Ecological risk assessment for a former radium production site in Komi Republic, Russia

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Preface

This study is a part of EANOR project (Long-term consequences of enhanced radioactivity and conventional chemical pollutants for biota at the scale of individuals, population and communities) and was performed in cooperation with scientists from Komi Republic, Russia.

I would like to thank Belykh Elena and Maystrenko Tatiana, senior researchers and Vakhrusheva Olesya, PhD student from the Institute of Biology, Komi Scientific Center, Ural Division of Russian Academy of Sciences and Kaneva Anna, student at Syktyvkar University, for collecting the samples used in this study.

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Yevgeniya Tomkiv

Abstract

Extensive human activities in both nuclear and non-nuclear industries resulted in large number of areas polluted by technologically enhanced naturally occurring radioactive material all over the world. Protection of non-human biota inhabiting those areas from the effects of ionizing radiation has become an important issue in radiological protection. The Vodny site in Komi Republic of Russia is one of the longest radiologically exposed sites in Europe. Twenty-five years of radium production from groundwater and uranium ore, has resulted in contamination of this area with radionuclides and heavy metals. Measurements on Ge-detector, ICP-MS and sequential extraction were used to estimate radionuclide concentrations in the samples collected at the site. Calculation of doses that might be received by biota was performed using the ERICA Tool. Activity concentrations of Ra-226 in the soil samples were ranging from 164 to 11400 Bq/kg soil (dw). The highest estimated doses were observed in lichens and bryophytes (334 µGy/h) followed by soil invertebrates, flying insects and detrivorous invertebrates (150 µGy/h). Internal dose from Ra-226 was the main contributor to the total dose to all reference organisms. Mass concentrations of uranium in the soil samples ranged from 0,43 to 18 mg/kg soil (dw) and did not exceed predicted no-effect concentrations. There was no sufficient data to assess bioavailability of radionuclides in soil samples.

Contents

1. Introduction

Radiation protection has been centered on humans since the beginning of the nuclear era. Numerous studies have been performed to study radiation effects on man and to define safe radiation doses to the public. Assessment frameworks for defining radiation doses to humans and predicting the effects of those doses are in fact much more developed and sophisticated than frameworks dealing with doses and effects caused by non-radioactive contaminants (Hinton et al 2004). However, when it comes to non-human biota, it has been believed for a long time that levels of radiation which are protective for humans will also be sufficient to protect the natural environment. This was a position of the International Commission for the Radiological Protection for many years stated first in 1977:

> "*Although the principle objective of the radiation protection is the achievement and maintenance of appropriately safe conditions for activities involving human exposure, the level of safety required for the protection of all human individuals is thought likely to be adequate to protect other species, although not necessarily individual members of those species."* (ICRP 1977)

In its 1990 recommendations, ICRP was concerned with the environment as a radionuclide transfer media and believed that:

> "*... the standard of environmental control needed to protect man to the degree currently thought desirable will ensure that other species are not put at risk."* (ICRP 1991)

Although humans are considered one of the most radiosensitive species, criteria used for their protection do not necessarily apply for all situations (Brechignac 2003, IAEA 1992, Hinton et al 2004, Pentreath 1998). Exposure pathways of humans and biota vary greatly, even if they are living in the same environment, and so do the doses they receive from radionuclides (IAEA 1992, Hinton et al 2004). In environments where humans are absent (e.g. marine environments), doses to benthic biota can be very high, whilst doses to man remain below recommended limits (Pentreath 1998).

The risks to people tend to be seen differently in comparisons to risks to environment. For people it is important to value each individual, while protection of non-human species is usually aimed at populations (IAEA 1992, Linsley 1996). Only in some special cases of endangered species, rare species or those with low reproduction rates, is individual protection considered (Linsley 1996...). There is no unitary view on how the environment should be valued, how to measure harm caused to exposed populations or to which extent flora and fauna should be protected (Oughton 2003, Pentreath 2007).

Ecological risk assessment is used to evaluate the possibilities of adverse effects on biota caused by exposure to one or many contaminants and can be used for radionuclides in the same manner as for non-radioactive chemicals (Garnier-Laplace et al 2004, Suter et al 2007).

Assessing risks from radionuclide contamination of the environment has a major problem – lack of knowledge and data (Garnier-Laplace et al 2004). Most of the research performed on effects of ionizing radiation on biota have emphasized individual rather than population responses, mortality rather than reproduction (or other more sensitive endpoints), acute rather than chronic exposure, use of external gamma source rather than internal exposures or mixed radionuclides in the environment (Whicker & Hinton 1996, Whicker 1997).

Effects of low radiation doses have been observed at molecular and cellular levels. They include various chromosome aberrations, abnormalities, and DNA damage (Geras'kin et al 2007, Møller et al 2007). Molecular endpoints could give a very early indication of changes in the organisms caused by exposure to ionizing radiation and can be used as molecular biomarkers in risk assessment (Blaylock et al 1996). Molecular responses react rapidly to stress, but are measured on the individual level – lower level of organization. However, in order to be ecologically relevant, they need to be linked to populational responses (Blaylock et al 1996, IAEA 1992). Extrapolation from the individual to populational level is very complex. There are many factors that complicate this process: including biological factors (type of species, life-history traits etc.), abiotic environmental factors (temperature, salinity, sunlight etc.), and relationships between species (mutualism, competition, predation, parasitism etc.) (Harrison and Anderson 1996, IAEA 1992, Whicker & Schultz 1982).

Exposure to ionizing radiation in the natural environment is often combined with exposure to heavy metals. Some of the heavier long-lived radionuclides (e.g. uranium) can cause both radiological and chemical toxicity to living organisms (Ribera et al 1996, Sheppard 2005). Presence of multiple contamination agents, brings further complications to risk assessment (Mothersill C. & Seymour C. 2007, Oughton 2007).

Radiation protection is aimed at both creating safe standards for controlling possible releases from facilities which are using radioactive material; and on dealing with the legacy of past actions. There are many areas in the world which are characterized by high levels of natural occurring radionuclides. Extensive human activities in mining, extraction and production, for needs of both nuclear and non-nuclear industries, resulted in tremendous number of areas polluted by technologically enhanced naturally occurring radioactive material (TENORM). Only uranium mining itself has generated 938×10^6 m^3 of mill tailings all over the globe (IAEA 2004). Coal mining and burning (Dai et al 2007), all types of mineral mining, oil and gas industry (Ericksen 1999) and fertilizer industry (Pachoa and Godoy 2002) also contribute to radionuclide pollution all over the world.

One of the sites, where TENORM has become a problem of concern, will be the subject of this study. It is situated in the Komi Republic, Russia in the Vodny settlement. Different industrial operations involving radioactive substances have been performed on the territory during 25 years (from 1931 to 1956) (Taskaev et al 2003). They included radium extraction from ground water and uranium ore reprocessing (Evseeva et al 2000, Geras'kin et al 2007). A more detailed site description of the actual area investigated during the present study is included in Chapter 2. The following section gives some background on the history of the site and the sources of radionuclide contamination.

1.1 The history of the site (after Evseeva et al 2000)

The area has been used for oil extraction from the XVIII century. When oil extraction was stopped, twenty wells were left abandoned and these continuously poured radionuclide enriched groundwater onto the surface. As a result, at least 15-20 g of radium was spread on the territory, even before the industrial extraction of radium began.

In the 1920s, two independent scientific institutes analyzed groundwater samples and discovered high concentrations of ^{226}Ra (half-life, 1601 y) and ^{228}Ra (half-life, 5,75 y). The concentrations were estimated to be in the order of 300 and 200 Bq/l respectively (Bogoiavlenski 1928). This discovery gave a start to the radium industry in Vodny in 1931. By 1934, there were 59 wells constantly pumping groundwater. The development of the radium-extraction facility was so rapid that it influenced the quality of the process. Production was badly organized, equipment was poorly designed, and safety measurements were ignored.

Sources of environmental contamination included radioactive water that was spilled during its transportation in wooden pipes and barrels (Photo 1, 2). Reprocessed water which still contained high amounts of radionuclides was dumped straight onto the ground. About 15 g of radium were washed into the river with surface runoff.

