

5 References

[7] Rauret, G. and Firsakova, S. (Eds) Final report of the Experimental Collaboration Project n°2 to the Commission of the European Communities: The transfer of radionuclides through the terrestrial environment to agricultural products, including the evaluation of agro-chemical practices. EURO-report EUR 16528EN, 1995


How short rotation forest crops can be used for sustainable remediation of contaminated areas

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Abstract

In large territories of the CIS, it becomes obvious from the factual consequences of the Chernobyl environmental contamination that no successful remediation actions can be achieved without considering realistic technical and economical issues. In these conditions, the "Short Rotation Forestry" concept for energy purposes is proposed as an alternative and integrated approach for the recovery of agricultural practices on waste farm land. This corrective option will be examined with respect to its ecological, economical and social relevancy. Different aspects of the culture in contaminated areas and of biofuel production remain to be investigated, developed and validated in the light of radiation protection criteria. In particular, attention will be drawn on the opportunity of this new concept to be integrated in the development of "Site Remediation" research activities at SCK•CEN.

1 Introduction

Following the Chernobyl accident, many thousands of square kilometres have been severely contaminated in the CIS. The application of simple clean-up operations leading to a rapid return of such a large area to its normal state, is unrealistic and would, in addition, generate an enormous amount of radioactive waste. Consequently, an important part of the contaminated agricultural land may be left deserted for many years, despite its agricultural potential remaining intact.

When agricultural perspectives must be abandoned in such territories because of irretrievably high levels in food products or because of economically and technically non-realistic corrective options, an increasing interest arises in developing more integrated and ecologically-based approaches. In this regard, a realistic strategy is needed and must be performed by using the most suitable combination of efficient restoration techniques.

The selection and application of alternative practices can offer another solution for land recovery close to normal situations. Bioalternative cultures, already tested in the traditional agriculture, offer a possibility for adaptation to local conditions of the contaminated areas.

In this regard, Short Rotation Forestry for energy purposes is put forward as a particularly appealing and innovative bioalternative for remediation of contaminated waste farm land with restricted uses. Willow species can act as combined vegetation filter and biomass producer.

By transposing the existing information of "non-nuclear" environmental technologies in the radiation protection field, the SRC concept validation opens the way to a better integration of more effective and acceptable solutions in the "Contaminated Site Remediation". When dealing with the application of this corrective option in
contaminated areas, different aspects of the culture remain, however, to be examined considering the evaluation of their ecological, economical and social consistency in the light of radiation protection criteria.

2 The "Short Rotation Copse" (SRC) concept

The Short Rotation Forestry concept lies in cultivating fast growing plants for energy purposes. Willow species (Salix spp.) are good candidates among others (poplar, alder, ...) for biomass production. The copse vegetation is intensively managed and harvested for biomass in a three to five year cutting cycle and a 22-25 year rotation (fig.1). The produced biomass can be valorized by pyrolysis and/or gasification processing and generate a energy.

Fig. 1: The SRC chain as conceived in common agriculture (modified from J.M. Jossart-ECOP.UCL: personal communication)

Faced with the complexity of the radioecological situation in contaminated areas, the fast growing copse-vegetation concept represents interesting properties which could fulfill important basic requirements, underlined by the following assets of the culture:

- High density perennial with following characteristics:
  - High factor of soil protection: solution against radionuclide mobility (limiting the risk of secondary contamination by restraining erosion of the contaminated surface by wind or water).
  - The green vegetation limits considerably the risk of fires and activity resuspension.
  - Its cutting propagation and perennial character allow limited maintenance (limited dose to workers).
- Compatibility for integration in a dynamic management for large territories; strategic zone for filtering trials (watershed outlet); draining sludge repositories; filtering quickset edge around waste burials.

**Economical assets**
- Possibility of economical valorization of products from agricultural waste land (production of biomass for energy purposes, pulp, paper).
- Possibility of mechanization at all stages of the culture
- In some regions, copse might provide woody biomass for energy purposes to cover up for fuel wood requirements in attendance of finding solutions for the use of forests which have been too heavily contaminated by the Chernobyl accident.
- Favourable energy balance (superior to bio-diesel ex rapeseed, bio-ethanol ex beet...)

**Social assets**
- Technically easily applicable culture for farmers/workers without experience in the field
- Integration of the local population in remediation action: creation of jobs.
- Non-foodstuff production (psychological aspects).

### 3 Researches and developments needed

The SRC is like the controlled space-time miniaturization of biomass production compared to common forest stands. Forest ecosystem contribute efficiently to the stabilization of the contamination.

So far the forestation of contaminated agricultural fields has been recognised as a pertinent option, particularly in heavily contaminated areas where soils are not suitable anymore for agricultural production. The predictions of radionuclides transfer in forest stands demonstrate, however, that accumulation in usable standing wood is continuous and relatively important (10-15% of the total radionuclides inventory).

With the use of high density vegetations as SRC as an alternative, soil protection is ensured and the risk of secondary pollution due to forest fires is firmly reduced; moreover, the regular harvest of copse biomass at short intervals (3-4 years) is consistent with an expected reduction of the woody matter contamination (fig.2). This would also comply with the mitigation of exposure risks during culturing, subsequent handling and combustion operations.
% of initial inventory accumulated in biomass

10-15%

Forest

3 years cuttings cycle

vSRC

0

Time (year)

30

Fig. 2: Comparison of the potential immobilisation of radiocaesium activity in the biomass generated by forest and vegetation-copse ecosystem, respectively.

Actually, knowledge on processes involved in uptake and partitioning of radionuclides in the SRC ecosystem is deficient. A primordial step in validating the SRC relevancy compared to forests, common agriculture or other bioalternatives, deals with the determination of the radionuclides’ cycling in the culture and the modelling of radionuclides output by harvest.

The benefit of the SRC option has to be evaluated and expressed not only in terms of dose reduction, but also in terms of economical benefit. Indeed, the cost-benefit analysis of the countermeasures already tested in CIS showed that the cost of the averted man.Sievert is very expensive. It is now proposed that the cost of the averted dose could be partially compensated by the additional economical profitability of alternative options.

The main economical issue of the SRC chain is to provide energy through biomass combustion. The economical competitiveness of this source of energy will depend on the availability of other biofuel energy sources as well as the on quantity and the form under which the energy can be used.

Different techniques are likely to suit the combustion of contaminated wood: to burn it in big heating plants or to convert the wood to biogas in a gasification incinerator. The last option involves less combustion units and represents opportunities for better adaptation to localised contaminations: limited transport, use of other wood waste, flexibility of the energy conversion process by electricity generation.
First calculations of the SRC chain profitability show that the cost price of wood corresponds more or less to the domestic fuel price. When using contaminated biomass, the profitability of the different combustion options remains, however, to be considered in function of the distribution of radionuclides during the respective processes and of the cost for the related radiological safety measures: smoke filtration, contaminated ash removal, waste treatment, ...

The social situation of people living in contaminated areas has also an important impact on the management routes of the SRC which can be implemented. The copse-vegetation culture allows for the participation of the local population even if a high mechanisation level is preferred for economical optimization. Acceptability of the culture, cultural habits, labour cost, mechanisation possibilities, availability of competitive energy sources, energetic production framework, ... are factors with large uncertainty. The consideration of such local economical criteria or social-traditional preferences may put forward the difficulties in attempting to reduce the cost of the converted dose with the application of new economy enhancing options like SRC.

4 Conclusions

Although a consensus on the implementation of a priorization of remediation techniques seems difficult, the selection and application of corrective actions in contaminated areas remain a key issue in large territories of Belarus, Ukraine and Russia.

In these conditions, minimizing the economic and social disruption is a priority in site remediation of extended agricultural areas, especially for agriculturally based communities. The SRC concept, as an alternative culture for energetic purposes, represents several ecological and technical properties that comply with such an integrated approach of the problem of site rehabilitation. Combining and comparing the results of radioecological, energetic, economical and radiological evaluation is essential in validating the sustainability of the technique.

In the framework of future "Site Remediation" research activities at SCK-CEN, the impact assessment study linked to the application of the SRC in contaminated areas will be also the opportunity to:

1. valorize experiences gained in determining radionuclides transfer and cycling in forest and agricultural lands, in modelling the radionuclides' transfer, ...
2. open the way for a multi-disciplinary collaboration and new developments and application, in decision helping technique, in GIS methods, in radiological impact modelling, in waste treatment, ...
Impact assessment of shallow land burial for low-level waste: modelling of the water flow and transport of radionuclides in the near-field

Jan Walravens, Geert Volckaert

Abstract

The Belgian concept for disposal of LLW consists of storage of waste drums into a concrete vault backfilled with a cementitious grout. The vault is placed above the water table and will be covered with a multilayer cap of clay, gravel and sandy materials. The SCK•CEN is charged with the long-term performance assessment of the disposal site. The main processes and parameters determining the radioactivity release from the site are identified. The determinant processes are the infiltration through the top cover and the sorption of waste on the backfill. The release of radioactivity from the site was modelled with the PORFLOW numerical code.

1 Introduction

An option for the disposal of low-level radioactive waste (LLW) consists of the emplacement and burial at the surface or at shallow depths. The waste can be placed in trenches, engineered structures, rock cavities and abandoned mines. A concept proposed in various countries for disposal of LLW consists of the disposal of waste drums into a concrete vault backfilled with a cementitious grout. The vault is placed above the ground water table and will be covered with a multi-layered cap of clay, gravel and sandy materials.

To ensure an acceptable low risk to the critical group of the population, reference limits and design criteria have to be acquired. SCK•CEN has experience with the long-term performance assessment of disposal sites. In this paper special attention will be paid to the transport of radionuclides out of an engineered facility for LLW and to how impact assessment can play a significant role in optimising the design. The main processes and parameters determining the radioactivity release from the site are identified.

2 General disposal concept of shallow land burial

Low-level waste arises from activities by nuclear power reactors, radioisotope manufacturers, nuclear fuel cycle facilities, hospitals, universities etc.. The waste contains generally short-lived radionuclides and very low concentrations of long lived radionuclides. Compaction, conditioning in concrete matrix and encapsulation in concrete packaging or steel drums are required to guarantee safe handling, transportation and disposal. Conditioning in cementitious grout will limit the mobility of the radioactive component and thus the transport to the environment.
The near surface disposal of LLW implies the construction of an engineered structure to isolate the waste from water and the human environment under controlled conditions and for a period of time long enough to allow the radioactivity to decay to an acceptable level. As the radioactive waste will mainly escape from the disposal facility as water dissolved species, the availability and movement of water has to be reduced. Therefore the vault will be placed above the water table and will be covered with a multi-layered drainage system. To limit the mobility of the waste a largely cementitious backfill will be cast around the waste containers.

The interstitial water of the cement pores provides a high pH environment in the repository which reduces the solubility of many radionuclides especially the actinides. Also the corrosion rate of the metal canisters is low at high pH. However, the properties of cement may evolve a.o. through calcium release by the infiltrating water. Again, this emphasises the importance of shielding the repository from inflowing water. A reference design coping with the principal requirements is shown in Fig. 1.

Fig. 1: Cross section of reference design

The waste package consists of a concrete waste form sealed in a steel container. The bottom of the vault is a permeable layer which permits the drainage of infiltrating water and which prevents flooding. A concrete cap covers the vault and serves to protect the vault from excessive infiltrating water because of its low permeability (hydraulic conductivity < 10^-10 m/s). Whereas its physical integrity cannot be guaranteed for more than a few hundreds of years. After the life time of the concrete cap, shielding task will rely on the imperviousness of the clay layer.

