IDENTIFICATION AND CHARACTERIZATION OF RADIOACTIVELY CONTAMINATED SITES IN UKRAINE AND PLANNING FOR ENVIRONMENTAL RESTORATION ACTIVITIES

Y. SOROKA
Ministry of Energy of Ukraine,
Ukrainian R&D Institute for Industrial Technology,
Zhovty Vody, Ukraine

ABSTRACT

In the Pridniprovsk-Krivoy Rog region uranium, titanium, iron and manganese ores were mined and milled beginning in the 1950s. These activities have caused radioactive contamination of the environment at some sites. In recent times intensive works concerning the surveying of contaminated areas and substantiating the need for remediation have been initiated. The research methodologies applied and the results from radiation surveys are presented for the site of the first uranium mine in the Ukraine, for tailings originating from the Pridniprovsk Chemical Plant (PChP), for the recultivated dump-site of the former "O"-mine, as well as for the wastes, raw materials and production of the Nicopol Ferro-Alloy Plant. The planning procedure for the remediation activities at the town of Zhovty Vody is described.

1. INTRODUCTION

The regions where mining and milling enterprises are located are often considered as locations with technogene impacts on the environment. They are regions of potential pollution by natural radionuclides (NRN) due to the accumulation as industrial wastes on the ground surface, their involvement in the milling processes, and their utilization in some comercial applications. The relevant radionuclides such as uranium-238, thorium-230, radium-226, polonium-210, lead-210, protactinium-231 are classified as the most hazardous pollutants.

From this point of view, the Pridneprovsk-Krivoy Rog region is the most affected in Ukraine. There is a number of large mining and milling enterprises located in the region (Fig. 1):

— VostGOK Mining and Milling Enterprise;
— Industrial Corporation“Alexandria Coal”;
— Verchnedniprovsky Mining and Metallurgical Enterprise;
— Industrial Corporation “Krivbussruda” —;
— Industrial Corporation “Pridniprovsky Chemical Plant”;
— Large Mining & Milling Enterprises (InGOK, SevGOK, CGOK, NKGOK, UGOK).

The VostGOK M&M Enterprise has been active in uranium ore mining and processing for more than 40 years. As a result of its activity, large territories (about 14 km^2) were affected and have been contaminated. The dust from dumps and dry beaches of tailings ponds creates an additional pollution of atmosphere with radon and long lived uranium- and thorium-series radionuclides.

At present, mining for uranium ores is going on in the Kirovograd region of Ukraine at the Smolino and Ingoolsky Mines respectively.

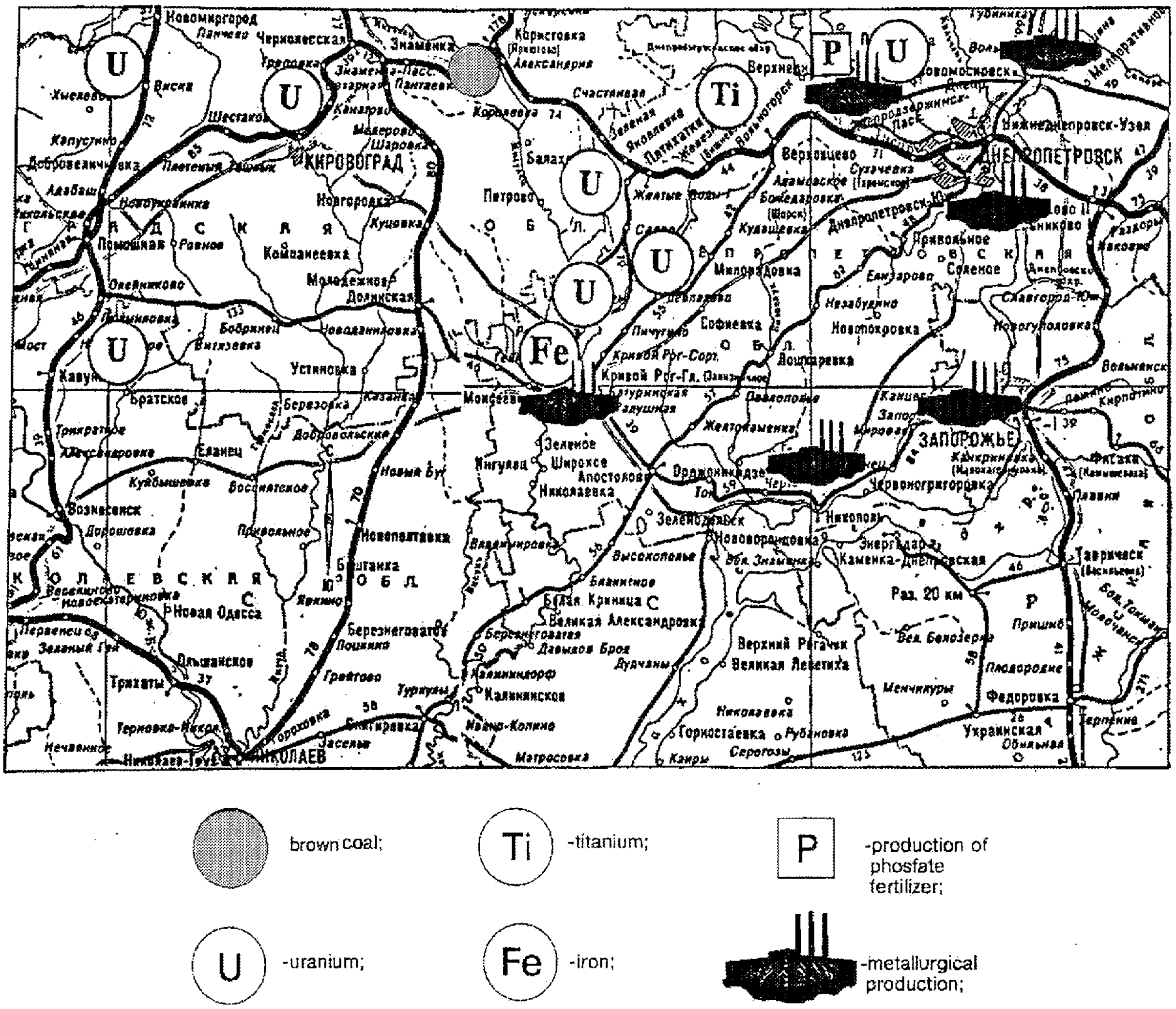

FIG. 1. Mining and ore processing in the region.