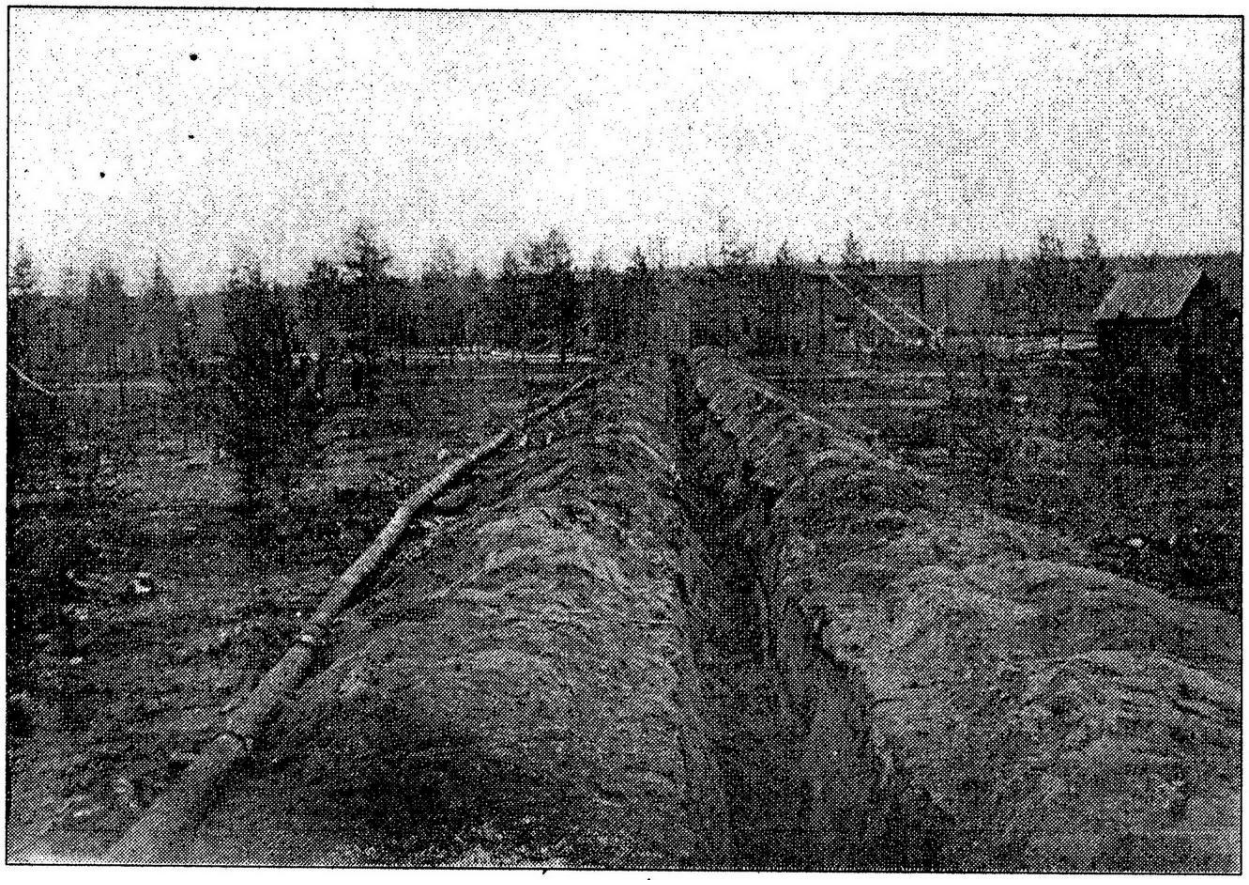

Photo 1 – Building of the wooden pipeline (from Evseeva et al 2000)

Photo 2 – Transportation of radium concentrate in wooden barrels (from Evseeva et al 2000)

In 1947, due to declining amount of radium in groundwater, extraction continued with the use of uranium ore. This caused enrichment of a variety of radionuclides in contaminated soils.

In 1956, radium-extraction on the site was declared unprofitable and all the operations on the territory were ceased. The industry was closed leaving 3000 km^2 of radioactively contaminated land in total.

1.2 Ecological studies on the site

The site represents one of the longest radiologically exposed sites in Europe. Starting in 1957, the Institute of Biology of the Komi Branch of the Russian Academy of Sciences performed numerous investigations of the radioecological situation on contaminated territory (Evseeva et al 2000, Geraskin et al 2007). However, due to secrecy (from both political and commercial reasons) surrounding this radium production, no scientific information about sources and levels

of radioactive contamination of the land have been published (Evseeva et al 2000, Taskaev 2007). The veil of the secrecy was lifted in the 1980s with the publishing of some of the technical papers about Vodny area. Large amounts of the information on radioecological situation at Vodny are still only available in Russian.

On the territory of the settlement, four distinct regions with different types of contamination were identified (Evseeva et al 2000, Geras'kin et al 2007, Taskaev et al 2003):

1. Krokhal – territory of the former chemical plant, contaminated mainly by discharge of Ra-rich groundwater

2. Otvally or Svalka (from Russian word "waste dump") - containing both residues from radium extraction and reprocessed uranium ore.

3. Obzhig (name originates from Russian "burning") - Former production of charcoal with residues similar to those of Otvally

4. Factory 10 – Remains of one of the radium concentration factories, contaminated by radium-rich groundwater

This research is going to be focused on the "Otvally" site (more about site characteristics in Chapter 2). This site presents a unique opportunity to investigate effects of ionizing radiation from a mixture of naturally occurring radionuclides. A number of studies have been carried out to map the radionuclide contamination (Evseeva et al 2011), to study biological effects on various organisms (Evseeva et al 2003, Evseeva et al 2009), to document the possible ecological impacts of exposures and to assess doses received by biota on this contaminated territory (Evseeva et al 2012)

However, any evaluation of the environmental impacts of radiation exposure is dependent on a good assessment of the doses to organisms. Many studies consider only the external gamma dose rates at the sites, and do not include the internal doses from radionuclides. With the increased focus on ecological assessments, a number of tools have been developed to carry out dose

calculations, such as RESRAD biota or the ERICA Assessment Tool (Brown et al., 2007). Since the ERICA tool is part of a larger risk assessment approach, this was applied in the present study.

1.3 The ERICA Integrated Approach and the ERICA Tool

The ERICA Integrated Approach (Environmental Risks from Ionizing Contaminants: Assessment and management) was the result of a EU project of the same name, and was created to «*ensure that decisions on environmental issues give appropriate weight to the environmental exposure, effects and risks from ionizing radiation with emphasis on ensuring the structure and the function of ecosystems*» (Beresford et al 2007). The structure of the overall approach includes three elements (Beresford et al 2007, Larsson 2007):

- Assessment, where activity concentrations in biota and environmental media are used to estimate radiation doses to selected reference organisms.

- Risk characterization, where results of the assessment are evaluated in order to estimate probable adverse effects on biota

- Management, where the pre- and post-assessment decisions are made.

The ERICA Tool is a software package supporting the ERICA Integrated Approach. Essentially, it contains a series of models allowing for a calculation of the radiation doses to a series of "reference organisms" in either aquatic or terrestrial environments. The dose calculations are in turn based on parameters detailing the concentration of radionuclides in organisms as compared to environmental media (concentration ratios, CR), radionuclide partitioning between sediment and water (distribution coefficients, Kd), and dose conversion factors (DCF) for converting Bq to Gy for a variety of different radionuclides. In order to put the observed doses into context, the ERICA Tool interacts with the FREDERICA radiation effects database. This database is based on FRED and FASSET radiation effect databases and is extended to include «*all the scientific literature on radiation effect experiments and field studies, organized around different wildlife groups and for most data, broadly categorized according to four effect umbrella endpoints: morbidity, mortality, reproduction, and mutation*» (Larsson 2008). Finally, the integrated

approach enables the observed doses to organisms to be compared to screening values representing expected no adverse effect levels of radiation doses.

The ERICA Tool can be run in three different tiers. Tier 1 is simple and conservative and doesn't require much input data. In this tier, input media concentrations of radionuclides are compared to Environmental Media Concentration Limits (EMCL) for each radionuclide and a risk quotient (RQ) is calculated as (Larsson 2008):

$$RQ=\text{Assessed value/screening value}$$

In Tier 2, *"total weighted doses to each reference organism are estimated and are compared directly with the screening value"* (default or selected by user) (Brown et al 2008). This tier is much more flexible and allows user to make changes and provide site specific concentration ratios, distribution coefficients, occupancy factors and radiation weighting factors. It also provides users with information on the effects that are likely to occur in each reference organism group at calculated dose rates. This tier gives users an overview of data availability for all groups of reference organisms which helps to identify knowledge gaps and need for further research. It also has a possibility for performing probabilistic assessment.