The use of geomembrane liners and clay covers is widespread in the containment of landfill sites. One advantage of using natural materials such as clay is that their long-term performance is assured, whereas the long-term behaviour of plastic membranes is not tested. In most cases a percolation rate of a few tens of litres/a/m² is allowed. If we consider a saturated layer whose flow through is controlled by Darcy's law a permeability coefficient less than 10^-9 m/s is required.
To protect the clay cap from environmental effects, it will be overlaid by an intrusion barrier (to prevent physical damage by vegetation or rodents) and a fertile top soil with vegetation (against rain fall erosion, frost penetration, wetting and drying cycles etc.). The water flowing off the clay cap will be carried from the repository by the drainage system.

3 Performance assessment

Since 1984 the SCK•CEN is involved in performance assessments of shallow land burial. Generic assessment assists in making decision between different possible disposal concepts and in predicting the safety of or in gaining acceptance for a particular disposal concept. Impact assessment studies aim to determine the behaviour and transport pathways of the radionuclides with use of the best available methods and information from field, laboratory and natural analogues:

- Knowledge of transport mechanisms of radionuclides
- Hydrological parameters (rainfall, infiltration rate, erosion of wind, ...)
- Hydraulic parameters of the site (porosity, hydraulic conductivity, suction potential, ...)
- Chemical composition of near-field (pH, Eh, T°, ...)
- Behaviour of the radionuclides (solubility limits, sorption behaviour, chemical reaction, decay, complexing reagents ...)

Considering the concept for the disposal of LLW into concrete vaults, as described above, two main conclusions resulted from previous impact assessments:

- For the majority of the radionuclides, advective transport seems to be the main mechanism for release out of the disposal facility. The radioactive flux shows linearity with the infiltration rate through the concrete vault.
- In case of poorly sorbed radionuclides such as tritium under aqueous form, diffusion through the walls of the vault and to the ground water table must be taken into account.

Because of the influence of the infiltration of water through the repository, the impact assessment of different design options was focused on the assessment of the performance of the drainage cover.

4 Performance of a multi-layer cover - a case study

In order to improve the performance of the general concept of Fig. 1, modifications were suggested:

- The operation of the drainage system cannot be guaranteed for the lifetime of the repository. Unless monitoring and maintenance is provided, the drainage system can fail through clogging. The alternative design will omit this weak point. The cover layers were lengthened so that off-flowing water cannot re-enter the vault.
- The clay cap will be divided and separated by a sand layer. The capillary break between these clay layers promotes the lateral drainage of any infiltration through the uppermost clay layer.
- To limit the lateral diffusion of tritium out of the vault, the walls will be surrounded by a gravel layer which will remain almost dry because of its low suction potential.
The alternative design is shown in Fig. 2.

![Fig. 2: Cross section of alternative design](image)

An infiltration analysis of both design options has been performed with use of the numerical PORFLOW code [Runchal, 1993]. The computer modelling of the flow in the multiple layer under unsaturated conditions contributes to understanding the hydraulic functioning of the structure. The main soil properties which control the hydraulic behaviour of the multi-layered structure are the water suction potential and the variation of permeability for water in function of saturation. Furthermore, climatological data such as precipitation and potential evapotranspiration were gathered.

The simulated steady state response of both systems to an average meteorological year are given in Tables 1 and 2.

Table 1 represents the infiltration through the different layers of the cover as percentage of the total input i.e. the net precipitation. The splitting of the clay layer reduces the final infiltration into the vault with 30 to 40%. Investigation of the flow paths ensures that no run-off water will re-enter into the vault.

As shown in Table 2 the gravel wall in contact with the vault remains nearly unsaturated.
### Table 1: Infiltration through cover layers

<table>
<thead>
<tr>
<th>Layer</th>
<th>Infiltration in % from net precipitation</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>reference design</td>
</tr>
<tr>
<td>Top soil</td>
<td>34</td>
</tr>
<tr>
<td>bio barrier (gravel)</td>
<td>23</td>
</tr>
<tr>
<td>upper clay layer</td>
<td>4.2</td>
</tr>
<tr>
<td>clay layer</td>
<td>sand drain</td>
</tr>
<tr>
<td>lower clay layer</td>
<td></td>
</tr>
</tbody>
</table>

### Table 2: Average saturation level of the cover layer and gravel wall

<table>
<thead>
<tr>
<th>Layer</th>
<th>Saturation level (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>reference design</td>
</tr>
<tr>
<td>Top soil</td>
<td>70</td>
</tr>
<tr>
<td>bio barrier</td>
<td>12</td>
</tr>
<tr>
<td>upper clay layer</td>
<td></td>
</tr>
<tr>
<td>clay layer</td>
<td>sand drain</td>
</tr>
<tr>
<td>clay layer</td>
<td></td>
</tr>
<tr>
<td>soil in contact with vault</td>
<td>gravel wall</td>
</tr>
</tbody>
</table>
5 Conclusion

The impact assessments of the burial site have suggested main improvements of the concept: with a simple modification of the barrier system, the flux of radioactivity, which is linear to the infiltration rate into the vault, can be reduced with 30 to 40%. Due to the lack of water in the gravel wall near the vault, diffusion in the aqueous phase will be limited.

Modelling repository sites in general and the PORFOW code in particular have proven their practical utility in assessing the impact to the environment of landfills, hazardous waste deposits and engineered facilities for waste disposal.

Acknowledgement

The author thanks NIRAS/ONDRAF for sponsoring our assessments of shallow land burial sites.

6 Reference

Runchal A., 1993
PORFOW code: a software tool for multiphase flow, heat and mass transport in fractured porous media
ACRI, Bel air, USA
**Soil bacteria for remediation of polluted soils**

Dirk Springael, L. Bastiaens, Marc Carpels, Max Mergeay, Ludo Diels

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1 Bacteria from polluted soils

For the remediation of polluted soils, soil bacteria may be used which are specifically adapted to pollutants and which have colonized industrial areas. In this perspective, the VITO has constituted a collection of strains isolated from a variety of severely contaminated areas.

This collection includes:
- degraders of mineral oils (*Pseudomonas sp.*, *Acinetobacter sp.*...)
- degraders of polycyclic aromatic hydrocarbons (PAHs): representative isolates belong mainly to the genera *Sphingomonas* (18) (α-Proteobacteria) and *Mycobacterium* (high GC gram-positive).
- degraders of Polychlorobiphenyls: representative strains belong to the genus *Ralstonia* (e.g. - "*Alcaligenes eutrophus*" and other strains related to β-Proteobacteria (*Burkholderia*, etc...) (15)
- metal resistant bacteria with plasmid borne resistances to Cd, Zn, Ni, Co, Cu, Hg and Cr.

Representative strains also belong to the genus *Ralstonia* (ex-*Alcaligenes eutrophus*) (17). Type strain is "*A. eutrophus*" CH34 (10). Most of these bacteria are hardy and suitable for survival in harsh soil conditions especially where vegetation is lacking (11).

Genetic and physiological studies on these bacteria focused on the mechanisms of degradation or resistance and on the construction of strains with improved remediation capacities or provided with reporter light producing genes allowing to track them in bio-reactors or in the environment.

Experiments with plasmid transfer have led to derivates which can express both efficient degradation of PCB and high resistance to heavy metals (15).

The main mechanism of resistance to heavy metals is efflux of toxic metals promoted by proteins located on the cytoplasmic and the outer cell membrane (3,6,9,12,13,14).

At high concentration of metals, bio-precipitation and/or sequestration of metals from the medium arose (post-efflux events) (5,6). This led to the development of membrane based bio-reactors which were designed to remove metals (but also PCB and chloro-aromatic compounds) from polluted effluents (7). Nevertheless, the major challenge is to use the bacteria adapted to the various pollutants, for soil remediation.

This requires the development of bench scale reactors to look at the performance of the various strains and at the feasibility for industrial application.

Batch Slurry Tank Reactors steered as Dry Solid Reactors for degradation of organic pollutants and Bacterial Metal Slurry Reactors for removal of heavy metals were designed in this respect.
2 Bio-remediation of oil contaminated soils in a dry solid reactor

Batch Stirred Tank Reactors (BSTR) were used to evaluate the efficiency of oil degraders (1). The microorganisms were first screened in smaller batches for breakdown of three oil components: dodecane (C12), pentadecane (C13) and octadecane (C18). Most of 25 test strains displayed efficient breakdown of the three tested alkanes. From the point of view of industrial feasibility, the strain which displayed the smallest pH drop and the fastest doubling time was chosen for testing in the dry solid BSTR.

The reactor system is built to enable working under wet and dry conditions. Biological breakdown under dry conditions is more efficient than slurry reactors because oxygen limitation is much smaller. The reactor has a working volume of 100 litres suspension and is completely closed. This volume is large enough to enable frequent sampling. Inox is used as building material because it is inert against aggressive nutrient mixtures and is known to have low interaction capacity with pollutants. Temperature is maintained at the desired temperature through a heated wall. Oxygen inlet and outlet are computer controlled and, if necessary, oxygen and CO₂ can be monitored on-line.

Additives like nutrients, new micro-organisms, detergents and water can be supplied automatically without opening the reactor. Sampling of gas is coupled with a GC for measuring hydrocarbons. Solid sampling is done without opening the reactor. Mixing efficiency can be controlled with a variable speed motor. A computer with Labview installed measures and controls all the functions described above.

The experiment was run at 28°C under the following conditions:
- water content 15% (w/w)
- diesel oil: 6000 mg/kg of dry matter
- aeration 16 l/hr
- inoculum: Acinetobacter calcoaceticus LH168

In about three days oil concentration dropped to 650 mg/kg. The analysis of gas components shows that about 500 mg oil/kg was removed by evaporation but 99% were metabolized due to the high mixing efficiency. The experiment provides a design to test a variety of pollutant degrading strains in similar conditions. In this respect is the microbial degradation of PAHs, which are especially recalcitrant, of special interest.

3 Bacterial degradation of fluorene in a dry soil reactor

*Stenomomonas* sp. LB126 was selected from an oil polluted soil nearby an oil refinery. The strain can grow on fluorene (C13H10, a tricyclic PAH) and efficiently co-metabolize dibenzothiophene, phenanthrene and to a lesser extent pyrene and anthracene.

Growth rate of *Stenomomonas* LB126 at the expense of fluorene is very slow (around 10-12 h⁻¹). In order to follow the fate of the strain in a dry soil reactor, the fluorene degrader was marked with reporter genes (8): the chosen marker was the lux operon (which promotes continuous bacterial luminescence via luciferase and fatty acid reductase).
Bacterial luminescence provided thus an easy way to track the fluorene degrader in viable counts from samples taken from the Dry Solid Reactor. Experiments with the Dry Solid Reactor had two purposes:

- to evaluate the endogenous degradation of fluorene in a recently polluted soil (70% dry matter) at a bench scale (50 kg soil).
- to assess the usefulness of adding a fluorene degrader and to optimize the remediation conditions.

Figure 1 shows the evolution of fluorene in the DSR in absence and in presence of the lux-marked strain of Sphingomonas sp. LB126 by adding inocula of growing size. The soil contained 500 mg fluorene per kg which is considered to be a substantial contamination. Elimination of fluorene is very slow in absence of the inoculum.