In the town of Zhovty Vody the uranium mining in the Novaya Mine has ceased, and the mine is used for the extraction of iron ores only. Uranium ores from Smolino and Ingoolsky Mines are still being processed in a hydrometallurgical plant in Zhovty Vody.

In 1950s and 1960s the mining of uranium ores took place in the Pervomayskaya Mine in the city of Krivoy Rog. After the uranium deposit were exhausted, the activities in the Mine switched to the development of iron ore body. At present, this Mine is also going to be closed.

Besides the traditional mining method, in situ leaching of uranium was employed in Ukraine at the Devladovo and Bratsky sites. They have been closed down and the site have been recultivated.

Uranium ores coming from both Ukraine itself and from abroad were processed between 1947 and 1991 at Pridniprovsky Chemical Plant (PChP) in the city of Dniprodzierzhinsk. When the PChP ceased to operate, many spots with radioactive contamination and the tailings storage facility remained at its former site. At present the plant has been switched to new products (zirconium and gold). The tailings storage facility is being investigated in detail.

2. REGULATORY BASIS

One of the basic documents regulating the work and the activities concerning the decontamination of radioactively contaminated sites is the “Sanitary Regulation for the Liquidation, Conservation and Conversion of Radioactive Ores Mining and Milling Enterprises” [1], established in 1991. Over the last few years new Ukrainian laws have been passed, regulating radiation safety at plants, in urban areas, as well as in the environment in general. These include:

- “On Nuclear Energy Utilization and Radiation Safety” [2];
- “On Human Protection against Ionizing Radiation Impact [3];
- “On Radioactive Waste Management” [4];
- “On Uranium Ore Mining and Milling” [5].

According to [3], an individual dose limit for the population in general must not exceed 1°mSv of effective exposure dose per year. Therefore, the average effective doses for humans belonging to the critical group must not exceed the established dose limits independent of the conditions and the way of the dose received.

In 1998, the new "Radiation Safety Standard of Ukraine" (NRBU-97) [6] has been put into force. It was drawn up on the basis of recommendations of both, the International Commission for Radiation Protection (ICRP) [7] [8] and International Safety Standards [9]. In this connection a transitional period is in force now in the Ukraine, during which relevant implementation regulations are updated and new one are developed.

3. METHODS OF CHARACTERIZATION AND INSTRUMENTATION

To characterize contaminated territories and radioactive wastes, a combination of field and laboratory methods was used.

The field method includes the following:

- dosimetric surveys;
- determination of α- and β-radioactive contamination on surfaces;
- emanation surveys;
- radon emission survey (measuring radon flux density from surface);
- measuring radon and its equivalent equilibrium volume activity, both in the atmosphere and inside dwellings.

For the gamma-radiation dose rate measurement occupational dosimeters and field radiometers were used.

Geiger-Müller counters were used as detectors in occupational dosimeters, and scintillation NaI (Tl) crystals were used as detectors in radiometers. The energy dependence of the dosimeters did not exceed ±2.5% within an energy range between 0.05 and 3 MeV. For the field radiometer this value was up to 60%. For this reason the radiometer was used only for detecting zones with abnormal radiation, and was not used for dose estimation. Both devices were calibrated at a special gauging organization in Zhovty Vody.

Surface contamination was determined with the aid of occupational dosimeters containing alpha and beta detecting removable units.

The measuring range for alpha-radiation was from 0.04 to 400 $Bq{\cdot}cm^{-2}$, and for beta-radiation from 0.04 to 4000 $Bq{\cdot}cm^{-2}$.

Emanation surveys were carried out with alpha-active gas field radiometers. Soil air sampling was carried out by drawing the air into a chamber of 50 cm^3 capacity from a depth of 0.6 m. Analyses took place 3 hours after sampling.

The density of ^{222}Rn flux from the ground surface was measured by gas sorption onto activated charcoal [10]. Special cartridges with activated charcoal discs were placed on the surface to be studied and exposed to it. After exposure, the charcoal was analyzed by gamma-spectrometry. The minimum measurable value of ^{222}Rn flux was 2 $mBq{\cdot}m^{-2}{\cdot}s^{-1}$.

For measuring the volumetric activity of ^{222}Rn in the air of houses a classic scintillation method (with a Lucas cell) was used [11]. The device consists of set of scintillation chambers, each with volume of 500 cm^3. The air was sampled by pumping it through a series of inter connected aerosol filters, a dryer and the chamber. Aerosol sampling was performed through the filter. Aerosol filters were made of ultra-thin perchlorovinyl fibres. The decay coefficient of these filters is 95% for Rn progeny. Markov's method was implemented using a low level alpha-counter and a portable air pump with a throughput of 20 l/min [12]. The alpha-counter detector includes ZnS plates and a photo-multiplier. The efficiency of the total alpha-irradiation registration by the detectors was equal to 43%:

$$\text{Efficiency of alpha irradiation registration} = \frac{\text{Count rate of standard source } [s^{-1}]}{\text{Activity of standard source [Bq]}}$$

The background reading of the counter did not exceed eight counts per hour. The detection limit for ^{222}Rn was 18.0 $Bq{\cdot}m^{-3}$ with a relative error of ±30%. In some cases, measurements were carried out with an "Alpha GUARD" (Germany) radon-monitor. Calibration of the devices for ^{222}Rn and its measurement of its progeny was performed with a standard Radon-chamber of 18 m^3 volume, which was located a special gauging organization in the town of Zhovty Vody.

Methods using field gamma-spectrometry with HPGe-detectors are still not widely used in Ukraine because of lack of domestically produced instruments, and of uncertainties connected with interpretation of results obtained by this method.

The following laboratory methods were used to determine the natural radionuclides (NRN) content in radioactive wastes, contaminated soils, raw materials and industrial wastes:

- radiometric measurements of total alpha-activity in samples;
- radiochemical analysis of NRN content;
- low-background gamma-spectrometric analysis of NRN content.

Radiometric measurement of total alpha-activity in environment samples were used for fast assessment of a site for alpha-emitting nuclides contamination. Two modifications of this method were available:

- measurement of sample alpha-activity in "thick" layer;
- measurement in scintillator layer.

Measurements of ^{238}U concentrations in soil and wastes were carried out in the laboratory using radiochemical methods with photometric detection.