Finally, Tier 3 of assessment is likely to be used in complex and unique situations. It's a *"probabilistic risk assessment where uncertainties within the results may be determined using sensitivity analysis"* (Larsson 2008).

1.4 Study aims

The aim of this study was to carry out an analysis of radionuclide concentrations in samples collected from the site, estimate doses that might be received by biota living on the contaminated site, and to identify the most dose-relevant radionuclides as well as the groups of organisms that are likely to receive the highest doses. In addition, the radiation doses have been compared with the possible chemical toxicity of uranium at the site.

2. Materials and methods

2.1 Site description

The study area is situated in the Komi Republic (Russia) beside the Vodny settlement (63ºN 53ºE) (Figure 1) in the North Taiga subzone, and represents the most highly contaminated areas within the 3000 km^2 of contaminated territory. It includes watershed, a first and second floodplain terrace and the floodplain of the Ukhta river and covers 1300 m^2. The major soil types of the site are Fluvisoils and Albeluvisoils. The vegetation on the area is herbaceous plants (Photo 3,4).

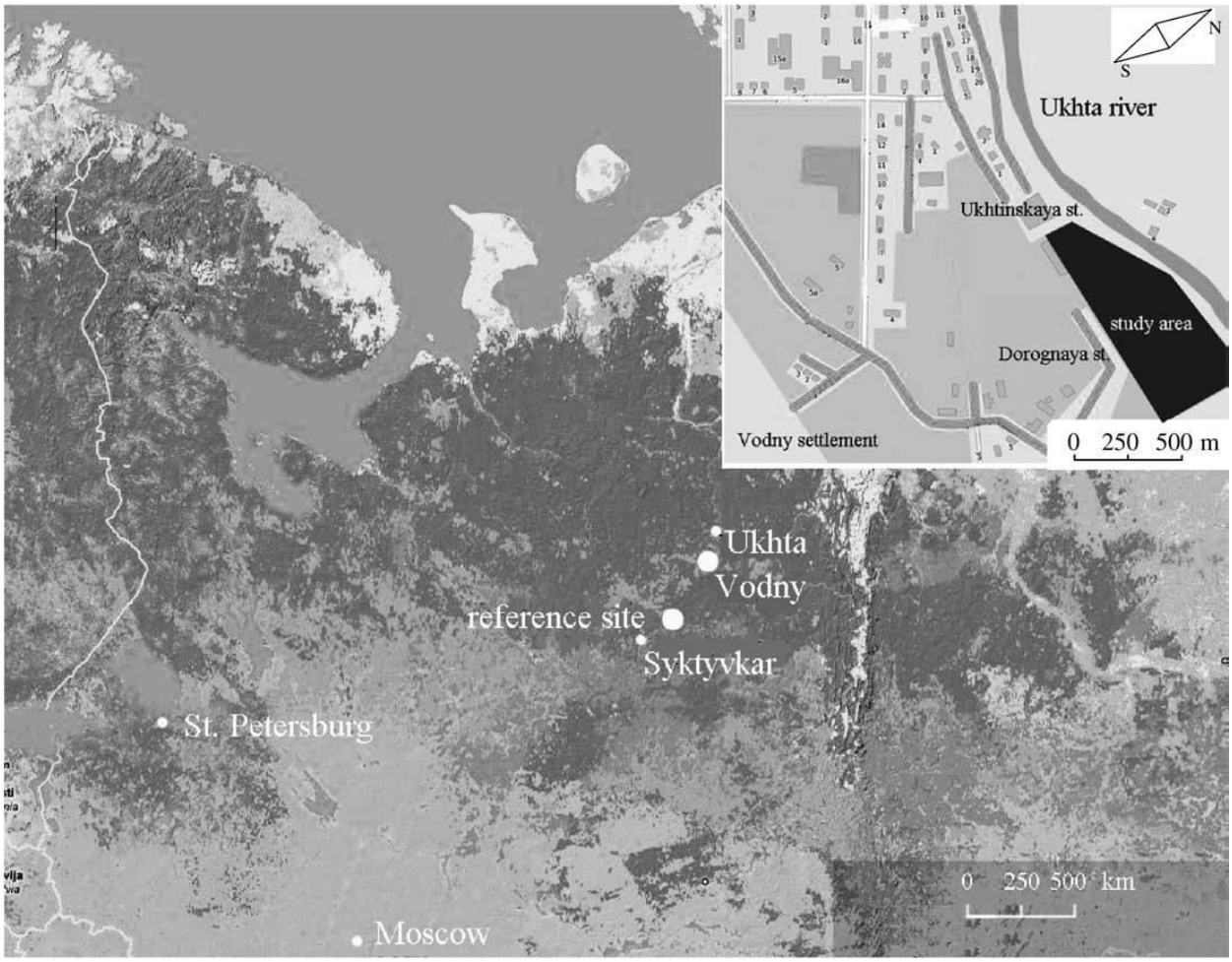

Figure 1 – Location of study area on the map (from Evseeva et al 2009)

Photo 3 – Study area, view north (Photo by Belykh E.)

Photo 4 – Study area, view south (Photo by Belykh E.)

As detailed above, twenty-five years of radium production from both groundwater and uranium ore extraction resulted in contamination of the whole territory with significantly enhanced concentrations of natural decay chain radionuclides. Sand-gravel mixture was spread on the soil in 1961 in attempt to rehabilitate the area. Due to water erosion and plant and animal activity, contamination of the top soil has increased, resulting in an uneven distribution of the radionuclides on the territory (Geras'kin et al 2007, Evseeva et al 2011).

2.2 Sampling and measurements

The study area has been divided into a grid, and coordinates of the sampling points (Figure 2) have been determined using Arcview GIS-3.2. Soil samples were collected at ten different sites, and three soil samples were collected at each sampling site to a depth of 0-20 cm. Samples were dried, cleaned from roots and stones and sieved through a 1 μm sieve.

Herbaceous species present on the area were sampled at three of the soil sampling sites, and taken from a 25x25 m square. Plants were sampled from the corners and center of the square. Samples were then cleaned, dried and milled to achieve a better counting geometry on the detector.

One soil sample from each soil sampling site and one plant sample from each plant sampling site were chosen randomly for analysis in this study. Those samples were vacuum packed with Mini Eco vacuum packing machine (HFE vacuum systems b.v., The Netherlands) and left for 1 month in order to achieve secular equilibrium between Ra-226 and its progeny (Photo 5, 6).