An inoculum of at least $4 \times 10^6$ cfu (colony forming units) is sufficient to promote the extensive degradation of fluorene. In five days most of the contamination had disappeared. The disappearance of fluorene had also as consequence the prompt decline in viable counts of the fluorene degrader. This is of special interest in the perspective of ecological bio-safety.
4 A slurry reactor for removal of metals from polluted soils

Historical emissions of former non-ferrous industry in the neighbourhood of the VITO, has lead to extended areas of contaminated sites. Zinc was the most abundant pollutant creating phytotoxicity while public health was mostly endangered by the presence of the toxic metal Cd. Besides Zn and Cd, also the metals Cu and Pb were present in the contaminated sandy soils.

"Alcaligenes eutrophus" CH34 (Ralstonia sp.) and related bacteria were isolated from soils and sediments strongly polluted by heavy metals. These bacteria contain megaplasmids coding for resistance against Cd, Co, Cu, Cr, Hg, Ni, Pb and Tl (4,6). The main resistance mechanism is energy mediated efflux of cations which is often accompanied by metal trapping or precipitation in and around the outer-cell membrane. These features together with a bacterially improved settling of soil were used to design a Bacterial Metal Slurry Reactor (BMSR) for the decontamination of sandy soils.

The Bacterial Metal Slurry Reactor (BMSR) consists of a continuous stirred tank reactor that is continuously fed with contaminated soil to which water and nutrients are added. During the start up period "Alcaligenes eutrophus" CH34 was added to the
slurry to a final concentration of 106 bacteria/ml. Bio-treatment was done with an hydraulic retention time of 10 hours.

The slurry was afterwards precipitated in a settler. The presence of CH34 improved the settling of the soil. The control soil contained about the same concentration of bacteria (indigenous bacteria in this case), but no good soil sedimentation was observed. The soils treated with CH34 showed a very good sedimentation after one hour, while the bacteria remained in the supernatant suspension. This characteristic was used to separate soil from bacteria. The bacteria could be recovered from the suspension via froth flotation process. The phenomenon of improved settling of soils in presence of CH34 was found quite empirically, yet may have promising applications in soil remediation. No explanation about this phenomenon is currently available: it would be of interest to test other strains or other mutants of CH34 affected in genetic determinants controlling the cell wall.

The results of the BMSR-treatment, presented in Table 1, give the Cd and Zn concentrations in the three final fractions: soil, water and biomass. The first example showed substantial reduction of Cd from 20 ppm to 1 ppm. In the second example, the Cd concentration could only be reduced from 13 ppm to 6 ppm. Other experiments (results not shown) showed again for some soils more than 95% Cd reduction and in other cases only 50%. Soils containing cadmium under metallic form or as sinters, could not be cleaned below governmental standards (16) used in The Netherlands (3 ppm for Cd) or in Belgium (2 ppm for Cd). The biologically unavailable Cd could hardly be removed. A second treatment or a longer hydraulic retention time could not further reduce the concentration. The results presented here show a very soft biological method to remove bio-available metals from sandy soils. After treatment, an intact soil is obtained. When metals are present under a more unavailable form more severe extraction methods are necessary. Most of the time, these aggressive methods irreversibly damage the soil structure and inhibit revegetation (data not shown).

Table 1: Cd and Zn concentrations in soil, water and biomass after a BMSR treatment. Metal concentrations are expressed in mg metal/kg soil.

<table>
<thead>
<tr>
<th>Fraction</th>
<th>Zn</th>
<th>Cd</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Example 1</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Soil before</td>
<td>2075</td>
<td>18.86</td>
</tr>
<tr>
<td>treatment</td>
<td>1000</td>
<td>1.15</td>
</tr>
<tr>
<td>Soil after</td>
<td>26772</td>
<td>250.40</td>
</tr>
<tr>
<td>treatment</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Biomass</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Example 2</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Soil before</td>
<td>881</td>
<td>13.00</td>
</tr>
<tr>
<td>treatment</td>
<td>563</td>
<td>5.70</td>
</tr>
<tr>
<td>Soil after</td>
<td>4200</td>
<td>1334.00</td>
</tr>
<tr>
<td>treatment</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Biomass</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
5 Future work

The data presented here show that various soil bacteria isolated from polluted soils (the oil degrading Acinetobacter LH168, the PAH degrading Sphingomonas LB 126 and the metal resistant Ralstonia sp CH34 (e.g. Alcaligenes eutrophus) may efficiently remove the corresponding pollutants in bench-scale bio-reactors filled with (non-sterile) real soils. The bench-scale reactors were designed to obtain industrial feasibility of bioremediation.

Next step will be further up-scaling:
- the Bio Metal Sludge Reactor (BMSR) is now developed at the level of a demonstration plant and is being built as a 1 m³ capacity reactor to continuously process 200 kg of polluted soil. Garden soil samples polluted with heavy metals processed through BMSR will be tested for revegetation with various plants and monitored for residual metal release via bio-sensors. Continuous BMSR, where the bio-available metal becomes concentrated in the bacterial fraction easily separable from cleaned soil, can also offer perspectives for decontamination of soils polluted by radioactive fall-out.
- Up-scaling of Dry Solid Reactors will be carried out via the construction of a 3 m³ reactor designed to continuously process 1 to 2 T of polluted material (mineral oils, PAHs, PCB..)

The problem of mixed pollutions (e.g. soils polluted with a mixture of recalcitrant organics and metals) will be addressed via bacteria able to degrade PAHs (Sphingomonas sp and Pseudomonas putida), in which heavy metal resistance genes coming from “A. eutrophus” CH34 (Ralstonia sp.) will be inserted.

Future genetical engineering will also focus on cyanide degraders. Mixed pollutions (cyanides, PAHs, PCB, heavy metals..) are of special concern around gas work stations taken as a striking example but they are in fact located all over the world. The present results of bioremediation using specific strains and bench scale reactors, encourage a further testing of combinations of bacteria and natural plasmids encoding for metal resistance or catabolic pathways.

6 References


Biotechnology for site restoration: scope of the problem
Olga Bitchaeva

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Abstract

The scale and the level of utilization of biotechnology in scientific and industrial environments, is one of the parameters, characterizing the level of scientific-technical progress and economical potential of any state. Use of biotechnological approaches for solving problems related with nuclear and industrial power, is considered today as one of the priority directions. The sharp increase of investments in such “innovative” or “break-through” technologies as biotechnology, supported by a favourable tax policy, as well as experience acquired in different leading countries, show the way for efficient economical development and provide the basis for governmental policy in the field of science and technology for the next decade. This presentation focuses on the potentials of modern biotechnology and its benefits for solving problems related with the nuclear industry (mainly for site restoration). Following items will be dealt with: a brief scope of the problem, the advantages of biotechnology, some current applications in Russia, main points of international collaboration and some political considerations in view of further development of this promising direction.

1 Introduction

Choice of strategy and of effective technologies for solving problems of site restoration and management, for the disposal of radioactive waste and spent nuclear fuel in countries and for the development of nuclear energy, are both determined by specific national factors, as well as by the world status of scientific-technical progress in this field. Such factors are: the scale of national nuclear programmes, in accordance with existing energy resources and energy demands, the existence of a mineral-resource base (for example, presence of U available at low prices), the peculiarities of economical development of a country, the size of territory and sociopolitical factors. Presently, 13 countries, including Russia and Belgium, with a strong nuclear policy, rely on closed nuclear fuel cycles. This policy opens perspectives for biotechnology as one of the promising alternatives for remediation purposes in terms of economy, social acceptability and rational technology in practical realisation of current nuclear programmes.

2 Modern biotechnology

Modern biotechnology refers to the application of living organisms and their cellular and molecular components for the creation of new compounds and to the use of microbial processes for research, industrial purposes and practical uses. The application of microorganisms in site remediation is based on their potential to transform, accumulate and degrade a wide spectrum of compounds and forms of elements.
About 20 different technical and scientific disciplines including microbiology, ecology, genetic engineering, biochemistry, classical genetics, protein chemistry and hybridoma technology, provide the foundation of modern environmental biotechnology.

Biotechnology has obtained a new dimension over the past two decades with various techniques for in vitro recombination of DNA fragments from different organisms and with the discovery of new plasmides encoding for biodegradation (D-plasmides). The underlying multidisciplinary character of modern biotechnology simultaneously offers a new dimension of broad applications because desirable characteristics can be introduced or amplified in existing biological species more rapidly, precisely and safely, than has previously been possible. There is a broad range of applications from basic research through industrial uses of environmentally relevant techniques.

3 Main problems to be solved

One of the most complex current ecological problems is the management of accumulated radioactive waste (RAW) resulting from past activities: nuclear weapons production, use of nuclear energy for peaceful purposes and the restoration of contaminated sites. The end of the cold war has led to a considerable exchange of data on the status of this problem and has risen concern about provision for ecological safety in different states of the world.

In Russia, the acuteness and importance of the problem are presently dictated by the following main factors:

- the existence of huge amounts of RAW and spent nuclear material (SNM) accumulated at enterprises of different federal organs of executive power due to the development of nuclear industry and power. Furthermore, the existing industrial facilities do not provide treatment and reliable isolation of accumulated and newly generated RAW and SNM. Populations and state authorities are anxious about the potential radiation hazard to people and to the environment;
- the existence of large contaminated areas, resulting from different nuclear accidents, explosion tests, temporal storage and disposal, from activities of nuclear plants and from the whole nuclear fuel cycle.
- the active and growing influence of the public on issues connected with the ecological aspects of the activities of nuclear enterprises.
- the necessity to implement new or to improve existing techniques for site restoration and RAW management and adapt them to modern ecological and safety requirements.

4 Reasons for choosing biotechnology in site remediation

The growing interest for using new non-traditional methods for site restoration, based on biotechnology, is determined by considerations such as international liabilities in the field of radioactive and toxic wastes disposal, the problem of import of “dirty” technologies and wastes for some countries, the social unacceptability of certain traditional technologies, the permanent increase of landfill costs in European countries, the search for economically efficient and ecologically clean, preferably closed technologies.
The most significant advantages of biotechnology, based on the use of microorganisms can be enumerated as follows:

- a wide action spectrum
- energy and resource saving
- economical benefit
- low primary cost of production of biomass at an industrial scale
- utilization of secondary raw material from the different industries
- reduction of ecological risk
- reduction of social anxiety in connection with the utilisation of biotechnology
- availability
- absence of undesirable side-effects from by-products

The basis of these biotechnological advantages is:

- the high potential of microorganisms to transform, accumulate and degrade a wide spectrum of compounds and forms of elements
- the mechanisms of self-cleaning of natural systems
- the high adaptational abilities of microorganisms for extreme conditions
- the versatility of physiologic-biochemical reactions of microorganisms
- the high microbial growth rate
- the low ash content of substances of microbial origin

The foundation of such processes lies in the mechanisms of microbial transformation of elemental forms. Microbial processes may result in:

- change of pH and redox conditions which influence the valency and ionic states of elements
- formation of chelates which in turn interact with elements and influence the valency and ionic states of elements
- dissolution and leaching of elements by action of microbial metabolites and decomposition products
- bioaccumulation and biosorption
- biomethylation
- formation of gaseous components such as CO₂, H₂, CH₄, H₂S
- isotopic fractionation, etc.

5 Biotechnology for site restoration

Presently, utilization of biotechnology for site restoration is considered as one of the priority and promising directions in modern science. Investigations in this area are carried out very efficiently in the USA (on the basis of DOE Programmes: "Subsurface Science Programme", "Biodegradation Programme", Microbial Physiology Subprogramme" etc.), in Japan, Germany, Finland as well as in other countries. In Russia this field is recently also considered as priority for the next decades in some regional programmes. Biotechnology is also included in main federal programmes, in a conversion programme of MAE RF and in some regional programmes of the North-West region of the RF.