The contents of ^{226}Ra, ^{232}Th, ^{40}K were determined by a gamma-spectrometric method with low-background gamma-spectrometer. The content of ^{210}Po was determined by a radiochemical method with radiometric detection using a low-background alpha-counter. Concentration of ^{210}Pb were determined by both methods: a radiochemical one with radiometric detection using a low-background beta-counter, and a spectrometric one with low-background gamma-spectrometer using a high-purity germanium detector. The relative errors of measurements (at the 95% confidence interval) were ± 20% for the radiochemical methods, and did not exceed ± 25% for the gamma-spectrometric and radiometric methods.

4. RADIATION SURVEY EXPERIENCE

4.1. Radiation survey of the “Pervomayskaya” Mine area

The first uranium ore mining enterprises in Ukraine was the “Pervomayskaya” Mine. Uranium ore mining began in the late 1940s at one of its sites (“2/6”). The mining activities were extended by opening the mines “Obyedinennaya” and “Severnaya-Ventilation” of the same enterprise. In the late 1960s, uranium ore mining ceased, but at “Obyedinennaya” mining for iron ore continued. In early 1998 the enterprise also ceased to mine the iron ore and transferred into liquidation.

Over the past 30 years all surface buildings of the mine "2/6" have become unused and derelict, as is the case for almost all main and auxiliary buildings and installations of the "Severnaya-Ventilation" mine (Fig. 2). The total area of the mining enterprise territory to be rehabilitated is 68.5 ha. The enterprise activities have resulted in contamination of environment, particularly from the following: scattered ore; natural radionuclides leaching from the ores; uncontrolled utilization of wastes for other purposes; dispersion of dusts etc. In the course of time, the ores and soil have been intermingled thus increasing the concentration of natural radionuclides in soils.

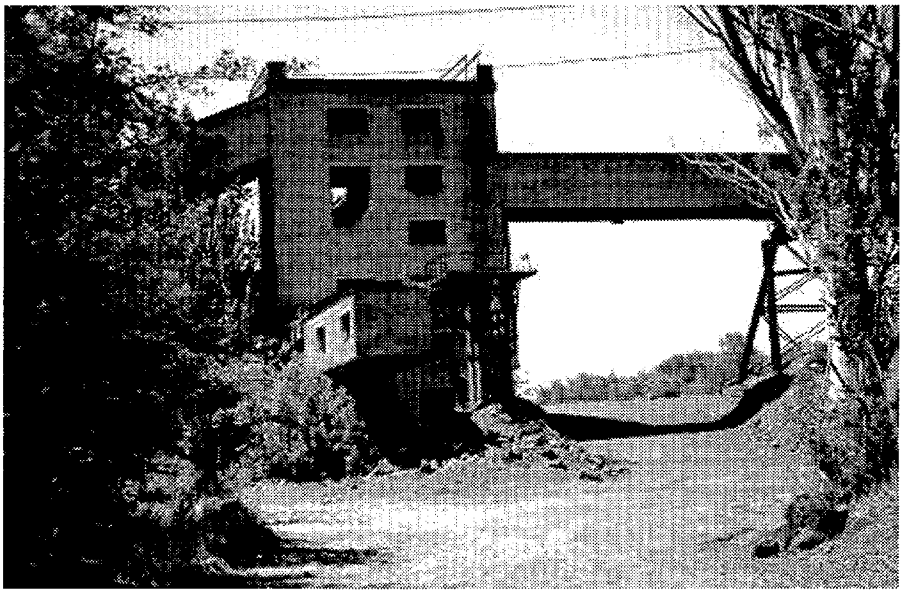

FIG. 2. Derelict ore processing building at the "Severnaya-Ventilation" Mine.

In order to assess the sites' contamination, a comprehensive range of characterization methods have been applied. Initially, dosimetric gamma-surveys were carried out, which helped to reveal and to contour in general occurrences of contamination. Then followed a radiometric determination of surface alpha- and beta-contamination. The same method was used to distinguish between contaminated and non-contaminated mine equipment, as well as construction materials. Radon emanation and radon emission surveys have allowed to detect contamination sources located at shallow depths (0.4 ÷ 0.5 m), which had not been identified with gamma-surveying.

Laboratory methods have shown the total alpha-activity of the mixture ore and soil ranges from 813 Bq/kg to 4.06×10^5 Bq/kg, radium-226 ranges from 110 to 5.35×10^4 Bq/kg (see Table I). At soil spots with radioactive contamination one can notice a considerable amount of radon exhalation to atmosphere, as well as increased radon levels in the soil air (Table II). Exhalations exceeded background values by 1.5–140 times over contaminated spots, while the radon concentrations in the soil air at abnormal spots are 40–815 times higher than background values.

Applying both field and laboratory methods for the characterization of affected areas has allowed to determine the contamination parameters and to develop respective recommendation for the remediation of the land.

TABLE I. NATURAL RADIONUCLIDE CONTENTS IN SAMPLES OF ORE AND SOIL DUST, SILT, AND DETRITAL SOIL MIXTURES TAKEN AT THE "OBYEDINENNAYA", "2/6", AND "SEVERNAYA-VENTILATION" MINE SITES

Sample number	**Sampling place and sample type**	**Natural radionuclide contents [Bq/kg]**			**Total specific alpha-activity [Bq/kg]**
		^{232}Th	^{226}Ra	^{40}K	
"2/6" Mine					
1	Ore loading site, ore and soil mixture	10	239	244	6340±610
2	Ore loading site, ore sample	–	53500	–	$(4.06\pm0.05)\cdot10^5$
4	Ore loading site, ore and soil mixture	14	705	307	7190±690
"Obyedinennaya" Mine					
5	Abnormal spot 1/1, ore and soil mixture	18	753	240	6750±610
"Severnaya-Ventilation" Mine					
17	Left sump. Depth 0.5 m. Silt	28	110	494	813±190
22	Dust from gallery #7	25	2412	1441	$(2,47\pm0.08)\cdot10^4$
26	Abnormal spot 3. Depth 0.1÷0.3 m. Detrital soil	25	601	542	6811±850
39	Sample of background soil	15	20	169	588±176