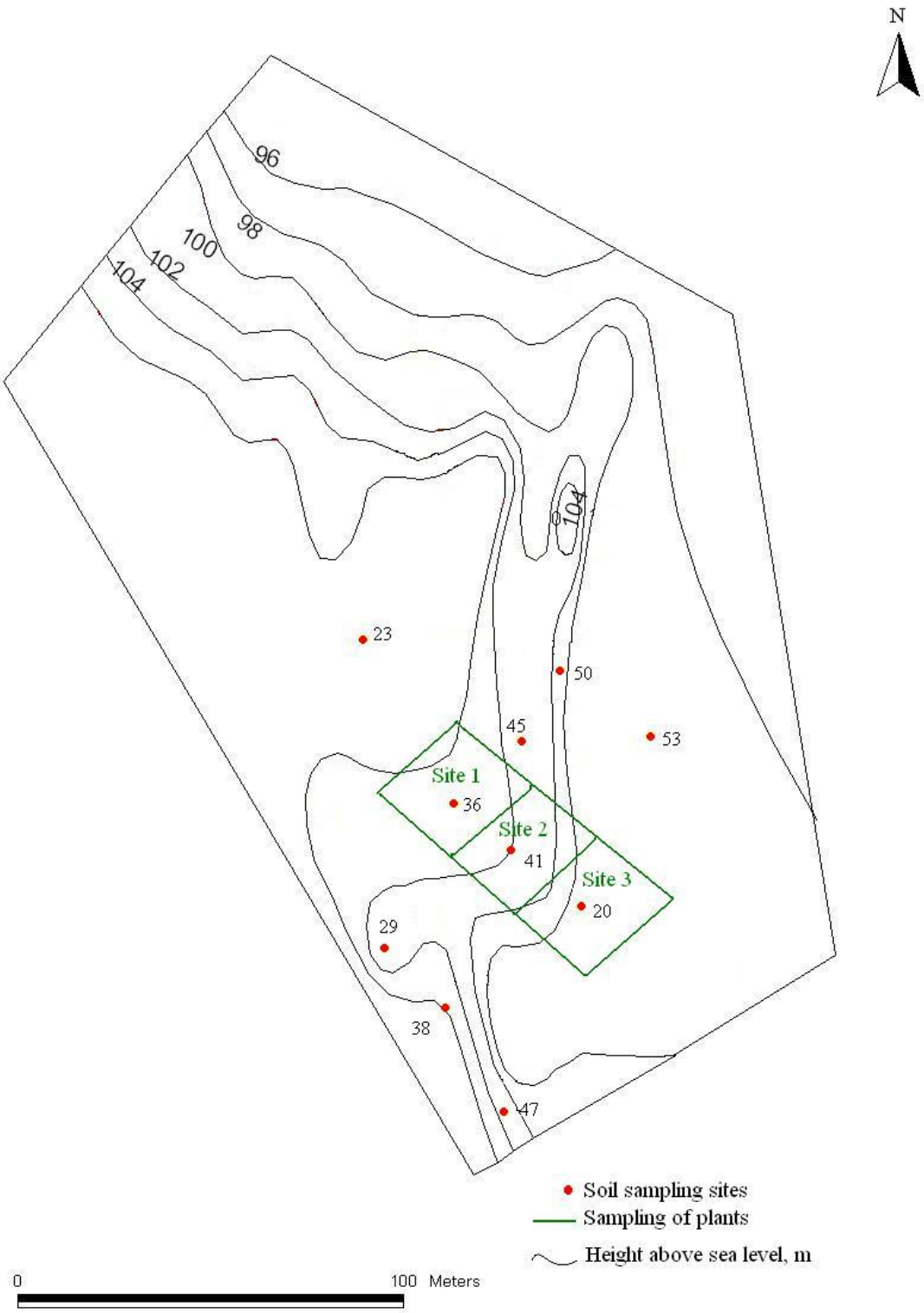

Figure 2 - Sampling points on the site (modified after Evseeva et al 2011)

Photo 7 – Mini Eco vacuum packing machine

Photo 8 – Vacuum packed samples

Since the radon daughter is a gas, its removal from samples following ^{226}Ra decay, means that concentrations of the subsequent daughters, including the ^{214}Bi and ^{214}Pb isotopes, are usually decreased relative to the parent ^{226}Ra activity concentrations. Vacuum sealing keeps the ^{222}Rn gas in the sample and allows the decay chain to reach equilibrium (Figure 3).

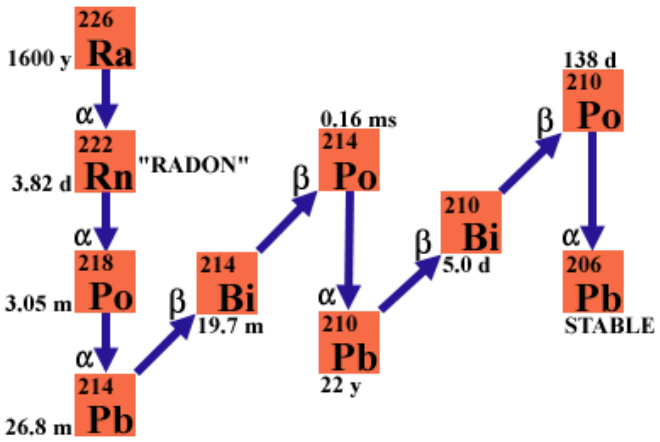

Figure 3. Radium decay chain (from http://www.nist.gov/pml/general/curie/1927.cfm)

All samples were counted using gamma spectrometry on high purity Ge-detector (energy resolution – 1,9 keV for 1332,5 keV peak, 20% efficiency) produced by Canberra, Meriden, CT. Analysis of the gamma spectra have been performed with Genie 2k gamma spectrum analysis software (Canberra, Meriden, CT).

Measurement of ^{238}U concentrations by mass in the samples was performed using ICP-MS analysis (Perkin Elmer Sciex Elan 6000, Norwalk, CT). Prior to analysis, 0,25 g samples were digested with the ultraclave (Milestone Inc., Ultraclave High performance detector, Shelton, CT) using high grade purity acid HNO_3 and HF for soil and HNO_3-HF-H_2O for plant samples. An internal standard with Tl was been added to all the samples before decomposition.

Standard reference material (Rocky Flats 4353a), calibration standards, reagent and method blanks have been used to ensure accuracy of measurements.

Detection limits for the ^{238}U ICP-MS measurement was calculated as ten times standard deviation of blank measurements and was 0,02µg/L.

Four randomly chosen soil samples were subjected to sequential extraction procedure (Oughton et al 1992) for determining bioavailable fractions of radionuclides and heavy metals. Samples (2 g) were weighed into centrifuge tubes, and shaken with 20 ml of exractant (Table 1). The supernatant was separated from the solid by high-speed centrifugation at 10.000 g (Beckman Coulter Allegra 64R). The solution was filtered to a vial with the use of blue band filter. Solid phases were washed with 20 ml Milli-Q water and centrifuged between each extraction step.

Soil samples, supernatants and wash solutions were counted on Ge-detector, but the activity concentrations were below detection limit. Since performing ICP-MS measurements on this samples wasn't possible due to technical issues, no results based on them is going to be presented in this study.

Table 1. – Sequential extraction procedure

Part of procedure	Extractant	Time of extraction	Conditions
Step 1	Milli-Q water	1 hour	-
Step 2	Amonium acetate (NH_4Ac)	2 hours	at soil pH
Step 3	NH_4Ac	2 hours	at pH~5
Step 4	Hydroxylamin($NH_2OH \cdot HCl$)	6 hours	at pH~2 water bath at 80°C
Step 5	H_2O_2+ NH_4Ac	6 hours	water bath at 80°C
Step 6	HNO_3	6 hours	water bath at 80°C

2.3 Data analysis

Statistical analysis of data was performed using standard statistical formulas in Microsoft Excel spreadsheet.

Radium and its progeny were assumed to be in equilibrium after samples were left vacuum packed for 30 days. That's why average activity concentrations of ^{226}Ra and its daughters ^{214}Bi and ^{214}Pb in soil and plant material were taken to be the most reliable estimate of the ^{226}Ra concentrations, and used as the input data for dose assessment. If the direct measurements of ^{226}Ra were not achieved, the average activity concentrations of ^{214}Bi and ^{214}Pb were used.

Since the chemical toxicity of uranium is not included in the ERICA tool, concentrations in the environmental media were compared with available predicted no-effect concentration (PNECs). However, the equivalent activity levels of ^{238}U were included in the ERICA dose assessment. When data was lacking on the relative ^{235}U/^{238}U ratios, these were taken to be equivalent to natural uranium isotope ratios.

2.4 Dose Assessment

The ERICA Tool (Tier 2) was used for calculation of radiation exposure doses to biota. The default dose rate screening value of 10 µGy/h, which represents a generic predicted no-effect dose rate for all organisms, was kept. The input data included measured concentrations in

environmental media and default parameters in The Erica Tool. All the available reference organism groups for terrestrial ecosystems were included in the assessment. Given the heterogeneity of the radionuclide distribution on the site, the assessment was run with both mean and maximum measured soil activity concentrations.

Since the aim of this study was to make a real-life estimation of possible doses that organisms living on the contaminated area might receive, rather than to model the probability of those doses exceeding the screening value, the uncertainty factor was set to 1. Default weighting radiation factors for all types of radiation (10 for alpha emitters and 3 for low level beta emitters) were retained together with default occupancy factors for the organisms. The ERICA tool also includes default parameters of concentration ratios (CR) for organisms and radionuclides as based on reviews of available experimental data. In the case of terrestrial organisms these are all expressed as (Bq/kg fw organism)/(Bq/kg dw soil). When no data is available, the tool gives a number of options for filling the data gaps, such as similar organism or biogeochemistry. In the case of the present study, experimental data was missing for bird eggs, amphibian and flying insects (for Ra Th U), detrivorous invertebrates and gastropods (for Th U), reptiles (for Pb, Ra, Th, U), and soil invertebrates (for Ra, Th).

The following methods were used when deriving ERICA default concentration ratio values (for filling data gaps): similar taxonomy, similar reference organisms, similar biogeochemistry and taxonomy, allometric or other modelling approaches (Table 2).