A list of a hundred different techniques, including recent biotechniques for site restoration/remediation can be found in the annex of the paper of G. Collard. Among them are classical solutions, starting from control by natural processes up to land farming, pump-and-treat, soil washing, in situ bioremediation, combination of biological and physico-chemical techniques, biosorption techniques, use of biosubstances with multipurpose action etc. USA experts estimate, the most promising categories of technologies to be developed in the nearest future, should provide in situ, effective and economical remediation. Given these three criteria, and from the
From the point of view of ecological safety, durability and socio-psychological acceptability, biotechnology for site restoration takes a leading place. The main applications of biotechnology for site restoration in Russia include techniques which use:

- microbial biomass from wastes of biotechnological, microbiological, food industry
- microbial strain from genetic and microbiological collections
- phytosorbents
- chitin-chitosan biosorbents (different modifications and derivatives)
- sorbents on the base of natural and synthetic polymers
- sorbents on the base of wastes from wood-treatment industry
- complexed biosorbents
- biopreparates on the base of microbial biomass cultures
- biomolecules as metallotheonines
- microbial cells, encapsulated in polymerized hydrogels
- biopolymers
- gene-engineering developments
- bioextraction from pulp
- immobilisation on different carriers
- vermicultures

PS: bold letters: to be considered the most promising today

All these solutions, being applied for site restoration (decontamination of water, soil, sediments), can provide decontamination meeting the required norms. Their efficiency depends mainly on characteristics of contaminants and specific environmental conditions. Technological characteristics could be improved by making use of the influence of biochemical processes on biomaterial, redox and other conditions, as well as by choosing other microbial strains or biosubstances, by gene-engineering constructions etc. These biotechniques could also be used in combination with other techniques depending on the problems to be solved.

Since 1994 Russian CBNIP collaborates with SCK-CEN, UCL, VITO and other Belgian Institutes in this promising direction. Promising results were obtained on

- physico-chemical mechanisms of microbial transformation of forms of radionuclides in soil samples from the (Chernobyl NPP zone and Briansk regions)
- the dissolution of fuel compounds
- sorption of elements by biosorbents with
- decontamination of areas by means of microbial biomass and by means of biosubstances in laboratory and microscale field experiments. Joint expeditions, seminars, reports on conferences, publications, exchange of information were provided.

The main scheme of the Russian-Belgian collaboration in the frame of an international programme (IP) and in accordance with national federal and regional programmes, includes three stages:

2. Development and Adaptation (D&A), including D&A of promising techniques and effective biomaterial.
3. Semi-pilot and industrial production, including testing, certification and development of joint techniques and production of promising biomaterials.
- upgrading of transfer of joint effort from R&D up to industrial scale (commercialization)
- complex of marketing-technological works
- elaboration of system of international certification of biomaterials and biotechniques
- preparation of technological regulation and design for pilot production.

The main objectives for the immediate future are:
- to provide a retrospective assessment of existing solutions and experience.
- to demonstrate the perspectives of biotechnology for site restoration and management and disposal of RAW for concrete situations.

Another important issue of collaboration deals with the protection of intellectual rights in view of transferring know-how, the coordination of running collaborations with foreign partners, coordination of tasks in the frame of federal and other programmes. Russian CBNIP prepared a concrete programme of Russian-Belgian collaboration on application of biosorption techniques as well as on adaptation of other biomethods. Series of biotechniques, based on utilization of effective biosubstances and biosorbents are planned to be applied for decontamination of soil, water, sediments, for decontamination of buildings, for liquid RAW treatment, as well as for other tasks in joint research.

To succeed in effective realization of this programme dealing with production and effective utilization of biomaterials and biotechnological techniques, it is necessary that efforts of industrial-technological and scientific research institutions are joined. Presently, in Russia and Belgium there is a joint methodological, factological and material-technical base for successful realization of programme concerned.

The overview of detailed information about some techniques, developed and applied by CBNIP was submitted in the report "Biotechnology for site restoration" (O. Bitchaeva, CBNIP-MAE RF. 1996) for the Report of SCK•CEN "Site restoration" (G. Collard, 1996) and in the book "Biotechnology for Site Restoration and Waste Management: scientific, technological, educational, social, economical, political aspects" Editors: O. Bitchaeva, C. Ronneau, G. Collard. Kluwer Academic Publisher (in press).

6 International collaboration

The international scale of ecological problems resulting from activities of the nuclear industry, points to the priority needs in international collaboration for their solving. A previous collaboration (1993), between the Russian CBNIP and the SCK•CEN, UCL, VITO and other research centres, allowed to elaborate on an international programme (IP), dealing with the utilization of biotechnology for solving the problems related with the nuclear industry. Areas of concern are site restoration and management and disposal of wastes.

The main goal of the IP is:
"Creation of a modern park of biotechnologies and environmental technologies for nuclear and industrial power, suitable to international demands to technologies and providing a management with developed technologies and products on external and internal markets on all stages of their developments, improvements, tests and innovations."

This IP includes proposals from about 300 centres from 33 countries. The IP has been presented at different symposia, conferences, at joint meetings with the EC, with Ministries, and European firms. In 1994 NATO proposed to sponsor the NATO ARW, dedicated to the first version of this international programme. Scientific,
technological, educational, business, social, economic, political aspects were considered. The main conclusions of the ARW, formulated as advises for future tasks, included the completion and publication of the IP, preparation of a book giving an overview of the present state-of-the art of the problem concerned, the organization of the steering committee of the IP, and the organization of an international association (IA) and international centre (IC) in this field, and the continuation and enlargement of running and establishing a new collaborative research. Presently, all these activities are in a progress.

The main mission of the IC is as follows:

"The Centre as a multinational, multibranch (Industry-Universities-Research Centres-Companies), multidisciplinary, multipurpose and multipotential consortium, will be able to offer a concrete fundamental solution to problems, dealing with waste management, prevention, storage, disposal and site restoration on all stages of its life cycle, in all aspects, from the research & development stage up to technological schemes, its innovation into the practice, industry and market, focusing on advancement of biotechnology and other perspective technologies."

The participation of different countries in the IP and IC could be a stimulus for the promotion, realization and consolidation of this promising direction also at national level. The IC, as coordinator of IP, must provide the coordination of the scientific-technical policy and the optimal use of the potentialities of different countries-executors of the IP and should accelerate the transfer of biotechnology as fundamental science into the area of its practical use for site restoration and RAW management in close cooperation with the National Centres.

The scope of main problems, main points and objectives of International Programme and International Centre (IC) are submitted:

- in Statute of IC (prepared by G. Collard and O. Bitchaeva. 1996) and

7 Perspectives

In a report of the Environmental Protection Agency (EPA) on environmental applications of biotechnology it was stated (1990), that:

- The EPA recognizes that new innovative technologies are needed to effectively prevent, treat and remediate the growing environmental problems facing the nation.
- Biotechnology can be used to achieve pollution prevention. Naturally occurring and genetically engineered microorganisms (GEMs) offer great potential in reducing the use of chemicals for agricultural applications.
- There is a need to encourage basic and applied research on the use of biotechnology as a new innovative tool for pollution prevention and to understand the full potential for its safe and effective use.

Recent endorsements by the EPA and other organisations are once more an indication of the increased level of acceptance of this technology. In Europe, such a clear policy statement to encourage and promote the use of innovative technologies, including modern biotechnology, has not yet been agreed upon. Recently, however, a serious reorientation of priorities in the European scientific-technical policy towards environmental technologies, including biotechnology is in a progress. The
8 Conclusions

Presently, biotechnology is applied in a broad range of industries and markets. The bioremediation market is just being developed and its potential appears quite large. The growing demand for new approaches to mitigate environmental contamination has resulted in the increased use and acceptance of biotechnological approaches for remediation by commercial and military sectors. In general, from the estimations prepared by the Freedonia Group, Inc., the annual bioremediation market, estimated of $155 million for 1992, will grow by an average of 15.2% through 1997 to $315 million. The regulatory and cost advantages for the application of in situ biological treatment are playing a significant role in the expansion of this market. A significant portion of the market will continue to be involved in the clean-up of closed military facilities. The largest growth is, however, predicted to occur in the areas of in-process pollution treatment with bioreactors, and biofilters since large industrial manufacturers attempt to reduce process emissions and cut pollution abatement costs. It is obvious that the biotechnological approach will primarily focus on the nuclear and heavy industry as the first source of contaminants.

For Europe, the EC should integrate, on a specific basis, all promising technologies and should improve and coordinate the options to apply modern biotechnology for benefit of man and environment:
- to elaborate a concept for the utilization of biotechnology for site restoration and prevention and management of wastes, following international and national laws and in the frame of international concepts on waste management and site restoration
- to stimulate additional quality research and development in environmentally relevant biotechnology
- to encourage public understanding of the (environmental) benefits of biotechnology through education and training
- to use legislative and other means to encourage and establish biotechnology as effective and environmentally “soft” business opportunities
- to realize appropriate and efficient technology transfer from the various R&D programmes into practical applications.

Whether or not the economical and ecological benefits of the biotechnology revolution will apply to Europe, will ultimately depend on the current and future ability of European industry to invest and to compete on national and international markets. High profitability, economy and efficiency of biotechnology for nuclear industry justify the necessity of investments by the government in the creation, development and improvement of modern parks for biotechnology. This is important for the development of an ecological and safe modern nuclear industry of world standard. The International Programme and the International Centre could play an important role in this process.
Unrestricted reuse of decommissioned nuclear laboratories
René Cornelissen, Luc Noynaert, Sven Harnie, Jozef Marien

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Abstract

The common building decommissioning strategy consists in reaching the green field. Chronological decommissioning steps are dismantling of the equipment, removal of contaminated parts of walls and floors using appropriate techniques and finally demolishing of the remaining structures using conventional techniques. It was decided by the SCK•CEN to differ from this common strategy by limiting the decommissioning works to the radioactive parts of the nuclear installation and to obtain an attestation of unrestricted reuse for the building after removal of all the radioactivity. After refurbishment, the building can be used for new industrial purposes outside the nuclear field. Four buildings were decommissioned according to this strategy. Three of them already received their attestation for unrestricted re-use.

1 Introduction

In the past, the SCK•CEN (the Belgian Nuclear Research Centre) was involved in nuclear and non-nuclear research programs. In the early nineties, the Belgian government decided to restrict the operation field of the SCK•CEN to the strictly nuclear programs. A new research centre, the VITO (Flemish Institute for Technological Research) was founded and took over all non-nuclear activities. The VITO is housed partly in former SCK•CEN buildings. Besides highly equipped non-nuclear laboratories and offices these buildings contain laboratories and installations with a radiological history.

In general, the goal of the common decommissioning strategy is reaching the green field by dismantling the equipment, removing contaminated parts of walls and floors using appropriate techniques, and finally demolishing the remaining structures using conventional techniques. Since important parts of some VITO buildings (former SCK•CEN) contain valuably non-nuclear infrastructure and equipment, the SCK•CEN decided to differ from the common strategy by limiting the decommissioning works to the radioactive parts of the nuclear installation and to obtain an attestation of unrestricted reuse for the building after removing all radioactivity. After refurbishment, the building can be used for new industrial purposes outside the nuclear field.