TABLE II. EMANATION AND EXHALATION MEASUREMENTS ON THE TERRITORIES OF "2/6" AND "NORTHERN VENTILATION" MINES

Sampling point	Sampling location	Radon exhalation [$mBq \cdot m^2 \cdot s^{-1}$]	Radon concentration at a depth of 0.5 m [$kBq \cdot m^{-3}$]
"2/6" Mine			
12	Abnormal spot 1	355	50.5
13	Abnormal spot 1	459	48,4
14	Abnormal spot 2	1850	459
15	Boundary of abnormal spot 1	93	–
16	Abnormal spot 1	801	–
17	Abnormal spot 2	4050	–
18	Abnormal spot 3	614	–
"Northern Ventilation" Mine			
5	Temporary storage of uranium ore mining wastes	860	960
8	Temporary storage of uranium ore mining wastes	169	–
21	Left sump of mine cleaning water system	43	0.18
19	Background value	28	1,18

4.2. Wastes, raw materials and production of the Nicopol Ferro-Alloy Plant

Steels are alloys of iron with manganese, chromium, silicon etc. One possible raw material for the ferroalloys production are ores which already are naturally enriched with the oxides of the alloying constituents and processed ore concentrates. In some cases these may also contain increased concentrations of natural occurring radioactive materials, for the genesis of many ore deposits resulted in radioactive elements being present as accessories to the main elements being mined.

The Nikopol Ferroalloy Plant (NFP) utilizes raw materials delivered from a range of deposits in the Ukraine, Russia and other countries. In terms of total amount, the production share of ferroalloys with manganese is around 50%, and the share of those with silicon is around 30°%. Table III shows laboratory analysis results for samples of both raw materials and products of the NFP. The data presented give evidence of high contents of natural radionuclides in rutiles, zircons and baddeleyites. In the remainder of the raw materials the natural radionuclides content is at background values levels. Concentrations of all radionuclides are almost equal to background values in products, except for flux AN-65-U. The increased ^{226}Ra concentration in the flux AN-65-U is explained by utilizing rutile and zircon for its preparation.

Solid production wastes (casting slag, granulated slag) containing ^{226}Ra, ^{232}Th, and ^{40}K can be used with restrictions for the production of constructing materials. Slimes, i.e. wastes from de-dusting and gas scrubbing systems, are stored in a slime sump.

TABLE III. NATURAL RADIONUCLIDES CONTENT [Bq/Kg] IN RAW MATERIALS, PRODUCTS AND WASTES OF THE NIKOPOL FERROALLOY PLANT.

Type of sample	^{238}U	^{226}Ra	^{232}Th	^{40}K	^{210}Pb	^{210}Po
Raw materials						
Rutile (Ukraine)	169±62	266±32	27±6	70±9	620±100	450±50
Zircon (Australia)	590±120	694±190	1150±420	600±80	4580±310	8240±400
Zirkon (New Zealand)	3110±800	9990±200	1100±400	500±30	6800±800	8941±400
Baddeleyite concentrate (Russia)	2870±370	117-±130	307±130	-	1080±140	1150±60
Manganese ore (Ukraine)	87±30	78±28	8±3	179±90	-	188±50
Boron slag	2,5±2	12±6	7±2	180±82	1074±300	73±30
Products						
Flux AN-65-U	-	374±70	80±30	20±02	-	61±25
Flux AN-60	-	51±18	33±12	149±46	-	39±13
Flux AN-348-V	-	48±10	16±5	159±46	-	51±20
Flux AN-47	-	224±10	42±6	111±30	210±60	189±82
Flux AN-67-A	22±8	48±8	15±4	61±15	450±92	20±4
Wastes						
Casting Slag (silicomanganese)	-	194±28	35±12	607±128	-	-
Granulated slag (silicomanganese)	-	135±42	25±10	477±122	-	-
Granulated slag (ferromanganese silicon)	-	161±30	30±10	617±130	-	-
Slime (slime storage 1)	24±8	65±40	24±6	1206±300	6011±200	3200±1300
Slime (slime storage 2)	-	66±32	7±2	1250±300	2100±650	1420±400
Slag of sintering shop 1	205±68	154±42	26±8	513±160	-	191±32

The investigation of the slime storage gamma signals have shown no considerable increases over background values. To the contrary, laboratory analyses have identified high total alpha-activities of the slime samples. More detailed laboratory investigations, using radiochemistry techniques and low-background spectrometry with a HPGe-detector have shown that the slime is rich in both ^{210}Pb and ^{210}Po (Table III). A considerable shift of equilibrium between uranium decay-chain radionuclides in the slime bears witness to the enrichment of trans-radon chain elements in the wastes due to the technical process (involving high temperature treatment) compared to their background concentrations in raw materials. Thus, the method of choice for detecting and characterizing radioactive contamination at such facilities is to measure total alpha-activity in the samples, rather than gamma-dosimetric surveying.

According to the standards under development [13], slime are considered as slightly radioactive wastes. In this connection it will be clamed special requirement during recultivation activities.

4.3. Preliminary results of radiation surveys on the capped tailings storage of Pridniprovsk Chemical Plant.

There are capped tailings on the site of the PChP with an surface area of about two hectares. They have been created during the 1950s and do not conform to the present standards and regulations for radioactive wastes management. The tailings resulting from uranium ores processing have a thickness of 12.5 to 15.5 meters and are covered with loess loam layer. About 770,000 tones of tailings with a total activity exceeding 0.2 PBq are stored here. The tailings surface is covered with turf, but the slopes might still be subject to erosion.

Gamma-radiation dose rates on the site of the tailings impoundment fluctuate between 0.14 to 7.29 μSv/hour. Along the perimeter of the tailings impoundment contaminated spots have been found, where gamma-radiation dose rates reach a value of 15.76 μSv/hour. The ^{226}Ra content in the wastes varies from 789 to 1.72×10^6 Bq/kg (Table IV). The concentrations of ^{226}Ra in surface water samples taken from locations in the vicinities of tailings impoundment site range from 24 to 99 mBq·dm^{-3}, while in the Dnjepr-river and in groundwater they range from 8.5 to 18.5 mBq·dm^{-3}. Values of ^{222}Rn flux from the surface of tailings impoundment, where covered by a protective layer, is around 0.32 to 1.5 Bq·m^{-2}·s^{-1}, while radon exhalation background levels are only around 9 to 18 mBq·m^{-2}·s^{-1}.