Table 2. - Methods used to derive ERICA default concentration ratio values

Reference organism **Radionuclide**	**Amphibian**	**Bird egg**	**Detrivorous invertebrates**	**Flying insects**	**Gastropod**	**Reptile**	**Soil invertebrates**
Cs		Allometric or other modelling approaches					
Pb		Similar taxonomy				Similar reference organism	
Ra	Similar reference organism	Similar taxonomy		Similar taxonomy		Similar reference organism	Similar taxonomy
Th	Similar reference organism	Similar taxonomy	Similar biogeochemistry and taxonomy	Similar biogeochemistry and	Similar biogeochemistry and taxonomy	Similar reference organism	Similar biogeochemistry and taxonomy

				taxonomy			
U	Similar reference organism	Allometric or other modelling approaches	Similar taxonomy	Similar taxonomy	Similar taxonomy	Similar reference organism	

Measured activity concentrations in the plant samples were also used in the assessment if available. Input data was entered in Bq/kg dry weight for soil and Bq/kg fresh weight for plant samples.

3. Results and discussion

3.1 Activity concentrations of radionuclides in samples

Results of the sample measurements showed a variety of radionuclides of natural decay chains. Activity concentrations of radionuclides in soil and plant samples are given in Table 3. More detailed information about radionuclide content in each sample is given in Annex A and B.

Although there have been efforts to remediate the area by covering it with a sand and gravel mixture, radioactively contaminated soils have been gradually uncovered due to erosion (Evseeva et al 2009, Geras'kin et al 2007). This has resulted in a higher activity in the top layer of soil in some places. Also there has been some run-off from higher altitude to lower altitude sites. This explains heterogeneity of the contamination, where relatively «clean» patches border with highly polluted ones.

A comparison of the activities measured in the samples before and after packing showed that ^{226}Ra, ^{214}Pb and ^{214}Bi activity concentrations values in most of the samples have become more even given that they reached equilibrium. The range of activity concentrations of ^{226}Ra in comparison to ^{214}Bi and ^{214}Pb is skewed since direct measurements of ^{226}Ra, especially on the lower activities, are harder to obtain. However, given time to reach equilibrium, both ^{214}Bi and ^{214}Pb give a more reliable indication of the ^{226}Ra, and as such are used for the most reliable estimation of ^{226}Ra content.

The main problem with measurement of ^{226}Ra is interference from ^{235}U at 185,7 keV peak (branching ratio > 50%). There is only one gamma peak for ^{226}Ra, at 186 keV, (with a branching ratio of only 3.28 %). While it is possible to compensate for ^{235}U if a reliable measurement can be made at one or more of the ^{235}U peaks (e.g. at 146 keV), the high number of gamma peaks in the region from naturally occurring radionuclides, combined with the low branching ratios means that estimates of both ^{226}Ra and ^{235}U can be flawed. In such cases, activities of the ^{226}Ra daughters which we assume are in equilibrium give a better estimation of the actual ^{226}Ra concentration, since they have multiple gamma energy peaks to support the analysis.

Plant material contained low activity concentrations of most radionuclides (except for ^{40}K, although this is probably an anomaly). Therefore, only one direct measurement on ^{226}Ra was received. However, activities of ^{226}Ra progeny suggest that there had been some accumulation of ^{226}Ra in plants, and these were used to calculate the eventual ^{226}Ra concentrations.

Although the site is considered clear of Chernobyl fallout (Evseeva et al 2009), small concentrations of ^{137}Cs were measured in some soil samples. These were much lower than concentrations seen at another site in Kirovo-Chepetsk (Russia) which was thought to be contaminated by Chernobyl and had much higher ^{137}Cs activity levels in soils (1470±1047 Bq/kg). Also the Kirovo-Chepetsk site had comparatively low levels of ^{226}Ra and U isotopes.

Table 3. Activity concentrations of radionuclides in the samples measured before and after vacuum-packing

Radionuclides	Before the inpacking		After the inpacking	
	Mean activity concentrations in soil, Bq/kg dw (STD) Min-max	Mean activity concentrations in plants, Bq/kg dw (STD) Min-max	Mean activity concentrations in soil, Bq/kg dw (STD) Min-max	Mean activity concentrations in plants, Bq/kg dw (STD) Min-max
^{40}**K**	758±266 433-1245	2625±247 2466-2909	596±231 251-907	1346±575 863-1982
^{137}**Cs**	24±8 20-33		31±16 20-49	
^{210}**Pb**	4913±5100 492-15112		3697±3783 414-11106	
^{212}**Pb**	82,6*		34±32 12-57	
^{214}**Bi**	3529±3532 174-10508	125,8*	4092±3622 178-10856	265±8 260-271
^{214}**Pb**	4333±3781 1254-11359	91±59 49-133	4150±3746 150-11126	171±95 65-249
^{219}**Rn**	944±2 943-946			
^{226}**Ra**	7249±4193 2687-13425	384,7*	5968±3696 2574-11361	458,2*
^{227}**Th**	1186,5*			
^{231}**Pa**	1615,2*			
^{231}**Th**	1399±1012 176-2631		1887±1372 1036-3469	
^{235}**U**	520±240 237-815	23,3*	348±80 292-405	27,6*

* - radionuclide was determined in only one sample

The concentration of ^{238}U in the soil ranged from 0,43 to 18 mg/kg, which is equivalent to between 5,3 and 223 Bq/kg soil (Table 4).

Table 4. Measured concentrations of ^{238}U in the soil and plant samples

Sample No	Sample type	Mass concentration in sample, mg/kg	Activity concentration in sample, Bq/kg
20	Soil	0,86	10,7
23	Soil	0,43	5,3
29	Soil	18,01	223
36	Soil	4,67	57,9
38	Soil	14,57	180
41	Soil	1,95	24,1
45	Soil	2,81	34,8
47	Soil	0,51	6,3
50	Soil	13,08	162
53	Soil	2,25	27,9
1_3	Plant	0,01	0,11
2_4	Plant	-*	-*
3_5	Plant	-*	-*
Reference material	Soil	3,16	39,23**

* - below detection limit

** Certified massic activity of ^{238}U in the reference sample – 39,6±3,0

If the U isotopes were in equilibrium and present at ratios consistent with natural abundance, one would expect a corresponding 0,2-10 Bq/kg ^{235}U and 5,3-223 Bq/kg ^{234}U (Table 5). As can be seen from the results, the ^{235}U activities were often higher than one would expect from the ^{238}U measurements. Since the ^{235}U measurement is known to be rather unreliable due to interference with ^{226}Ra, and uranium chemical analysis showed excellent agreement with standard reference materials, the ^{238}U measurement is probably the most reliable.

Table 5. Natural abundances of uranium isotopes (after Cordfunke 1969, and Ewing 1999) and ranges of their concentration in the soil samples

Isotope	Isotope abundance, %	Half-life	Activity in the samples, Bq/kg
^{238}U	99,275	$4{,}5\times10^{9}$ y	5.3 – 223
^{235}U*	0,7200	7×10^{8} y	0.2 - 10
^{234}U*	0,0055	$2{,}5\times10^{5}$ y	5.3 – 223

* - estimated from measurements on ^{238}U

Concentration of ^{238}U in the plant samples was very low or below detection limit. This can be explained by the varying accumulation ability of different plants as it was shown in Salbu et al (2011) where ^{238}U content in plants sampled from Kadji Sai mining area in Kyrgyzstan varied from 0,3 to 416 Bq/kg depending on plant species. Another explanation for such low concentrations of ^{238}U could be no accumulation of uranium in the above-ground parts of this particular plant species (Pereira et al 2009). The size of the plant samples measured on ICP-MS could also be insufficient in order to get results on uranium content.

Although all results show the concentrations of radionuclides and uranium in the soil, there is no information on its bioavailability. While four soil samples were subject to sequential extraction for determination of radionuclide mobility, the concentrations were below detection levels. Lack of information can lead to overestimation of the chemical and radiological risk caused by contaminants. Furthermore, there may be a number of other heavy metals present on the site which may act as multiple stressors together with ionizing radiation.

Previous analysis has shown that the mean activity concentration of ^{238}U in the soil of the site were about 2 times higher than its natural content in the soils of Russia. As for ^{226}Ra, they exceed natural levels by 650 times (Evseeva et al 2011, UNSCEAR 2000), reaching over 100 kBq/kg in some areas. Although the samples analyzed in this study have been taken from the less contaminated part of the site, with expected activity levels of less than 10 kBq/kg (Figure 4), one soil sample did reach this maximum level with 10 kBq/kg of radium and that soil sample should be considered radioactive waste according to Russian standards (Sanitary rules 2003).