2 Arrangements prior to the decommissioning activities

Definition of the unrestricted release and/or reuse limits

In the absence of a well-defined Belgian regulation for unrestricted release of suspected and/or decontaminated materials, the SCK•CEN Safety Service, in association with the Authorized Control Organism for Radiation Protection (AVN), specified the following release limits:
• 0.4 Bq/cm² surface contamination for $\beta\gamma$ emitters
• 0.04 Bq/cm² surface contamination for $\alpha$ emitters
• release the residual radioactivity of the radionuclides present in representative samples of the construction materials of the building must be similar to that of corresponding construction material originating from a non-nuclear zone
SCK•CEN and AVN based the draw up of these limits on already existing Belgian rules and regulations in preparation by IAEA.

They also defined the methods that must be used to analyse and monitor the radioactivity from the start of the decommissioning until the final release, namely:
• monitoring the entire wall and floor surfaces
• radiological analyses of the washwater of walls and floors
• measurement of selective core samples

When the results of these measurements and analyses are below the above-mentioned limits, the Safety Service establishes the attestation of unrestricted reuse and submits it for approval to the independent Control Organism.

The buildings that need to be decommissioned are the Physics Building, the Metallurgy Building, Bloc 3 of the Chemistry Building and two Radiobiology buildings.

The Physics building consists mainly of laboratories, offices and a waste storage room. Only a few laboratories were used for experiments and measurements with radioactive materials such as $^{14}$C, $^{137}$Cs, $^{60}$Co, $^{133}$Ba and $^{85}$Sr. The total wall and floor surface of these laboratories covers approximately 700 m². Average contamination levels were below 2.5 Bq/cm² with hot spots up to 30 Bq/cm² for $\beta\gamma$ emitters and 0.1 Bq/cm² for $\alpha$ emitters.

The decommissioning of the Metallurgy Building is, compared to the Physics building, somewhat more complicated. Besides a number of conventional laboratories, a large hall for material testing, some cellars and a storage room were contaminated. In these laboratories characterization programs on different kinds of fuel, fissile material and waste were carried out resulting in contamination with Thorium and Uranium isotopes. The total wall and floor surface of the potentially contaminated areas is approximately 4 800 m². Average contamination levels were below 2 Bq/cm² with hot spots up to 60 Bq/cm² for $\beta\gamma$ emitters and 0.5 Bq/cm² for $\alpha$ emitters.

Bloc 3 of the Chemistry Building consists mainly of laboratories and offices. The laboratories were used for experiments and measurements with radioactive materials, fuel and fissile materials. The total wall and floor surface of these laboratories covers approximately 4 300 m². Average contamination levels were below 500 Bq/cm² for $\beta\gamma$ emitters and 0.12 Bq/cm² for $\alpha$ emitters.

Two Radiobiology buildings needed to be decommissioned, namely the Bio-lab and the Bio-animals I. Both buildings contain laboratories, offices and storage rooms. In addition, the Bio-animals I building contained several animal cages. Extended experiments and measurements of the impact of radiation and contamination on plants and animals were carried out in these laboratories. A whole range of isotopes, including $^3$H, $^{14}$C, $^{32}$P, $^{238}$U, $^{239}$Pu and $^{241}$Am, were used. The total wall and floor surface of these laboratories and cages covers approximately 3 500 m². Average contamination levels for the surfaces were below the limits for unrestricted release/reuse. Hot spots of 1.5 Bq/cm² for $\beta\gamma$ emitters and 0.15 Bq/cm² for $\alpha$ emitters were found.
3 Decommissioning activities

**Safety Report** Before starting with the actual decommissioning activities, a safety report has to be established and approved by the Safety Service. This safety report describes the installation to be decommissioned, identifies the problems in terms of radiation doses and contamination and also describes the organization of the decommissioning works. When the decommissioning activities are carried out by an external company, the safety report also serves as a schedule of conditions for the call for tenders.

**Dismantling methodology** The strategy used for decommissioning the above described facilities is the following.

The zone to be decommissioned is first isolated from the rest of the building and equipped with hand/foot monitors and an air-monitoring device. In this zone, an area is set up for decontamination, material reduction (photo n°1), sorting out and packaging of the waste produced during the dismantling and demolition activities.

The first step in the decommissioning process exists in scanning for αβγ contamination on loose materials and equipment inside the controlled area. Not contaminated objects are evacuated to the outside of the controlled zone. Suspected or contaminated items are brought to the decontamination area and treated.
Next, the utilities anchored in the walls and floors such as ventilation pipes (photo n°2), fume hoods, waste-piping etc. are demolished and removed to the decontamination area for treatment.

Based on the results obtained by scanning the dismantled and/or decontaminated items, the agent of the Safety Service decides whether the objects can be freely released or whether they have to be disposed of as industrial waste on a public dumping ground. Materials that cannot be decontaminated are evacuated as radioactive waste following the specifications of NIRAS (the National Institute for Radioactive Waste and Enriched Fissile Materials).

The next step includes vacuum cleaning and washing the floors and walls, followed by mapping all surfaces to identify which parts are contaminated and have to be removed by cutting and drilling. This sequence of washing, mapping and cutting is repeated until all contamination has disappeared.

Then, all the surfaces are washed again. The washwater is collected and analysed by $\alpha$ and $\gamma$ spectroscopy. Core samples are taken at random from floors and walls for further measurement by $\alpha$ and $\gamma$ spectroscopy.

Finally, the demarcation of the zone is removed.

The decommissioning is carried out from the lowest contaminated lab to the most contaminated one. Continuous supervision of the Safety Service personnel is required.

The choice of the decommissioning tools was based on their simplicity in manipulation and the ability of minimizing cross contamination.

Standard saws were used for dismantling and size reduction of contaminated pieces. Ventilation pipes were cut by means of electrical nibblers and scissors. Contaminated wall plaster, concrete, stone and tiles were removed using electric and pneumatic needle scalers and drills. Water and common decontamination products were used for washing walls, scrubbing floors, cleaning apparatus and equipment etc... When using these tools, the intervention crew wore tyvec overalls, safety shoes, gloves and full face masks when needed.
Waste handling

All solid radioactive waste was packed and evacuated according to the specifications of the National Institute for Radioactive Waste and Enriched Fissile Materials (NIRAS). Washwater collected during decontamination were sampled, measured by α and γ spectroscopy and evacuated as liquid waste.

4 Results obtained

The decommissioning of the Physics Building, executed by the intervention crew of the SCK•CEN, was used as a test case. During the decommissioning works, the daily activities in the non-contaminated parts of the building were going on as usual. This caused some stress for members of the personnel not familiar with radioactivity. No major problems were encountered during the decommissioning of the building itself. Nevertheless, the contamination level of some parts of the infrastructure such as window shelves and doors was higher than expected. The attestation of unrestricted reuse of the Physics building was delivered in August 1993.

The Metallurgy Building, Bloc 3 of the Chemistry Department and the two Radiobiology buildings were decommissioned by an external company selected on basis of a call for tender. All the activities were supervised by the SCK•CEN Technical Liabilities team and the Safety Service. Before each decommissioning phase, a meeting was held to inform the employees of the VITO about the decommissioning activities and the safety conditions.

After complete decommissioning of the buildings, the transferable αβγ contamination was below 0.001 Bq/cm². The core samples have a similar radionuclide spectrum as corresponding material coming from a non-nuclear zone. The total αβγ activity measured on those samples was below 1 Bq/g.

The decommissioning attestations were obtained in 1994 for the Metallurgy Building and in 1995 for Bloc 3 of the Chemistry Building. The attestation for the Radiobiology is expected before the end of this year. Recently, the VITO has officialised its agreement for the transfer of the decommissioned buildings.

5 Costs

The dismantling and waste costs for the different buildings (FYS = Physics Building, MET = Metallurgy Building, SCH = Chemistry Building and BIO = Radiobiology building) are given in table 1. The dismantling costs include the expenditures for SCK•CEN personnel and external personnel. Waste costs are calculated according to the NIRAS waste tariff for the different types of waste.

<table>
<thead>
<tr>
<th>Table 1: Dismantling and waste costs (MBEF)</th>
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<tr>
<td>Dismantling</td>
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<tr>
<td>Waste</td>
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</tbody>
</table>
6 Future works

The decommissioning of the outside waste water piping for the buildings on the Radiobiology site is foreseen in the near future. Before the year 2000, it is intended to restore the surrounding site that contains pastures, a greenhouse and a farm where experiments were done to study the transfer of radionuclides like $^{90}$Sr, $^{137}$Cs, $^{14}$C and $^3$H to plants and animals.

7 Conclusions

The SCK·CEN has demonstrated that decommissioning and measurement techniques are available to clear, by the legal rules, old laboratories to the level of unrestricted reuse so that they can be accepted by the non-nuclear world.
Technology for site remediation: Availability, needs and opportunities for R&D at SCK•CEN
Guy Collard

SCK•CEN
Waste & Dismantling
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Abstract

A lot of experience has been gained over the past several years in the use of control and treatment technologies, applied to different configurations and types of contaminated sites and environments. Although available technologies are adequate in many cases, it is recognized that many technologies being utilized can be too costly to set up, and often, they are inadequate to address the multitude of contaminant problems. This recognition has led national and international organizations, as well as private organizations and university communities, to sponsor environment technology development programmes to address technology needs. For example, the U.S. DOE has initiated an aggressive environmental technology development programme and the Commission of the European Union is sponsoring environmental technology development. This presentation focuses on the more innovative and emerging technologies that may be important for the future and on the opportunities for the SCK•CEN in the needed Research, Development and Demonstration programs (RD&D).

1 Introduction

Substantial contamination of air, soils, water, vegetation and structures has occurred due to activities related to the development and the application of nuclear energy and the use of radioactive substances such as the fabrication and testing of nuclear weapons, nuclear power production and its fuel cycle, mineral processing of ores and radionuclides for industrial and medical applications, and waste management practices and accidents. Some principal contaminant radionuclides which may be of concern are $^{226}$Ra, $^{222}$Rn and daughters, $^{60}$Co, $^{137}$Cs, $^{90}$Sr, $^{239}$Pu, $^{241}$Am.

As a result of this contamination, several countries have initiated programmes to assess and remediate radioactively contaminated sites. For example, the U.S. DOE has allocated more than 18 billion U.S. dollars to identify and, manage the problem, and assess the degree of remediation necessary for the various sites and facilities. The Chernobyl Nuclear Power Plant (NPP) accident, which resulted in extensive contamination transcending national borders, has led to an international effort to restore contaminated environments. More recently, there has also been growing concern regarding radioactive contamination resulting from sources outside the nuclear fuel cycle, which is leading to development of appropriate regulations, assessments and action in several countries.

The aim of this presentation is to provide some technology information and to show opportunities of Research, Development and Demonstration (RD&D) for the SCK•CEN. Therefore emerging and promising technologies for restoration and remediation of radioactively contaminated sites get more attention than well-established techniques and processes.
2 Terminology

This paragraph does not intend to fix the general terminology to be used in the field but rather defines our interpretation for the most common terms encountered when dealing with Site Restoration. Table 1 summarises the main definitions as understood for soil treatment.

Table 1: Terminology applied to soil treatment.