On the basis of the results from the completed research, recommendations have been developed concerning the improvement of the tailings capping.

TABLE IV. CONTENT OF NATURAL RADIONUCLIDES IN WASTES OF PChP

Sample number	**Concentration [Bq/kg]**	
	^{226}Ra	**^{232}Th**
342	39330	-
358	27230	5169
324	3888	-
84	2738	54
38	789	25
189	1.72×10^6	-
53	69970	-
330	1.32×10^5	-
157	92080	-

4.4. "Olkhovskaya" mine recultivated territory radiation survey

At the end of the 1990s one of the first projects for restoration of contaminated land was realized at the former uranium ore mine "Olkhovskaya" site [14]. The work included the sorting of the waste rock on the site and the sanitation of contaminated land. From the waste rocks sorted, such parts containing rich ores were processed at the hydrometallurgical plant, the remainder was removed to "Novaya" mine. After rehabilitation post-remediation monitoring was performed, including:

- dosimetric surveys;
- emanation surveys;
- radon emission surveys;
- soil sampling and analysis for natural radionuclide contents.

In general, measured values of gamma-radiation equivalent doses rate on the remediated land is within 0.1 to 0.22 μSv/hour, which is in the range of natural background radiation of the region.

Emanation surveys were conducted for determining and contouring residual contamination occurring at depth and not detected by gamma-surveys. Minimal values of soil radon content were observed in the middle of the site. This can be explained by the blanketing of the soil under "Olkhovskaya" mine waste rock. On the major part of the site, the average radon concentration in the soil was within $10\text{-}20 \times 10^3$ Bq m^{-3}, which conforms to the background values of the regions. Maximum values of soil radon content were observed at southern part of the site, where the concentrations exceeded 25–50 times background values. Abnormalities determined, in general, are caused by the deposition and accumulation of radionuclides in topographical depressions (ditches, hollows etc.).

At the most conspicuous points of the site under survey, the radon exhalation from soil was measured. Five measurements at three points gave a background value of 15–25 $mBq{\cdot}m^{-2}{\cdot}s^{-1}$. In the centre of abnormal point 1, determined during the emanation survey, the radon exhalation reaches 553 $mBq{\cdot}m^{-2}{\cdot}s^{-1}$. The same situation is observed at abnormal point 2.

To specify contamination parameters at the abnormal points, boreholes have been drilled and soil samples have been taken. Results of radiation surveys at abnormal Points 1 and 2 are shown in Table V. At abnormal Point 1 soil contamination extends down to a depth of four meters, and is caused by leachates penetrating into the ground.

TABLE V. RESULTS OF RADIATION SURVEYS

No. of abnormal spot	**Radon exhalation [$mBq{\cdot}m^{-2}{\cdot}s^{-1}$]**	**Rn conc. in soil [$kBq{\cdot}m^{-3}$]**	**Sampling depth [m]**	**Soil description**	**Concentration in soil [$Bq{\cdot}kg^{-1}$]**		
					^{238}U	**^{226}Ra**	**^{232}Th**
Abnormal 1	553	579	0.0 - 0.25	Grey poured soil	37	34	38
			0.25 - 0.5	Grey poured soil	40	37	45
			0.5 - 0.75	Grey poured soil	45	56	57
			0.75 - 1.0	Grey poured soil	45	61	46
			1.0 - 1.5	Black loam	626	568	57
			1.5 - 2.0	Light-brown loam	248	146	35
			2.5 - 2.8	Light-brown loam	422	208	40
			3.5 - 4.0	Pale-yellow loam	434	42	33
			6.5 - 6.8	Brown loam	26	37	30
Abnormal 2	313	136	0.0 - 0.2	Poured loam	103	79	71
			0.2 - 1.4	Rocks soil	868	735	58
			1.4 - 1.6	Buried soil	75	35	32
			2.0 - 2.5	Light-brown loam	47	38	37
			3.5 - 4.0	Brown loam	33	34	33
			5.3 - 6.0	Brown loam	40	33	30

At abnormal Point 2 no significant contamination of the soil by natural radionuclides was noticed. The elevated values of radon content in the soil were caused by an one-and-half meter layer of waste rock. Following the survey results, some additional remediation measures at the contaminated spots were implemented.

4.5. Radioactive situation in the town Zhovty Vody

In the town of Zhovty Vody (Fig. 3), as in many of similar such settlements (for instance as in Grand Junction, USA), in the first studies of industrial zones, the harmful influences of low active wastes have being underestimated [15]. It has led to uncontrolled use of the dumped rocks for commercial activities of the population. This in turn led to serious contamination of some parts of the town. In the 1980s decontamination was organized for some parts of the town, but it appears not to have been sufficient; at present the average gamma- background on the area of the town is:

- for 88% of the area between 0.16 and 0.4 μSv/hour;
- for 8% of the area between 0.4 and 1.5 μSv/hour;
- for 3% of the area between 1.5 and 2.5 μSv/hour;
- for 1% of the area above 2.5 μSv/hour.