In general the observed ^{226}Ra concentrations were in line with those expected, ranging from 164 to 10400 Bq/kg from the 1-10 kBq/kg area; and both samples from the 100-1000 Bq/kg area were below detection limits. Two samples apparently taken within the 10-100 kBq/kg area were lower than expected, but they were both on the very edge of the contours, and hence the map locations may not be completely accurate.

When compared to other areas in the world with high levels of TENORM, concentrations of ^{238}U in soils of Vodny site were generally lower than those observed in soils of uranium mining sites in Central Asia. Uranium concentrations in those areas were in the range of 71-1455 Bq/kg, 1082-5858 Bq/kg and 296-590 Bq/kg for Kazakhstan, Kyrgyzstan and Tajikistan mining sites respectively (Salbu et al 2011). However, the samples for this study have been taken from a less polluted part of the site and a cartographical investigation performed by Russian scientists has showed that up to 1140 Bq/kg of ^{238}U can be measured at some spots (Evseeva et al 2011). A comparison of ^{238}U concentrations values received in current study with uranium levels measured in the Fen area in Norway (one of world's largest deposits of thorium) showed that they were in the similar range (Popic et al 2011).

Concentrations of radium were generally similar to those received in studying mining sites of Kazakhstan and Kyrgyzstan; they were measured to be 114-2185 Bq/kg and 1285-4990 Bq/kg respectively. However, this wasn't true for one sample where more than 10 kBq/kg of ^{226}Ra were measured.

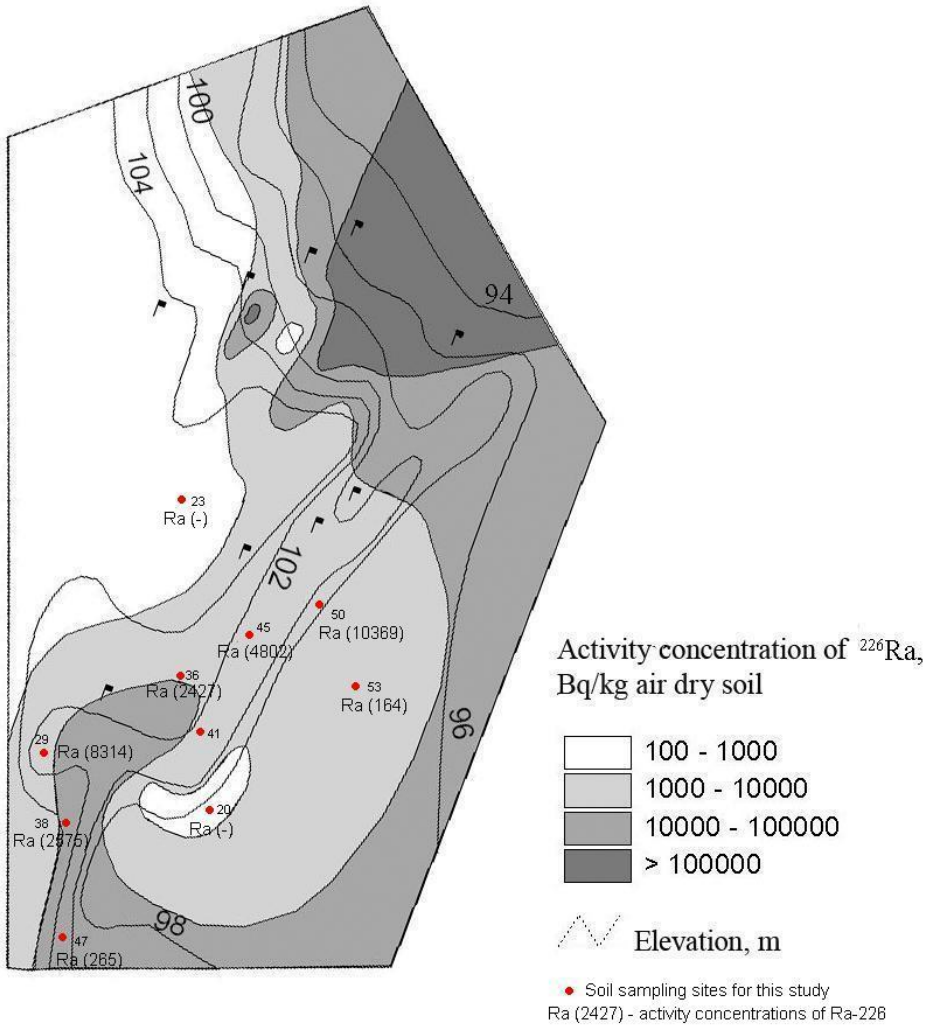

Figure 4 – Sampling sites for soils used in this study in relation to Ra distribution on the whole contaminated territory (modified after Evseeva et al 2011)

3.2 Dose and risk assessment

Radionuclide measurements confirm that extraction of radium from the groundwater and subsequently from uranium ore has resulted in significant radioactive contamination of the area. Apart from low concentrations of Cs-137, all the radionuclides of interest include those of natural decay series. These series include both beta and alpha-emitters, which means that in addition to humans, natural plant and animal populations are also being exposed to α-, β- and γ-

radiation. The territory is also contaminated with heavy metals and rare-earth elements, in addition to radionuclides, which complicates assessment of the risks to the environment.

The assessment of doses to terrestrial organisms was carried out using Tier 2 of the ERICA tool. Although the tool asks the assessor to consider stakeholder involvement as part of the assessment, at this initial stage, stakeholder involvement was not deemed to be necessary. However, one would hope it could be included as part of the next phases of the assessment.

The maximum calculated doses to all the reference organisms (Figure 5) was highest for lichens and bryophytes, at 334µGy/h, followed by detrivorous invertebrates, insects and soil invertebrates, at about 150 µGy/h. Doses to the rest of the reference organisms (except for trees) in a range from 48 to 75 µGy/h. Only trees did not exceed the screening dose value.

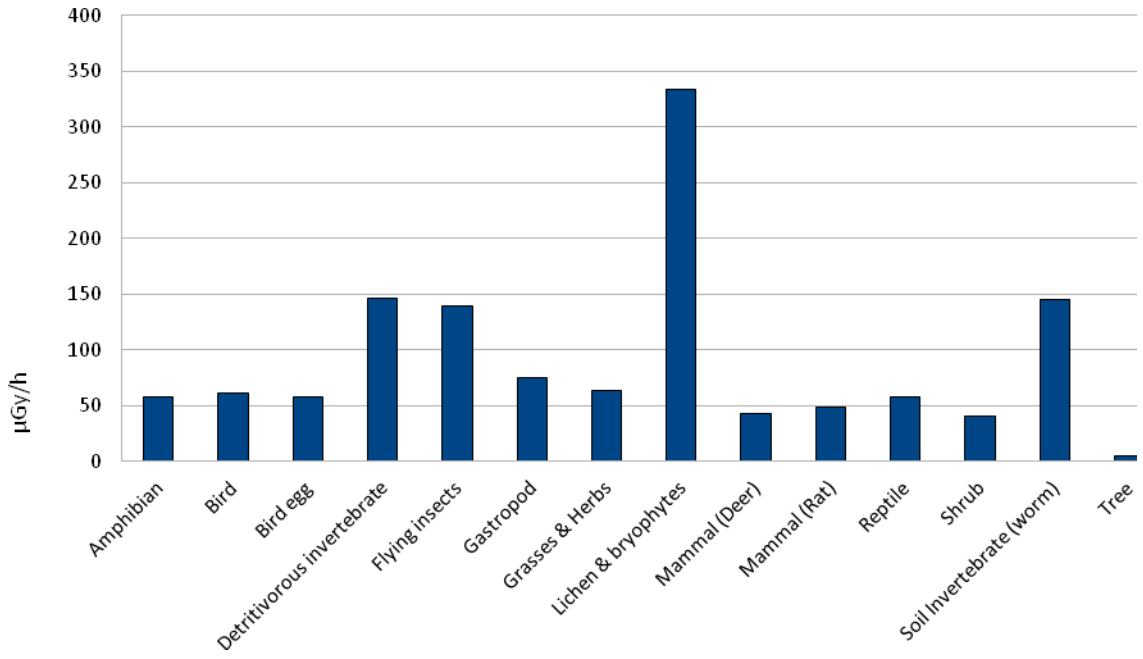

Figure 5 – Calculated total doses to reference organisms (maximum soil concentrations were used.)