<table>
<thead>
<tr>
<th>By:</th>
<th>We understand:</th>
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<tbody>
<tr>
<td>Fixation/Immobilization</td>
<td>The action of preventing the spreading of the contamination into the soil and the action of preventing the transfer of the contaminant to the food chain or to the population by any mean. This does not imply the removal of the isotopes from the soil.</td>
</tr>
<tr>
<td>Remediation</td>
<td>The application of any technological method to fix or immobilize the contaminants.</td>
</tr>
<tr>
<td>Extraction</td>
<td>The action of separating the contaminant from the soil.</td>
</tr>
<tr>
<td>Clean-up</td>
<td>The application of any technological method to separate the contaminants from the soil.</td>
</tr>
<tr>
<td>Restoration</td>
<td>The application of any technological method that leads to the final clean-up of the site. This may imply a first step leading to immobilization but it should always be followed by clean-up to finalize the whole operation.</td>
</tr>
<tr>
<td>Countermeasure</td>
<td>A set of preliminary and immediate actions helping in decreasing the immediate risks of transfer of the contaminants to the food chain or the population. Countermeasures are provisional in nature.</td>
</tr>
</tbody>
</table>

3 General approach to remediating technology

The first selection of potential technologies will mainly depend on the type of contamination and on the type of contaminated zones. The contamination can be aerially extensive or localised. Contaminated zones can be limited (nuclear site, incidental contamination) or extended. Extended zones may be urban, rural, forestal, etc.

The decision to implement the clean-up of a contaminated area may be made on the basis of the Derived Intervention Levels (DILs) for this protective measure. Once the decision has been made, then clean-up criteria should be available to define the specific radionuclide concentration limit or gamma exposure level which should be achieved by remedial action in a particular area. In addition, reentry criteria should be established by which it can be decided whether to allow the return of the population and/or reuse of the land for agriculture, etc.

The development of such criteria which relate the dose to humans to contamination levels using pathway analysis, is difficult for small sites and extremely difficult for large diverse regions. In practice, different acceptance criteria may be set for different zones or situations in large contaminated areas. Fortunately, by the time large scale
Selecting preferred remediation action

During or after the preliminary characterization an engineering study should be implemented to investigate remediation options to address this specific contaminant problem and to reduce radiological and chemical exposure. A preliminary selection of options may be made based on several factors including:
- site end use,
- technical and institutional considerations,
- public acceptability,
- cost,
- meeting regulatory requirements, etc.

A further focused investigation of one or more particular method(s) may be also conducted, which may include conducting a bench-scale and pilot-scale tests of a specific technology. These tests would be designed to collect sufficient information to design, procure, and operate a full-scale system. A site specific remediation concept would be developed from these particular methods. A final decision to adopt the preferred remediation action would be made by the competent national authority.

Once a decision has been taken, clean-up criteria should be available to define the specific radionuclide concentration limit or gamma exposure level which should be achieved by remedial action in a particular area. In addition, reentry criteria should be established by which it can be decided whether to allow for the return of the population and/or for the reuse of the land for agriculture, etc.

The development of such criteria which relate the dose to humans to contamination levels using pathway analysis, is difficult for small sites and extremely difficult for large diverse regions. In practice, different acceptance criteria may be set for different zones or situations in large contaminated areas. Fortunately, by the time large scale clean-up is initiated, only a few longer lived radionuclides would need to be considered in setting criteria. It is advisable that the criteria are based on risk levels translated into acceptable dose limits. For rural areas, concentration limits for radionuclides in soil, water, air and food or acceptable radiation levels can be derived using suitable pathway analysis and, where possible, realistic site specific parameters. For urban areas, an integrated evaluation of the radiation from various surfaces should be undertaken.

Sites with mixed contamination introduce added complexity in the selection and design of a remediation system and strategy. First, the mixture may affect the actual extent of contamination. For example, at the Hanford site, the mixture of carbon tetrachloride with plutonium in the effluent disposed to the soil (Rohay and Hagood) has led to deeper penetration of the plutonium in the soils due to the complexation of the organic components with the radionuclides. Secondly, the design of the remediation system itself must consider the cocontaminants. For example, sorbents used to collect radionuclides may preferentially select the chemical contaminant over the radioactive contaminant, affecting the efficiency of the system. Thirdly, packaging and disposal of "treated" material may also be affected as certain countries maintain different requirements for hazardous waste, radioactive waste, and mixed wastes.

Implementation

Implementation of remedial action should be based on sound principles of project management and the ALARA principle. Implementation includes the design of the remediation system is completed; procurement is initiated; site preparation is conducted; a health and safety plan is developed; operations procedures are developed;
staffing is determined and training initiated; disposal is conducted; etc. At the end of this phase, the remediation is verified and the site is restored.

4 Remediation and restoration technology

Many technologies are or can be used depending of the types of sites, environment and contamination.

Some of them are listed here below:

Self-remediation processes

- Decontamination of structures
- Motorized sweeping and vacuum sweeping
- Firehosing and flushing
- Aqueous methods incorporating chemical additives
- Abrasive jet cleaning
- Road planing/grinding
- Strippable coatings
- Clean-up of indoor contamination
- Decontamination of equipment
- Guidance on the selection and application of decontamination methods

In-situ remediation technologies

- Containment/immobilization
  - Capping
  - Fixants
- Subsurface barrier
  - Sheet Piling.
  - Grout Curtains.
  - Slurry walls.
  - Viscous Liquids.
  - Flowable Grout Techniques.
  - Soil Freezing Technology Applications.
  - Circulating Air Barrier.
- Passive vertical walls
- Encapsulation/stabilization
  - Stabilization (or solidification)
  - In Situ Vitrification of Contaminated Soils

In-situ treatment

- Subsurface bioremediation
  - In situ immobilization of uranium by bacterial reduction of U(VI) compounds to uraninite U(IV).
- Permeable Treatment Barrier
- Chemical Osmication

- Subsurface manipulation/process control
  - Electrokinetics
  - Soil Flushing

Agricultural methods

- Deep ploughing
- Revegetation
- Soil additives
Crop selection
Material removal technologies
Removal of vegetation
Removal of surface soil
  Standard excavation
  Remote excavation
  Cryogenic retrieval
  Dust control
  Other methods
Ex-situ treatment technology
  General
  Radon treatment in air
    Active vapour extraction
    Passive soil venting
    Passive soil vapour extraction.
Soil treatment
  Physical and chemical separation of radionuclides from soil
  Chemical extraction of radionuclides from contaminated soil.
  Magnetic separation
  Incineration
  Solidification
Water treatment
  Pumping and treatment systems
  Precipitation
  Ion exchange
  Membrane separation
  Adsorption
  Aeration

It is beyond the scope of this presentation to describe how these technologies are or can be applied to site restoration and remediation. Nevertheless, the authors can provide more information on request.

Most of the current technologies will appear as antiquated in the 21st century as technologies of 50 years ago seem today. In 1940, scientists had to use photographic plates to spectrally characterize molecules. Current technology has advanced to allow chemical mapping of the human body. The same degree of advancement should be made about restoring the environment in the next 20 years. The effort will require the use of present resources and the development of new resources to address and solve the problems resulting from past disposal practices. [Ref: DOE Applied Research, Development, Demonstration, Testing and Evaluation, November 1989]. To meet their commitments, responsibles of the environment must develop the ability to process increasingly dilute solutions.

The nuclear industry, like all other production entities that relied on shallow land burial, cribs, ponds, lagoons, and underground storage tanks and transfer lines for disposal, has created a system that has affected surface water and ground water. The forces driving subsurface contamination include percolation, vapour transport, and transportation of contaminants by leaked and spilled solvents. The subsequent migration of organics, inorganics, and radioactive materials has resulted in wide dispersion of waste material at low concentrations. Thus, the environmental remediation challenge must focus on the source of the material and on restoring and preventing contamination of the waters under
and around facilities. Confining contamination, removing the source of contamination, and restoring water containing low concentrations of contaminants pose problems of enormous magnitude.

The challenge is to process high volumes of low value (concentration) materials and to enhance natural degradation: the earth's own technology. Present technology has been developed to extract valuable materials from ores or process streams. The production of metals, pharmaceuticals, organics, and weapons is based on extraction of valuable components from high-value (concentration)/low-volume and high-value/high-volume basic materials. This kind of process not only produces a usable material but also produces a side stream of low-value/high-volume waste. Generally, this waste has been disposed of into shallow land burial sites, has migrated, and has ultimately been intercepted by surface water or ground water. The result has been lower and lower value/higher and higher volume streams. Technology does yet not exist to extract efficiently and effectively or remove these diluted contaminants from water.

Technologies must be developed to ensure that responsible of the environment do not have to revisit these contamination problems in a few decades. By then, postclosure monitoring under the national and international laws and regulations should show that the responsible will have achieved their long-term goal. Technologies to be applied (and in many cases to be developed) must address anticipated environmental regulations of the future, not the present. Coupled with such technologies must be the development of technologies, processes, and materials that reduce the amount of waste generated requiring disposal. When disposal is necessary, all assurances must be developed to ensure minimum environmental impact.

5 Opportunities for the SCK-CEN

5.1 Experience of the SCK-CEN

For more than forty years, one of the missions of the SCK-CEN has been: "To inform, encourage, promote, and support the use and development of effectively demonstrated innovative technology for environmental investigation and remediation."

The SCK-CEN developed technologies for

• waste reduction, treatment and conditioning,
• waste disposal,
• site characterization,
• decontamination of structures,
• decommissioning,
• ecological use of contaminated sites, etc....

Site restoration and remediation is thus no new field of RD&D for the SCK-CEN. The only difference with the recent past is that it is now considered as a discipline, just as nuclear fuel cycle or waste disposal.

In some cases, the SCK-CEN developed knowledge and resources which could be straightly applied to site restoration and remediation. One example is the migration of radionuclides.
"The early phases of a hazardous waste site remediation include an assessment of the extent of contamination, and a characterization of the relevant physical, chemical and biological processes governing the fate and transport of the contaminants at the site. Accurately delineating the location of these contaminants is an important first step in the process. A second, more difficult, step involves obtaining an estimate of how much contaminant resides in the various phases (soil, water, vapour), and how quickly, under natural or engineered circumstances, this contaminant might be transferred out of a given phase, or transformed into different chemicals. Unfortunately, the former procedure alone can often require months (or years) of soil sampling and laboratory analyses. Accurate completion of the more detail-oriented latter procedure is even more involved, and, at many sites, is a procedure that is poorly understood." [Remediation and Restoration at UCLA's Center for Clean Technology, June 1995]

In the frame of the project "Waste disposal", the SCK•CEN is actively pursuing the development of faster and more accurate methods for assessing and understanding the distribution, transport and potential transformation of contaminants in the deep and subsurface environment. An increase in the accessibility and in the detail of such information would add substantially to the utility of the mathematical models used to explore the feasibility of different remediation strategies. Such models can be applied to explore the feasibility of different remediation strategies.

5.2 Philosophy of the SCK•CEN

The SCK•CEN has always placed a continuous emphasis on innovative technologies. In site restoration and remediation, the centre will also adopt a coordinating strategy addressing enhanced development and application of innovative technologies. Potential benefits include reduced risk, more permanent solutions, lower cost, and greater community acceptance.

5.3 Opportunities

It is beyond the scope of this presentation to describe promising innovative technology which could be applied to site restoration and remediation. The alphabetic list of innovative technologies being supported by the DOE (Annex 1) gives an idea of the scope of RD&D opportunities for a Research Centre such as the SCK•CEN [DOE, 1996].

The SCK•CEN is cleaning up its own site. This operation implies the solving of problems for which no industrial solution is available. In general, these problems do not need an urgent solution. Therefore, the SCK•CEN can orientate its RD&D programme so that it leads to technology which is presently needed elsewhere. Some examples are given here below.

**Decontamination of concrete**

The decommissioning of the BR3 reactor could lead to large quantities of waste in the absence of suitable decontamination or of removal of the contaminated fraction.