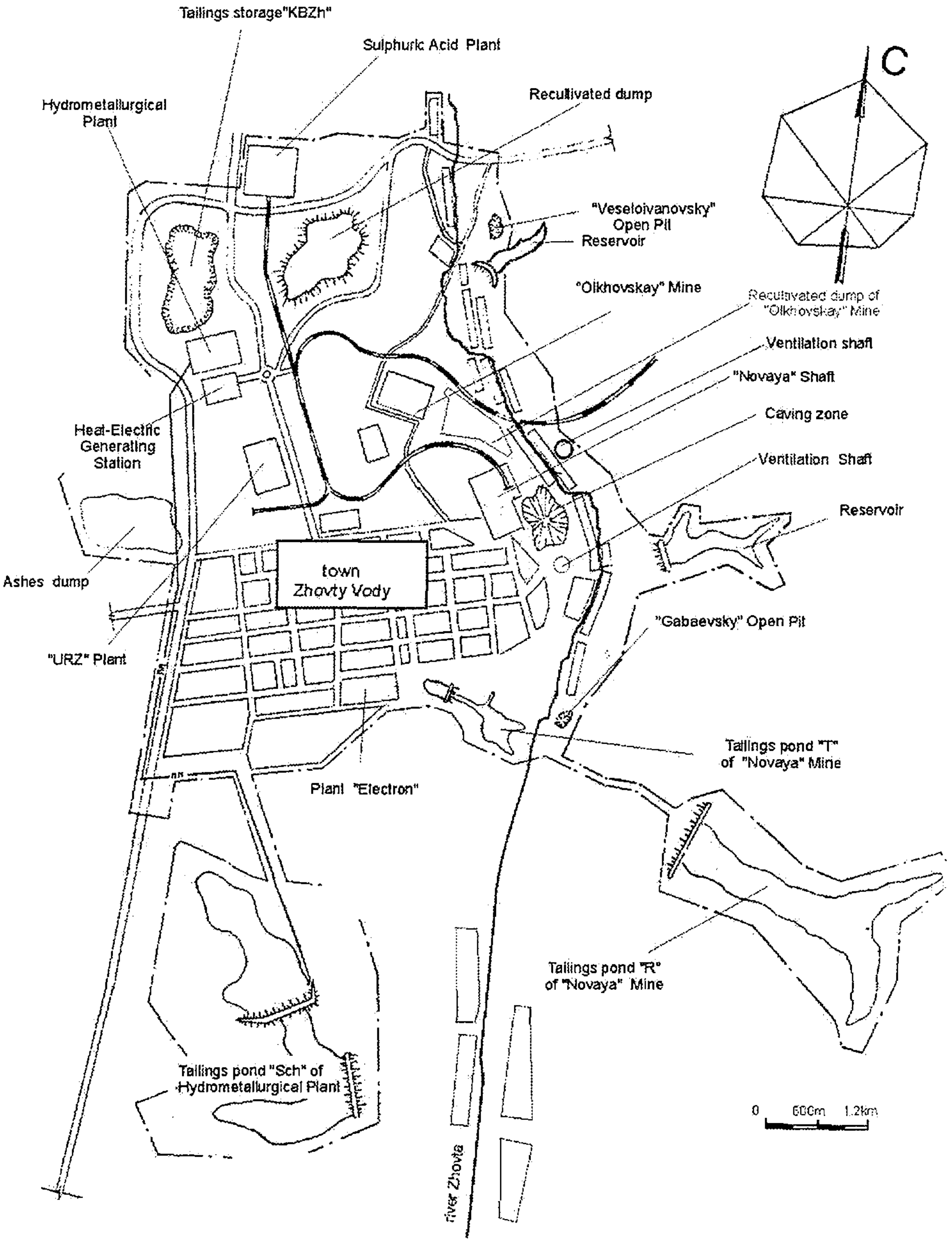

FIG. 3. Location of objects of potential ecological hazard in Zhovty Vody.

In radiation surveys more than 5,400 abnormal locations and places, where the gamma-radiation exceeds 1.2 μSv/hour, have being detected. At 460 locations the gamma dose rate exceeds 10 μSv/hour, and at another 57 locations it is higher than 30 μSv/hour. All these irregularities are related to rocks which contain elevated levels of natural radionuclides.

The abnormalities according to their type are distributed as follows:

- in house foundations — 1334 (= 25% of the total number of observed) abnormalities;
- roads and squares surface coverings — 2440 (= 44%) abnormalities;
- isolated spots — 1253 (= 23%) abnormalities
- sites with undefined localization — 430 (= 8%) abnormalities.

In 1994, two thousand measurements of the radon-gas equivalent equilibrium concentrations (EEC) in the living spaces of the town were performed. For 14% of all houses the EEC of radon was 100 to 200 Bq/m^3, for another 19% it was between 200 and 1000 Bq/m^3, and for 3.5% in excess of 1000 Bq/m^3.

5. THE PLANNING OF REMEDIATION WORK (USING THE EXAMPLE OF ZHOVTY VODY TOWN)

5.1. Choice of method

An important element of the planning for remediation work at a contaminated site is the choice of the optimum strategy of the implementation of the work. According to standards in force at present [6], both radiation safety and protection against occupational exposure are built on following principles:

- any practice involving the exposure of people must not take place if its benefit for the people being exposed and for society as a whole is less than harm (principle of justification);
- exposure dose levels from all types of practice must not exceed established dose limits (principle of non-exceedance);
- individual dose levels or the number of individuals being exposed, regardless of the source of irradiation, must be as low as can reasonably be achieved, taking into account economical and social factors (principle of optimization).

The above principles were used when planning the cleanup measures for radioactive contamination on the territory of Zhovty Vody town.

5.2. Determination of norms for urban residual contamination.

Norms for permissible urban residual contamination are determined by starting from the exposure of critical population groups according to following general steps:

- external exposure;
- exposure from Rn and it daughters by inhalation;
- exposure from long lived natural radionuclides in dust taken up by organisms.

No indigestion path for long lived natural radionuclides was considered because of its insignificance under the conditions at Zhovty Vody. Critical groups include the following three categories of population living on land with radioactive contamination:

- infants;
- children under 17 years of age;
- adults.

A background value of 20 μR/hour is accepted as gamma dose rate on the town's territory. An average value of 60 μR/hour is accepted as gamma dose rate on contaminated sites.

For the purpose of assessing the impact of doses on the general population caused by radioactive site contamination three values of gamma dose rates (30, 40, 60 μR/hour) are defined according to international standards [9] [16]. The results of the estimations are shown in Table VI. Taking a 30 μR/hour residual gamma dose rate from a contaminated site, the dose limit for member of the public will not be exceeded when a recommended dose margin coefficient of 2 is used [13]. By this, a certain dose safety margin for the impact from the contamination on the town area (Hydrometallurgical Plant, "Novaya" Mine, "Sch"-tailings storage) is provided.

The above accepted criteria (≤30 μR/hour) are in accordance with existing national and international ones:

- the ^{226}Ra concentration in soil shall not exceed 150 Bq/kg, which conforms to the criterion of a specific alpha-activity in soil equal 1200 Bq/kg, which is accepted for remediation by way of afforestation;
- the average Rn volume concentration over a given site shall not exceed 18.5 Bq/m^3.

TABLE VI. ANNUAL EFFECTIVE DOSES FOR CRITICAL GROUPS OF THE POPULATION AS CAUSED BY RADIOACTIVE SITE CONTAMINATION AND BASED ON THREE DIFFERENT GAMMA DOSE RATES.