The mean calculated doses to reference organisms were about 2,5 times lower but showed a similar pattern of dose distribution (Figure 6), and still most of the organisms were above the 10 uGy/hr screening dose.

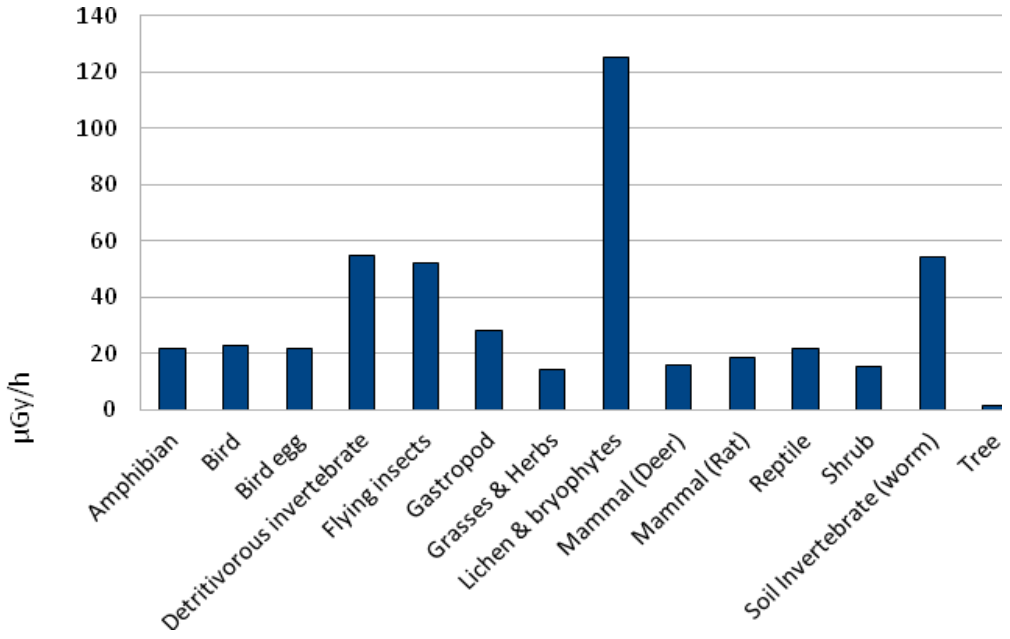

Figure 6 – Calculated total doses to reference organisms (mean soil concentrations were used)

Ra-226 was shown to contribute most to the total estimated dose for all the reference organisms (Figure 7) and this is consistent with results received in previous research in the area (Evseeva et al 2009, Hosseini et al 2011). Internal alpha doses from Ra-226 contribute from 80 to 93 % to the total dose (26% for trees).

According to the summary on PNECs for U as a chemical toxin performed by Sheppard (2005), no effects are expected to occur on terrestrial plants and soil biota under 250 and 100 mgU/kg dry soil respectively. However recent study shows that already at concentrations of 5-15 mg/kg, «DNA damage and adverse effects on lysosomal membrane stability were observed» in earthworms Eisenia fetida (Giovanetti et al 2010).

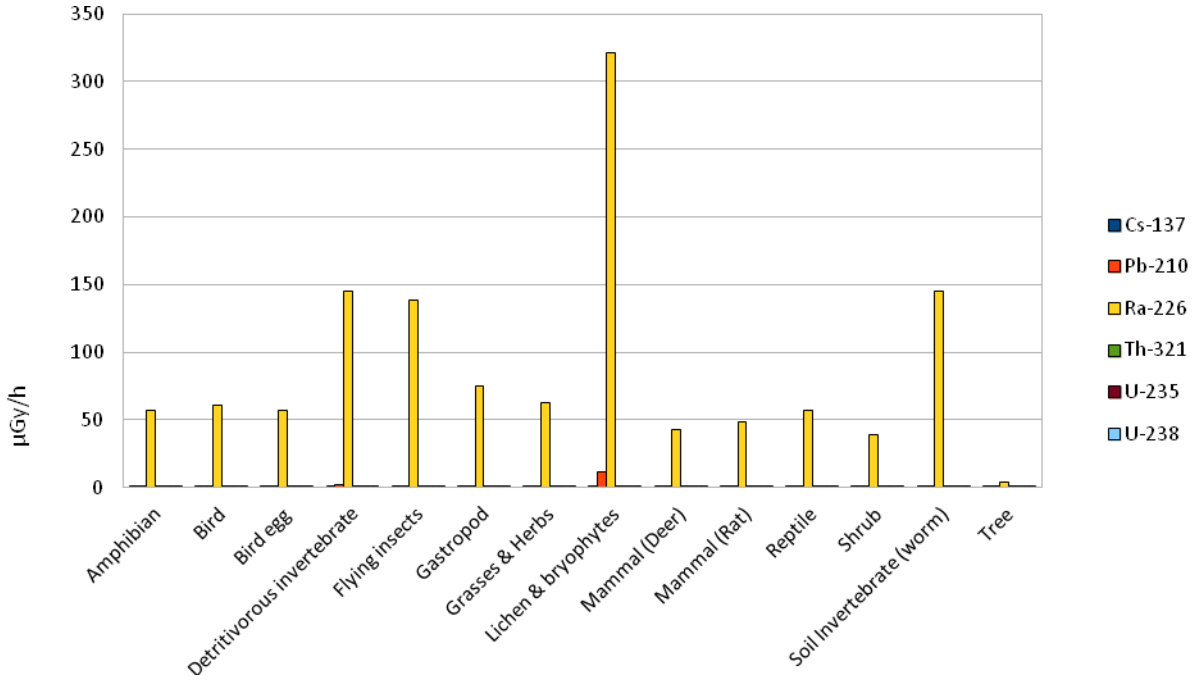

Figure 7 – Calculated doses for reference organisms from different radionuclides (maximum soil concentration were used)

Despite of little data on radionuclide concentrations in plants, it was possible to perform comparison of the concentration ratios (CR) for plants from the site with the CR in the ERICA (Table 6). Concentration ratios were in good agreement in case of ^{226}Ra, but calculated CR for ^{238}U was much lower than default CR in the ERICA Tool. Site-specific CR might not be very reliable, since ^{238}U was measured in just one plant sample. However, Thørring er al (2007) noted that *"there seems to be a general over-prediction of U activity concentrations using default CRs in most cases"*

Table 6. - Comparison of soil and plant activity levels and concentration ratios

Sampling site	Radionuclides	Soil Bq/kg (dw)	Plant Bq/kg (fw)	CR plant/soil	ERICA CR
Site 1	Pb-210	1627			
	Ra-226	2427	111	4,6E-2	3.94E-2
	U-235		11,9		
	U-238	57,91	0,05	8,6E-4	1.46E-2
Site 2	Pb-210	1633			
	Pb-212	11,8			
	Ra-226	2480	89,3	3,6E-2	3.94E-2
	U-238	24,17			
Site 3	Ra-226		30,7		
	U-238	10,67			
Average	Ra-226	2453	77	4,1E-2	3.94E-2
	U-238	30,91	0,05	8,6E-4	1.46E-2

4. Conclusions

Soil activities on the site showed a heterogeneous distribution of radionuclides. Activity concentrations of ^{226}Ra in the samples ranged from 164 up to 11400 Bq/kg dry weight, despite the fact that the samples analyzed were taken from the less contaminated part of the site (according to previous studies).

The maximum doses on the site were observed in lichens and bryophytes (334 µGy/h), although they are considered as one of the radioresistant plant groups compared to trees, shrubs and herbaceous species (Whicker 1997). There are no effects stated in the database for this group of species which means they should probably be sampled on the further assessment and studied more carefully. Doses to soil invertebrates, detrivorous invertebrates and flying insects were about 150µGy/h. The internal dose from ^{226}Ra contributed most to the total doses expected for organisms. Although calculated concentration ratios for radium in plants were in good agreement with the default CR value in the ERICA tool, lack of data on CR for radium for many of the other organisms represents a large source of uncertainty.

The number of biota samples and organisms available for measurements were limited (only three plants samples) so further research should be recommended with the sampling of all possible groups of reference organisms to achieve more site-specific calculated doses.

While mass concentrations of uranium in the soil did not exceed predicted no-effect concentrations based on chemical toxicity, these are probably not the highest levels at the site and it cannot be ruled out that other soils would exceed both chemical and radiological no-effect levels. This may further complicate effect analysis due to mixture toxicity. Furthermore, measurements of other chemical compounds that might act as multiple stressors were not carried out in the present study, but would be essential to provide an overall picture of the potential ecological risk. Sequential extraction of the soils did not give sufficient data to assess the bioavailability of the radionuclides, but should be a focus for future research.