*Problem* Concrete surfaces contaminated by radioactive isotopes, heavy metals and organics create severe problems for conventional decontamination methods. Surface clean-up by mechanical scabbling or physical abrasion can result in the generation of large quantities of secondary wastes and highly toxic fine particles. Use of these methods also makes it necessary to employ stringent precautions to protect working personnel during the actual procedure. Disposal of large volumes of contaminated concrete and other wastes is expensive and absorbs limited resources.
Potential solutions exist such as bacterial decontamination, microwaves, electrokinetic decontamination, ....
Let us concentrate on electrokinetic decontamination.

Solution
A solution among others: electrokinetic decontamination.
This technology for the decontamination of concrete uses an electrical field to induce the migration of ionic contaminants from porous concrete. An electrolyte solution through which an electrical current is run is applied to the concrete, and the contaminants migrate from the concrete to the solution, with which they are then removed.

State of the art
Field pilot scale demonstrations are in process at Oak Ridge and are planned in support of the D&D Integrated Demonstration Program. Phase I will be completed with a thorough evaluation and report on the technical feasibility, clean-up levels, and cost analysis.

Remediation of the banks of the Nete river
The Nete river has been contaminated by liquid effluents containing low concentrations of radioisotopes. The banks are contaminated with trace quantities of Plutonium but this does not create radiological problem. Nevertheless since cases of more severe Pu contamination exist on river banks in Eastern Europe & the CIS, it is worth investigating in remediation research. And for the sake of fo the exercise, one could pretend that a remediation action would be requested.

Problem
Removal of the soil would lead to high amounts of slightly contaminated soil and an ex-situ treatment would be too costly.

Solution
A solution among others: Electrokinetics
If contaminants are to be removed from soil in situ, effective methods for moving the contaminants through the soil to collection points must be developed. Clays and tight soils offer the greatest challenge; hydraulic flow through fine pores will be nearly zero, making these soils non-responsive to traditional soil washing.
Electrokinetics is an attractive process for moving water and ionic materials through fine soils, because both electro-osmosis and electrolyte migration are transport processes that are independent of pore size. The SCK•CEN is familiar with this technique which is used it to accelerate migration of nuclides in the frame of the “geological disposal” programme.

State of the art
Electrokinetic remediation is incompletely understood because of the complexity of parameters and the interactions that occur when one applies a direct current between buried electrodes in contaminated soil.
Contaminants can move through the soil by three different processes induced by the applied field: electro-osmosis, electrophoresis, and electromigration. Electrolysis reactions that occur at the electrodes induce pH changes that can affect contaminant speciation and solubility. Contaminant mobility can also be influenced by soil permeability and the degree of water saturation. A mathematical model of all of these effects operating simultaneously will necessarily be complicated, and effects of parameters measured in isolation may not be the same as when changes occur together.

Opportunity for RD&D
The SCK•CEN is collaborating with the Russian CBNIP.
Technological advances made by Russian scientists in this area of environmental remediation could be used as much as possible. A selected part of the banks of the river
Technology need should be such as to allow for easy access for testing, be representative for the problems present elsewhere, and be accessible to industry, regulatory agencies, and academia.

**Conditioning of plutonium**

**Problem** Aged not irradiated plutonium, residues of MOX post irradiation studies led, to not negligible quantities of metallic Pu and plutonium oxide. A solution has to be found, according to the future disposal in a clay formation.

**Solution** Potential solution: Plutonium stabilization and packaging
It is necessary to stabilize and package plutonium metal and oxide according to their future disposal. However, none of the Belgian facilities has all of the equipment necessary for completing this task in-place. A Plutonium Stabilization and Packaging project could be initiated by the SCK•CEN for scoping, specifying, and conducting a procurement of a standardized set of packaging and stabilization equipment. Although this effort would be coordinated by NIRAS/ONDRAF, the SCK•CEN could contribute the necessary technical and systems engineering support and solve its own problems.

**Technology need** The resulting system would be obtained from one design effort and one procurement package, and yet meet the needs of each site. This would provide necessary standardization of stabilized materials and reduce duplication of efforts and costs that would be incurred if individual sites continued development. The standardized stabilizing, packaging, labelling and transfer system would simplify handling, accountability, inspection, identification, and transportation. The equipment would be operable under glove-box conditions, automated to reduce exposure, and stabilize and package material to acceptable standards, which will increase safety.

**Conclusions**
These few examples illustrate that the SCK•CEN could combine innovative RD&D with the solving of existing problems.

### 5.4 Long-term programme: development of biotechnology

Apart from punctual operations, the SCK•CEN could also develop an extended knowledge in order to prepare its long-term future. Although the development of biotechnology is not the only possibility, I take it as an example because biotechnology could be the only economically acceptable solution for large contaminated areas.

Bioremediation is the use of living organisms, primarily microorganisms, to degrade environmental contaminants. This innovative, "green", and versatile (multidisciplinary) technology is being used for many different kinds of site clean-ups. Bioremediation techniques can be adapted to meet clean-up needs: approaches can be ex situ or in situ; systems can be aerobic or anaerobic; and the bacteria can be indigenous, imported, or genetically engineered. Bioremediation can be a stand alone technology or combined with other technologies in a treatment train.

Ex situ and in situ is one of the major differentiations for bioremediation techniques. Ex situ processes require the additional cost for the excavation or removal of contaminated media and transportation to treatment facilities. In situ processes which involve treatment in place, are more difficult to evaluate for the resulting clean-up. By means of collaboration with other institutions dealing with biotechnology or/and by building its own skill the SCK•CEN could valorize its knowledge in nuclear waste and contaminated material treatment as well as in other technologies by developing
biotechnology or mixed technologies for waste treatment, decontamination and remediation.

6 Conclusions

Substantial contamination of air, soils, water, vegetation and structures has occurred due to activities related to the development and the application of nuclear energy and the use of radioactive substances such as the fabrication and testing of nuclear weapons, nuclear power production and its fuel cycle, mineral processing of ores and radionuclides for industrial and medical applications, and waste management practices and accidents. Technologies to be applied (and in many cases to be developed) must address anticipated environmental regulations of the future, not the present. Coupled with such technologies must be the development of technologies, processes, and materials that minimize the amount of waste generated requiring disposal. Site restoration and remediation is no new field of RD&D for the SCK-CEN. Indeed the SCK-CEN developed knowledge and resources which can be straightforwardly applied to site restoration and remediation. This new "science" could be a road for the future for our Research Centre.

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Annexe I  DOE Innovative Technology Alphabetical Listing