Population category	**Exposure paths**	**Annual effective doses [mSv/year] for gamma dose rates [μR/hour] of**		
		30	**40**	**60**
Infant	External ionizing irradiation	0.142	0.284	0.568
	Radon and its decay products	0.107	0.232	0.357
	Re-suspended dusts	$1.7\cdot10^{-5}$	$2.1\cdot10^{-5}$	$2.9\cdot10^{-5}$
	Total	0.249	0.516	0.925
Children less then 17 years old	External ionizing irradiation	0.122	0.244	0.488
	Radon and its decay products	0.107	0.232	0.357
	Re-suspended dusts	$4.2\cdot10^{-5}$	$5.2\cdot10^{-5}$	$7.2\cdot10^{-5}$
	Total	0.229	0.476	0.845
Adults	External ionizing irradiation	0.113	0.226	0.452
	Radon and its decay products	0.107	0.232	0.357
	Re-suspended dusts	$3.9\cdot10^{-5}$	$4.7\cdot10^{-5}$	$6.7\cdot10^{-5}$
	Total	0.23	0.478	0.809

5.3. Estimation of the efficacy of remediation measures

The exposure doses to the population from radioactive site contaminations are estimated assuming the following conditions [16]:

- a dose coefficient, which is the ratio of effective dose rate and absorbed dose rate in air, of 0.74 for adults, 0.8 for children, and of 0.93 for infants;
- staying outdoors for 20% of the total time.

In Table VII the results of estimations for the effective dose from the soil on the town population are given.

TABLE VII. EFFECTIVE DOSE ON POPULATION (μSv/year) FROM DIFFERENT LEVELS OF EXTERNAL GAMMA DOSE RATE AT THE AREA.

Category of population	**Exposure dose rate [μR/hour]**				
	20	**30**	**40**	**50**	**60**
Infants	284	426	568	710	852
Children less than 17 years old	244	366	488	610	732
Adults	226	339	452	565	678

The average weighted population distribution in the town is as follows:

— infants	— 570 individuals;
— children less then 17 years old	— 11200 individuals;
— adults	— 48230 individuals;
— total	— 60000 individuals.

The weighted average dose E_a^n on every category of population is calculated according to Equation 1:

$$E_a^n = \frac{E_b^n \times (S_b - S_r) + E_r^n \times S_r}{S_b} \qquad (1)$$

where

- E_b^n is the effective dose for category n of population residing on town land with a background value of gamma dose rate of 20 μR/hour;
- E_r^n is the effective dose for category n of population residing on contaminated land with an average gamma dose rate as 60 μR/hour;
- S_b is the total area of non builtup urban are in ha, i.e. S_b = 643 ha;
- S_r is total area of radioact ive contaminated sites in ha, i.e. S_r = 51,3 ha;

From the above calculations it has been estimated, that the weighted average effective doses are:

— for infants	— 330 μSv/year;
— for children before 17 years old	— 282 μ Sv/year;
— for adults	— 262 μSv/year.

The results of estimating the collective dose for the town's population under different remediation dose rate targets are shown in Table VIII.

As the calculations show, in case of remediation down to the background values of gamma-radiation exposure dose rates, a decrease (from 15.982 man Sv) of the collective effective dose of 2.188 man Sv will be achieved.

In case of partial decontamination to 30 μSv/year of an average gamma-radiation exposure dose rate, the decrease will be 1.631 man Sv, and accordingly for 40 μSv/year it will be 1.079 man Sv.

TABLE VIII. COLLECTIVE EFFECTIVE DOSE FOR THE POPULATION FROM EXTERNAL GAMMA-IRRADIATION [man Sv]

Population category	Annual dose [mSv/year] for average of gamma dose rates [µR/hour] of			
	Background	**30**	**40**	**60**
Infant	0.162	0.175	0.183	0.188
Children less than 17 years old	2.732	2.842	2.950	3.158
Adults	10.9	11.334	11.770	12.636
Total	**13.794**	**14.351**	**14.903**	**15.982**

In addition to the exposure from gamma-radiation sources, a contribution to the total dose to the population is received from Rn and its daughters, as well as by re-suspended dusts. Table IX gives values for collective effective doses assuming background values for gamma dose rate and residual value of gamma dose rate from contaminated land of 30, 40 and 60 µR/hour respectively.

TABLE IX. COLLECTIVE EFFECTIVE DOSE TO TOWN POPULATION FROM ALL SOURCES ON LANDS WITH RADIOACTIVE CONTAMINATION [man Sv].

Sources of impact to population	At background γ-dose rate	At average residual value of γ-dose rate [µR/hour]		
		30	**40**	**60**
External exposure	13.794	14.351	14.903	15.982
Rn and its daughters	4.255	6.134	7.112	8.718
Re-suspended dust	-	0.002	0.0037	0.004
Total	**18.049**	**20.487**	**22.0187**	**24.704**

Remediation measures should result in a reduction of the dose for a long period of time. Radiation detriment V_c can be expressed in terms of cost [17] according to:

$$V_C = \alpha \cdot S_E, \qquad (2)$$

where α is the cost of exposure (cost of collective dose unit), a used in radiation protection; in the present case α =10,200 USD/ man Sv (the exchange rate for Ukrainian Hrivna to US dollar was at the time of this work 1 USD= 2 Hrivna); S_E is prevented collective effective dose in man Sv.

The cost of implementing the remediation measures will be less than the cost incurred from the radiation damage. As a result, the expenditure on the remediation measures is expected to give a net “profit” for a long time period. The ICRP recommends [17] time periods of 70 years for children and 50 years for adults to use in assessments. Table X gives calculated values for radiation detriments, prevented as a result of implementing remediation measures, for different scenarios.

The calculations show that conducting remediation measures prevents radiation detriments for one year from 27394 to 67861 USD/year, depending on the assumed/targeted residual gamma-dose rate.

TABLE X. MONETARIAN VALUE [USD] OF PREVENTED RADIATION DETRIMENTS FOR DIFFERENT RESIDUAL GAMMA DOSE RATES AND DIFFERENT PERIODS OF AMORTISATION.