Finally, Hosseini et al (2011) have mentioned that radiation responses of the organisms living in the boreal/Arctic regions are different of those living in warm climate. The combined effects of

low temperatures and ionizing radiation can lead to higher response of biota at the same doses and give an underestimation of the risks.

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Annex A - Activity concentrations of radionuclides in plant and soil material before vacuum packing

Sample No	Sample type	Weight, g	Time	Radionuclide	Activity, Bq	Uncertainity Bq	Activity, Bq/kg
20	Soil	13.5774	2 hrs	K-40	10.5	5.68	773
23	Soil	26.9503	2 hrs	K-40	13.5	5.58	500
29	Soil	11.2621	2 hrs	Pb-210	14.6	2.32	1296
				Pb-212	0.93	0.18	82.6
				Bi-214	22.7	0.6	2015
				Pb-214	24.3	0.6	2157
				Ra-226	151.2	5.57	13425
				U-235	9.18	0.4	815
36	Soil	19.8046	2 hrs	K-40	8.57	4.44	432
				Pb-210	57.1	7.79	2883
				Bi-214	33.9	0.76	1711
				Pb-214	36.1	0.75	1822
				Ra-226	94.2	4.41	4756
				Th-231	3.49	0.9	176
38	Soil	18.2472	2 hrs	Cs-137	0.37	0.16	20.3
				Pb-210	138.6	18.3	7595
				Bi-214	100	1.35	5480
				Pb-214	100.7	1.7	5518
				Rn-219	17.2	1.3	942
				Ra-226	49.03	16.8	2687
				Th-231	28.6	2.44	1567
				U-235	8.36	0.9	458
41	Soil	25.4308	2 hrs	Pb-210	52.5	7.1	2064
				Bi-214	30.1	0.7	1183
				Pb-214	31.9	0.7	1254
				Ra-226	99.2	4.4	3900
				U-235	6.03	0.3	237
45	Soil	25.3887	2 hrs	K-40	21.04	6.15	828
				Cs-137	0.5	0.17	19.7
				Pb-210	125.6	16.7	4947
				Bi-214	92.3	1.3	3635
				Pb-214	98.6	1.66	3883
				Ra-226	208.8	7.13	8224
				Th-231	31	2.4	1221
47	Soil	13.2453	2 hrs	K-40	16.5	5.93	1245
				Pb-214	2.3	0.2	173

Annex A - Activity concentrations of radionuclides in plant and soil material before vacuum packing (continued)

Sample No	Sample type	Weight, g	Time	Radionuclide	Activity, Bq	Uncertainity Bq	Activity, Bq/kg
50	Soil	17.0255	2 hrs	K-40	14.1	5.16	828
				Pb-210	257.3	33.9	15112
				Bi-214	178.9	1.91	10507
				Pb-214	193.4	2.99	11359
				Rn-219	16.1	1.32	945
				Ra-226	178.8	20.2	10501
				Th-227	20.2	0.9	1186
				Pa-231	27.5	5.11	1615
				Th-231	44.8	3.36	2631
				U-235	9.69	1.07	569
53	Soil	15.2462	2 hrs	K-40	10.6	4.27	695
				Cs-137	0.51	0.14	33.4
				Pb-210	7.51	1.41	492
1_3	Plant	8.6786	2 hrs	K-40	21.7	6.02	2500
2_4	Plant	8.4748	20 hrs	K-40	20.9	2.12	2466
				Pb-214	1.13	0.06	133
				Ra-226	3.26	0.7	384
				U-235	0.198	0.04	23.3
3_5	Plant	9.1429	42 hrs	K-40	26.6	1.76	2909
				Bi-214	1.15	0.06	125
				Pb-214	0.45	0.03	49.2
Ref.mat	Soil	19.442	19 hrs	K-40	18.8	1.63	966
				Cs-137	0.34	0.05	17.5
				Bi-212	1.87	0.32	96.2
				Pb-212	1.73	0.07	88.9
				Bi-214	1.23	0.08	63.2
				Pb-214	0.8	0.06	41.1
				U-235	0.18	0.04	9.2

Annex B - Activity concentrations of radionuclides in plant and soil material after vacuum packing

Sample No	Sample type	Weight g	Time	Radionuclide	Activity Bq	Uncertainity Bq	Activity Bq/kg
20	Soil	13.5774	4,5 hrs	-	-	-	-
23	Soil	26.9503	4,5 hrs	K-40	12.4	3.02	460
29	Soil	11.2621	3,5 hrs	K-40	9.17	3.52	814
				Cs-137	0.55	0.13	48
				Pb-210	11.49	1.87	1020
				Pb-212	0.64	0.16	56
				Bi-214	75.03	0.96	6662
				Pb-214	77.94	1.27	6920
				Ra-226	127.95	4.46	11361
36	Soil	19.8046	3,5 hrs	Pb-210	32.23	4.38	1627
				Bi-214	44.21	0.68	2232
				Pb-214	42.98	0.78	2170
				Ra-226	57.00	2.79	2878
38	Soil	18.2472	3,5 hrs	K-40	7.93	3.49	434
				Pb-210	104.17	13.63	5708
				Bi-214	99.09	1.11	5430
				Pb-214	98.28	1.57	5386
				Ra-226	46.98	11.92	2574
				Th-227	10.95	0.57	600
				Th-231	21.07	1.73	1154
				U-235	5.33	0.67	292
41	Soil	25.4308	3,5 hrs	K-40	6.38	3.32	250
				Pb-210	41.55	5.58	1633
				Pb-212	0.3	0.13	11.8
				Bi-214	55.16	0.76	2169
				Pb-214	53.02	0.94	2084
				Ra-226	81.08	3.35	3188
45	Soil	25.3887	3,5 hrs	K-40	23.04	4.34	907
				Cs-137	0.5	0.13	19.7
				Pb-210	110.94	14.56	4369
				Bi-214	124.99	1.27	4923
				Pb-214	129.89	1.99	5116
				Ra-226	169.82	5.49	6688
				Th-231	26.3	2.04	1035
47	Soil	13.2453	4,5 hrs	K-40	6.86	2.86	517
				Bi-214	3.73	0.21	281

Annex B - Activity concentrations of radionuclides in plant and soil material after vacuum packing (continued)

Sample No	Sample type	Weight, g	Time	Radionuclide	Activity, Bq	Uncertainity Bq	Activity, Bq/kg
47	Soil	13.2453	4,5 hrs	Pb-214	3.3	0.15	249
50	Soil	17.0255	3,5 hrs	K-40	14.2	3.6	834
				Pb-210	189.08	24.64	11105
				Bi-214	184.84	1.61	10856
				Pb-214	189.43	2.85	11126
				Rn-219	17.12	1.09	1005
				Ra-226	155.21	14.83	9116
				Pa-231	20.39	3.69	1197
				Th-231	59.06	4.15	3468
				U-235	6.89	0.78	404
53	Soil	15.2462	5 hrs	K-40	8.37	2.76	548
				Co-60	0.89	0.07	58.3
				Cs-137	0.37	0.09	24.3
				Pb-210	6.31	1.04	413
				Bi-214	2.72	0.19	178
				Pb-214	2.29	1.53	150
1_3	Plant	8.6786	17 hrs	K-40	7.5	1.6	863
				Bi-214	2.35	0.1	270
				Pb-214	2.16	0.08	248
				Ra-226	3.98	0.76	458
				U-235	0.24	0.05	27.6
2_4	Plant	8.4748	17 hrs	K-40	16.8	1.7	1982
				Bi-214	2.2	0.1	259
				Pb-214	1.7	0.07	200
3_5	Plant	9.1429	17 hrs	K-40	10.9	1.64	1192
				Pb-214	0.59	0.07	64.5
Ref.mat	Soil	19.442	119 hrs	K-40	21.6	1.01	1111
				Cs-137	0.33	0.02	16.9
				Pb-210	1.63	0.25	83.8
				Bi-212	1.37	0.14	70.4
				Pb-212	1.77	0.05	91.0
				Bi-214	1.74	0.05	89.5
				Pb-214	1	0.04	51.4
				Ra-226	2.27	0.37	116
				Ac-228	2.09	0.06	107
				Th-234	0.72	0.14	37.0