Acid Extraction Treatment System
Acoustic Barrier Particulate Separator
Adsorptive Filtration
Advanced Landfill Cover
Advanced Worker Protection System
Aerobic Microorganism for the Degradation of Chlorinated Aliphatic Hydrocarbons
Air-Sparged Hydrocyclone
Airborne Survey Drone
Airborne Unmanned Geophysical Survey System
Alternating Current Electrocoagulation Technology
Ambersorb® 563 Adsorbent
Anaerobic Thermal Processor
Anaerobic/Aerobic Sequential Bioremediation of PCE
Analysis of Vapor Phase Samples Using Pulsed Valves and a Pulsed Gas Discharge Apparatus for Incinerating Hazardous Waste
Arc Melter Vitrification
Asbestos Pipe-Insulation Removal Robot System
Associated Particle Imaging
Atomic Line Emission Analyzer for Hydrogen Isotopes
Audible Radiation Monitor
Augmented In Situ Subsurface Bioremediation Process
Automated Volatile Organic Analytical System
Autonomous Mobile Robot for Radiological Surveys
B.E.S.T. Solvent Extraction Technology
Base-Catalyzed Decomposition Process
Batch Steam Distillation and Metal Extraction
Below-Grade Bioremediation of Chlorinated Cyclodiene Insecticides
Biocube Biofilter
BioGenesis Soil and Sediment Washing Processes
Biological Aqueous Treatment System
Biological Sorption [AlgaSORB©]
Biological/Chemical Treatment
Bioremediation of Contaminated Ground Water
Bioscrubber
Bioslurry Reactor
Biotox Process
BioTreatSystem
Bioventing
Borehole Sampler
Bruker Mobile Environmental Monitor
Campbell Centrifugal Jig
Carver-Greenfield Process® for Solvent Extraction of Wet, Oily Wastes
CAV-OX® Process
CCBA Physical and Chemical Treatment
Ceramification for Stabilization and Immobilization of Plutonium Containing Compounds
Chelation/Electrodeposition of Toxic Metals from Soils
Chemical and Biological Treatment
Chemical Fixation/Solidification Treatment Technologies
Chemical Oxidation Technology
Chemical Soil Oxidation
Chemical Treatment
Chemical Treatment and Ultrafiltration
Chemotactic Selection of Pollutant Degrading Soil Bacteria
Chlorinated Gas Treatment Biofilm Reactor
Chromated Copper Arsenate Soil Leaching Process
Circulating Bed Combustor
Closed-Field Capacitive Proximity Sensor
Coherent Laser Vision System
Colloidal Borescope
Combined Chemical Precipitation, Physical Separation and Binding Process for Radionuclides and Heavy Metals
Cometabolic Bioreactor
Compact Processing Unit
Concentrated-Chloride Extraction and Recovery of Lead
Cone Penetrometer
Constructed Wetlands-Based Treatment
Contained Recovery of Oily Wastes [CROW]
Contained Recovery of Oily Wastes [CROW]
Contamination Containment System
Contamination Control Unit
Conveyor with Rotary Airlock Apparatus
Cross-Flow Pervaporation System
Cross-Flow Pervaporation System
Cross Borehole Electromagnetic Imaging Technology
Cryofracture
Cryogenic Crushing
Cryogenic Retrieval
Crystalline Silicotitanates
CURE®-Electrocoagulation Wastewater Treatment System
Cyclone Furnace
Cyclone Furnace
CYROCELL®
DARAMEND Bioremediation Technology
Debris Washing System
Dechlorination and Immobilization
Decontamination and Recycle of Concrete
Direct Current Graphite Arc Plasma Furnace
Direct Sampling Ion Trap Mass Spectrometry
Disposal Sludge Dewatering Container and Method
DNAPL Removal from Low Permeables
Double Containment Piping System
Down Hole Optical Probe
Dynamic Underground Stripping
Eimco BioLift Slurry Reactor
Electric Resistivity Tomography
Electrokinetic Decontamination of Concrete
Electro-Klean Electrokinetic Soil Processing
Electro-Klean Electrokinetic Soil Processing
Electro-Osmotic Remediation
Electrochemical In Situ Chromate Reduction and Heavy Metal Immobilization
Electrochemical Separation of Metal Ions Solution
Electrokinetics
Emission Monitoring Spectrometer
Entrained-Bed Gasification
EnviroGard PCB Immunoassay Test Kit
EnviroGard TM Immunoassay for PCBs in Soil
Environmentally-Safe Compact Radiation Shield
Environmental Test Kits
Equate® Immunoassay
Excavation Techniques and Foam Suppression Methods
Expandable Pipe Crawler
Expedited Site Characterization Methodology
Extraction of Polychlorinated Biphenyls from Porous Surfaces Using the TECHXTRACT Process
Fiber Optic Based Hydrogen Sensor
Fiber Optic Chemical Sensor
Fiber Optic Monitoring Device
Fiber Optic Probe Having Fibers with Endfaces Formed for Improved Coupling Efficiency
Field Analytical Screening Program PCB Method
Field Laser Metals Spectrometer
Field Portable Detection of VOCs Using a SAW/GC System
Field Raman Spectrograph
Field Screening Laboratory
Field-Usable Portable Analyzer for Chlorinated Organic Compounds
Filtration Stabilization Process for M-Area Waste Treatment (FIST Alternate A)
Flameless Thermal Oxidation
Flame Reactor
Flexible Pipe Crawler with Stabilizing Mid Section
Flow Measurement by Focused Ultrasonic Waves
Fluid Extraction - Biological Degradation Process
Fluid Flow Monitoring Devices
Fluidized-Bed Cyclone Agglomerating Combustor
FORAGER® Sponge
Forge Hammer
Formalex® FRC-3 TM
Fourier Transform Infrared Spectrometer
Frequency-Tunable Pulse Combustion System
Frozen Soil Barrier
Full-Scale Buried Waste Integrated Demonstration
Fungal Treatment Technology
Gas-Phase Chemical Reduction Process
Gas Chromatograph
Geoprobe Conductivity Sensor
GHEA Associates Process
GIS/Key Environmental Data Management System
Glass Material Oxidation and Dissolution System (GMODS)
Halosnif-Fiber Optic Spectrochemical Sensor
Heavy-weight Cone Penetrometer
Heavy Metals and Radionuclide Polishing Filter
High Efficiency Radiation Detector
High-Energy Corona
High-Energy Electron Irradiation
High-Energy Electron Irradiation
High Clay Grouting Technology
High Energy Electron Beam Irradiation
HNU-Hanby PCP Test Kit
HNU Source Excited Fluorescence Analyzer-Portable [SEFA-P] XRF Analyzer
Horizontal Well Air Stripping
Horizontal Well Insert for In-Situ Groundwater Remediation System
HRUBOUT® Process
Hybrid Directional Boring/Horizontal Logging
Hybrid Fluidized Bed System
Hydraulic Fracturing
Hydrolytic Terrestrial Dissipation
Hydrothermal Oxidation
Immobilized Cell Bioreactor Biotreatment System
Implanted Sensor Location Mapping
In-Line Rotating Torque Sensor with On-Board Amplifier
Integrated Surveillance System for Material in the Plutonium Focus Area
In-Situ Bioremediation Using Horizontal Wells
In Situ Electroacoustic Soil Decontamination
In Situ Enhanced Soil Mixing
In Situ Groundwater Treatment System
In Situ Metal Enhanced Abiotic Degradation of Dissolved Halogenated Organic Compounds in Groundwater
In Situ Mitigation of Acid Water
In Situ Remediation of Chromium in Groundwater
In-Situ Remediation System for Groundwater and Soils Using Reactive Fluids
In-Situ Remediation System and Method for Contaminated Groundwater
In Situ Solidification and Stabilization Process
In Situ Steam and Air Stripping
In Situ Steam Enhanced Extraction Process
In Situ Thermal Extraction Process
In Situ Vacuum Extraction
In Situ Vitrification
Infrared Analysis for Process Control of Wastes
Infrared Thermal Destruction
Integrated Spectroscopic Characterization
Integrated Vapor Extraction and Steam Vacuum Stripping
Intelligent Mobile Sensor System for Autonomous Monitoring and Inspection
Interactive Computer Enhanced-Remote Viewing System
Inverting Membrane Borehole Instrumentation
Ion Conduction Agglomeration System
Ion Exchange Mercury Removal From Contaminated Soil
Ion Mobility Spectrometry
Ion Trap Mass Spectrometry
Kelly Vac Wall Attachment
Large Volume Flow Thru Detector System
Lasagna (tm) Soil Remediation
Laser Induced Fluorescence
Laser Metal Emissions Monitor
Laser Surface Cleaning
Laser Spark Spectroscopy Metal Emissions Monitor
Leak Detection Aid
Light Absorption Cell Combining Variable Path and Length Pump
Light Duty Utility Arm
Liquid and Solids Biological Treatment
Liquified Gas Solvent Extraction [LG-SX] Technology
Long-Range Alpha Detection
Long-Term, Post Closure Radiation Monitor
Low-Energy Extraction Process
Low Temperature Thermal Treatment [LT3®] System
MAECTITE® Chemical Treatment Process
Magnetometer Towed Array
Measurement While Drilling (MWD)
Membrane Filtration and Bioremediation
Membrane Gas Transfer in Waste Remediation
Membrane Microfiltration
Membrane Recovery Of Contaminants (MRC)
Membrane Recovery Of Contaminants (MRC)
Metal Analysis Probe [MAP®] Portable Assayer
Metals Immobilization and Decontamination of Aggregate Solids
Metals Release and Removal from Wastes
Methane Enhanced Bioremediation
Methanotrophic Bioreactor System
Method and Apparatus for Data Sampling
Method for Thermal Processing Alumina-Enriched Spinal Single Crystals
Metorex X-MET 2P XRF Analyzer
Microbial Composting Process
Microbial Monitoring
Microwave Solidification
Mobile Workstation for Decontamination and Decommissioning
Minimum Additive Waste Stabilization (MAWS)
Mixed Waste Treatment Process
Mixed Waste Vitrification
Mobile Thermal Desorption System
Model 2 XRF Analyzer
Multiple Forearm Robotic Elbow Configuration
Multi-Staged Electrochemical Oxidation
Multi-Vendor Bioremediation
Near Infrared Spectroscopy
Oleophilic Amine-Coated Ceramic Chip
Online Fiber-Optic Spectrophotometry System
On-Site Anaerobic Bioremediation
On-Site Microbial Filter
On-site Radioactivity Detection
Organics Destruction and Metals Stabilization
Oxidation and Vitrification Process
Oxidation and Vitrification Process
Oxygen Microbubble In Situ Bioremediation
PACT® Wastewater Treatment System
Passive Soil Vapor Extraction
PCB- and Organochlorine-Contaminated Soil Detoxification
PCB D Tech TM Assay (PCBs in Soil)
PCB RaPID Assay TM for PCBs in Soil and Water
PCB Ris TM Soil Test System (Monocional Test)
PCP RaPID Assay® (Pentachlorophenol in Water and Soil)
Penta RISc Test System
Pentachlorophenol RaPID Assay
PhotoCAT Process
Photocatalytic Degradation of PCB-Contaminated Sediments and Waters
Photocatalytic Oxidation with Air Stripping
Photocatalytic Water Treatment
Photocatalytic Water Treatment
Photolytic and Biological Soil Detoxification
Photolytic Destruction for SVE Off-Gases
Photolytic Oxidation Process
Photothermal Detoxification Unit
Photovac S PLUS
Pillared Clay Cesium and Strontium Separation
Pipe Crawler Instrument Carriage
Pipe Crawler With Extendible Legs
Pipe Crawler with Suspension System
Pipe Explorer System
Plasma Arc Vitrification
Plasma Centrifugal Furnace
Plasma Hearth Treatment of Mixed Waste
Plutonium Recovery through Microwave Leaching or Dissolution
Plutonium Stabilization and Packaging
Pneumatic Bioremediation Support
Pneumatic Fracturing Extraction and Catalytic Oxidation
Pneumatic Fracturing/Bioremediation
PO*WW*ER Technology
Polymer Encapsulation of Debris
Portable Acoustic Wave Sensor
Portable Chlorinated Compound Analyzer
Portable Gas Chromatograph
Portable Gas Chromatograph
Portable Hazardous Waste Detector
Portable Liquid Chemical Analyzer (PLCA)
Portable Sensor for Hazardous Waste
Post-Closure Monitoring
Precipitate Hydrolysis Process for the Removal of Organic from Nuclear Waste Slurries
Precipitation, Microfiltration, and Sludge Dewatering
Precision Excavator (PE)
Process for Removing and Detoxifying Cadmium from Scrap Metal
Process Models for Predicting Viscosity and Electrical Resistivity of Glass
PurCycleVapor Treatment Process
PYRETRON® Thermal Destruction
PYROKILN THERMAL ENCAPSULATION Process
Radio Frequency Heated Extraction (RFHE)
Radio Frequency Heating
Radio Frequency Heating
Radioactive Pipe Characterization
Radiological and Hazardous Material Measurement
Rapid Geophysical Surveyor (RGS)
Rapid Optical Screen Tool
Rapid Surface Sampling and Archive Record System
Rapid Transuranic Monitoring Laboratory (RTML)
Reactor Filter System
Recirculation Well Aquifer Remediation
Reconfigurable In-Tank Mobile Robot
Reductive Photo-Dechlorination Treatment
Reductive Thermal and Photo-Thermal Oxidation Processes for Enhanced Conversion of Chlorocarbons
Remote Excavation System
Remote Monitoring and Analysis of Glass Melt Composition
Representative Geometry Phantom
Resonant Sonic Drilling
Road Transportable Analytical Laboratory (RTAL)
Robotic End Effector
Rochem Disc Tube Module System
Salt Distillation
SAREX Chemical Fixation Process
Scentograph Portable Gas Chromatograph
SEAMIST (tm)
Segmented Gate System [SGS]
Self-Checking Liquid Level Sensor
Self-Referencing Spectrophotometric Measurements
Self-Referencing Remote Optical Probe
Seismic Site Characterization
Sensors for Waste Glass Quality Monitoring
Shelf Life Studies
Single Point Groundwater Flow Monitor Based on Diffusion of Salt Out of a Semipermeable Cylinder
Site Characterization Analysis Penetrometer System [SCAPS]
Site Characterization Penetrometer
Site Sampling Planner
Six Phase Soil Heating
Small Volume Sample Transfer Pump
Smelting Lead-Containing Waste
Soil and Sediment Washing
Soil Recycling
Soil Saw
Soil Separation and Washing Process
Soil Washing Plant
Soil Washing System
Sol-Gel Indication (SGI) Program
Solidification and Stabilization
Solidification and Stabilization
Solidification and Stabilization
Solution Assay Monitor for Analyzing Small Samples of 235-U
Solvent Extraction Treatment System
Spectrace X-Ray Fluorescence Analyzer
Spent Ore Bioremediation Process
Steam Enhanced Recovery Process
Steam Injection and Vacuum Extraction-Linear Flow [SIVE-LF] Process
Stimulatory Activity of Tetrachloroethylene for Positive Chemotaxis by Bacteria
Stored Waste Autonomous Mobile Inspector
Subsurface Geophysical Data Fusion
Subsurface Volatilization and Ventilation System [SVVS®]
Supercritical Extraction/Liquid Phase Oxidation
Supercritical Water Oxidation
Supported Particle-Bound Sequestrant (3M/IBC)
Surface Contamination Detector and Recorder
Surfactant-Enhanced Aquifer Remediation
The SABRE Process
The SABRE Process
Thermal Desorption Unit
Thermal Desorption
Thermal Enhanced Vapor Extraction
Triox Ozone Treatment System for Cooling Tower Water
Tunable Hybrid Plasma
Two-Phase Extraction Process
Two-Zone, Plume Interception, In Situ Treatment Strategy
Two Stage Mixed Waste Oxidation and Reduction
Ultrasonic-Aided Leachate Treatment for Mixed Wastes
Ultraviolet Radiation and Oxidation
Underground Seismic Imaging
Underground X-Ray Spectroscope
Unsaturated Flow Apparatus
Use of Microalgae to Remove Pollutants from Power Plant Discharges
UVB - Vacuum Vaporizing Well
VaporSep Membrane Process
Variable Path Length Remote Optical Probe
Video Bar-Code Reader
Volume Reduction Unit
VYTAC 10F
Waste Acid Detoxification
Waste Inspection Tomography (a)
Waste Inspection Tomography (b)
Waste Vitrification Through Electric Melting
WES-Phix Stabilization Process
Wetlands-Based Treatment
X*TRAX Thermal Desorption
X-Ray Treatment of Aqueous Solutions
X-Ray Treatment of Organically Contaminated Soils
XonTech Sector Sampler
ZenoGemProcess
Zero Tension Lysimeters