Critical Group	**Reduction of γ-dose rate to background**		**Reduction of γ-dose rate to 30 μR/hour**		**Reduction of γ-dose rate to 40 μR/hour**	
	for 1 year	**for whole period of optimization**	**for 1 year**	**for whole period of optimization**	**for 1 year**	**for whole period of optimization**
Infants	697	48790	383	26775	206	14455
Children under 17 years of age	12857	900025	8151	570605	5185	362915
Adults	54307	2715350	34471	1723550	22003	1100150
Total	**67861**	**3664165**	**43005**	**2320930**	**27394**	**1477520**

5.4. Estimation of optimum dose reduction to population

Optimization of radiation protection is performed in the cases, when individual doses are less than the dose limits for members of the public (1 mSv/year). The calculations for selecting a strategy for optimizing the implementation are based on [6]:

$$R < \frac{V_C - X}{\alpha}, \qquad (3)$$

where X is the expenditures [in USD] for carrying out the remediation measures, and R is the collective risk of stochastic effects arising from the exposure:

$$R = r_E \cdot S_E, \qquad (4)$$

where r_E is the risk coefficient for a cancerous decease arising with mortal as well as not mortal outcome and for serious hereditary effects arising. For a population r_E is 7.3×10^{-2} Sv^{-1}.

Calculated values for the optimization of relevant remediation measures are presented in Table XI.

The optimization calculations performed show that the maximum return of investments should be achieved when carrying out the remediation measures until obtaining a residual gamma-irradiation rate of 30 μR/hour. Remediation of the land down to background values is not efficient.

6. CONCLUSIONS

When investigating contaminated land, it is important to select an optimum range of methods allowing a detailed characterization of the site.

The main methods for characterizing contaminated sites are:

- dosimetric surveys;
- radiometric fast measurements of total alpha-activity in sample;

- emanation surveys;
- radon emission survey (measuring the density of radon flux from surface);
- measuring radon and its equivalent equilibrium volume concentration in houses and in the ambient atmosphere;
- soil sampling and analysis for concentrations of natural radionuclides.

TABLE XI. CALCULATION RESULTS CONCERNING OPTIMIZATION OF REHABILITATION MEASURES CONDUCTING UNDER DIFFERENT SCENARIOS

	γ-dose rate reduced to background	**γ-dose rate reduced to 30 μR/hour**	**γ-dose rate reduced to 40 μR/hour**
Prevented detriment V_c, [USD]	3664165	2320930	1477520
Calculated value of expenditure for rehabilitation measures X [USD]	3950000	1100200	569720
Calculated index $(V_c - X)/\alpha$	-28	120	89
Collective risk R	26.23	16.61	10.58

As the experience from applying these methods shows, no single one can be used separately for characterizing the contaminated land. In any given case it is necessary to apply a certain set of methods, depending on the extend of expected contamination and radionuclides involved.

According to results from this work it is expedient to perform efficacy analyses before and during implementation of remediation measures.

The analyses were performed in the context of planning for remedial actions on the contaminated areas in the town of Zhovty Vody. From these it was concluded that it would be efficient to clean the land down to a residual gamma dose rate of 30 μR/hour. Under those conditions the maximum effect will be obtained from the remediation activities without exceeding dose limits for the population.

Participation in the CRP made it possible to learn from international approaches to the identification and characterization of radioactively contaminated sites. The experience gained will be used both for the design of a new regulatory basis for the remediation of the contaminated sites and for carrying out remediation measures themselves.

REFERENCES

[1] USSR MINISTRY OF HEALTH, Sanitary regulation for the liquidation, conservation and conversion of radioactive ores mining and milling enterprises, Moscow (1991).

[2] LAW OF UKRAINE, “On Nuclear Energy Utilization and Radiation Safety”, Kiev (1995).

[3] LAW OF UKRAINE, “On Human Protection against Ionizing Radiation Impact”, Kiev (1998).

[4] LAW OF UKRAINE, “On Radioactive Waste Management”, Kiev (1995)

[5] LAW OF UKRAINE, “On Uranium Ore Mining and Milling”, Kiev (1997).

[6] HEALTH MINISTRY OF UKRAINE, Radiation Safety Standards (NRBU-97), Kiev (1997).

[7] INTERNATIONAL COMMISSION FOR RADIATION PROTECTION, Radiation Protection, ICRP Publication 60, Pergamon Press, Oxford (1990).

[8] INTERNATIONAL COMMISSION FOR RADIATION PROTECTION, Protection Against Radon-222 at home and at Work, ICRP Publication 65, Pergamon Press, Oxford (1993).

[9] INTERNATIONAL ATOMIC ENERGY AGENCY, International Basic Safety Standards for Protection Against Ionizing Radiation and for the Safety of Radiation Sources, IAEA Safety Series No.115, IAEA, Vienna (1996).

[10] MOLCHANOV, A. et al., A Method for the Direct Measurement of Radon and Thoron Exhalation from Emanating Surfaces, Tehnicheskiy progress v atomnoy promyshlennosty, Seria gorno-metallurgicheskoye proizvodstvo. 5 (1990) 14–16 (in Russian).

[11] LUCAS H.F. “Alpha scintillation radon counting”, Methods for Measuring Radiation in and around Uranium Mills, (Proc. Workshop Albuquerque, NM, August 1977, Harvard, E.D., Ed.) Atomic Industrial Forum, Washington, DC (1977), 278–287.

[12] MARKOV, K.P., RYABOV, N.V., STAS, K.N., A rapid method for estimating the radiation hazard associated with the presence of radon daughter products in air, Atomic Energ. 12(4) (1962) 135.

[13] HEALTH MINISTRY OF UKRAINE. General sanitary rules of work with ionizing irradiation sources in ukraine. Draft. Kiev, (1998).

[14] KOSHIK, Y.I., "Solution of Ecological Problem for Uranium mining and milling in Ukraine", Report at the Third Workshop within IAEA Technical Co-operation Project RER/9/022 on Environmental Restoration in Central and Eastern Europe in Zhovty Vody (1996).

[15] SOROKA, Y.N., MOLCHANOV, A.I., ISAYEVA, N.G., BEDNARIK, O.N., "The rehabilitation program for territory of town Zhovty Vody after 40 years of uranium ores output", Sixth international conference on radioactive waste and management environmental remediation, ICEM'97, (Proc. of conf. in Singapore, 12-16 October 1997), ASME, New York (1997), 773–776.

[16] UNSCEAR, Sources and Effects of Ionizing Radiation, Report to the General Assembly, with annexes, UNSCEAR, New York (1993).

[17] ICRP, Cost-Benefit Analysis in the Optimization of Radiation Protection, ICRP Publication 37, Pergamon Press, Oxford (1993).