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On release of radionuclides from a near-surface radioactive waste repository to the environment

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Abstract. A closed near-surface radioactive waste repository is the source of various radionuclides causing the human exposure. Recent investigations confirm an effectiveness of the engineering barriers installed in 2006 to prevent the penetration of radionuclides to the environment. The tritium activity concentration in groundwater decreased from tens of kBq/l to below hundreds of Bq/l. The monitoring and groundwater level data suggest the leaching of tritium from previously contaminated layers of unsaturated zone by rising groundwater while ^{210}Pb may disperse as a decay product of ^{226}Ra daughters.

Key words: near-surface repository • radionuclides • groundwater • radionuclide release • forest environment

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Introduction

In 1963, the near-surface 'RADON' type radioactive waste repository was installed in Bartkuškis forest about 30 km from Vilnius, the capital city of Lithuania, in a location with boggy environment sensitive to radionuclide migration in the biota [1, 2]. Despite the fact that the repository was closed in 1989, and reconstructed in 2006, the accumulated quantities of radionuclides (Table 1), especially of long-lived radioisotopes, remain dangerous from the long-term perspective. The monitoring wells were installed and the leakage of tritium from the vault to groundwater was monitored since 1993; the highest activity concentrations were measured in the monitoring well no. 4 located close to the vault in the prevailing groundwater flow direction (see Fig. 1 for monitoring wells positions); the levels of tritium activity in surrounding soil cores were assessed in 2005 when the maximum activity concentration in the soil moisture of about 100 kBq/l was determined at 8 m depth very close to the vault at newly established monitoring well no. 42 [3]. Follow-up experiments carried out from 2006–2008 confirmed the release of radiocarbon and plutonium (^{239,240}Pu) from the vault to groundwater: the maximum activity concentrations of (5.7 ± 0.7) Bq/l and (0.32 ± 0.04) mBq/l, respectively, were determined in the monitoring well no. 4 [4, 5]. The tritium activity concentration in the monitoring well no. 4 started to decrease due to an effectiveness of new barriers installed during the site reconstruction in 2006. Under the EU PHARE project, the French companies THALES and ANDRA together with the Lithuanian Energy Institute and the

Table 1. Radionuclides stored in the repository. Estimated inventory [6] is given at repository closing date (14.02.1989) and is also decay-corrected to February 2014

Radionuclide	Half-life [7] [y]	Activity [Bq]	
		February 1989	February 2014
^{3}H	12.312	2.75E+14	6.73E+13
^{137}Cs	30.05	5.56E+13	3.12E+13
^{60}Co	5.2711	7.21E+12	2.69E+11
^{90}Sr	28.80	6.41E+11	3.51E+11
^{239}Pu/Be	2.41E+03	5.99E+11	5.99E+11
^{239}Pu	2.41E+03	3.16E+11	3.16E+11
^{14}C	5.70E+03	1.77E+11	1.76E+11
^{226}Ra	1.60E+03	1.11E+11	1.10E+11
^{152}Eu	13.522	5.35E+10	1.49E+10
^{63}Ni	98.7	4.14E+10	3.47E+10
^{85}Kr	10.752	2.19E+09	4.37E+08
^{36}Cl	3.02E+05	1.20E+09	1.20E+09
^{238}U	4.468E+09	4.31E+07	4.31E+07
^{133}Ba	10.540	4.10E+06	7.92E+05
^{207}Bi	32.9	6.72E+05	3.97E+05

Institute of Physics assessed the technical state of the repository and evaluated its safety [6]. To prevent an increase of the precipitation infiltration, a vault capping system was proposed and implemented. The system of protective engineering barriers was comprised of two layers of very low permeability high-density polyethylene (HDPE) membranes with the durability not shorter than 30 years. It has been constructed on the ferroconcrete vault of the radioactive waste. Later on, when implementing the project "Tritium and radiocarbon transfer from the near-surface radioactive waste repository to the ground-level atmosphere" with the Research Council of Lithuania from 2011–2012, it was found that both tritium and radiocarbon may escape from the repository through the different pathway, namely, via evaporation through the soil layers. The radiocarbon activity concentration in groundwater (9.1 ± 0.6) Bq/l was determined in the monitoring well no. 4 in 2012. The total beta-activity measurements in groundwater from the wells no. 41 and 42 indicated variations at meaningfully higher level than it was observed in remaining wells. The gamma-ray spectrometry revealed the presence of ^{210}Pb at considerably higher activity concentrations than they could be expected in groundwater. In order to determine time-dependent activity concentrations of ^{210}Pb in groundwater, the sampling program was changed with special emphasis on higher volume monthly samples from the monitoring wells no. 41 and 42, beginning with March 2013.

The aim of this work is to discuss recently collected data concerning groundwater and bioindicators.

Methods applied

Since 2005, the groundwater level has been measured and groundwater samples for tritium analysis have been taken from all monitoring wells on a monthly basis. A plumb-line is used for groundwater level determination ensuring measurement uncertainty not greater than 1 cm. The main groundwater flow direction is calculated from these data (Fig. 1).

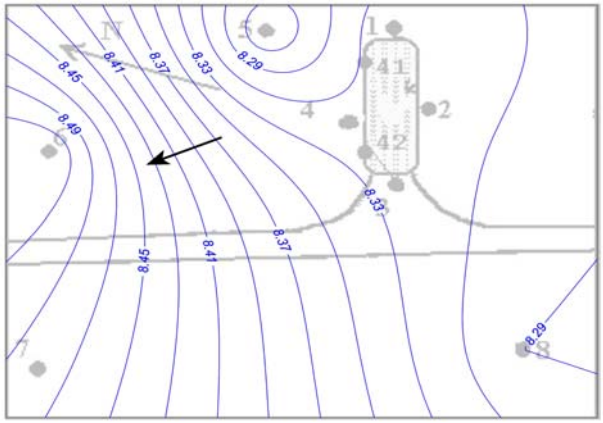

Fig. 1. Scheme of the site with installed monitoring wells no. 1–8, 41 and 42. The distance between monitoring wells no. 4 and 6 is 48.5 m. Grey lines encompass the asphalted area. Numbers at the isolines indicate the groundwater level. The groundwater flow direction determined on 4 September 2014 is shown by the shorter arrow.

The 0.5-l, 1-l and 10-l groundwater samples are taken for the analysis of tritium, radiocarbon and ^{210}Pb, respectively (Fig. 2).

Tritium and radiocarbon activity is measured with an ultra low-level liquid scintillation counter Quantulus-1220 from PerkinElmer. The external

Fig. 2. Groundwater sampling from monitoring well no. 42. Monitoring wells no. 4, 5 and 41 are well seen in the picture.

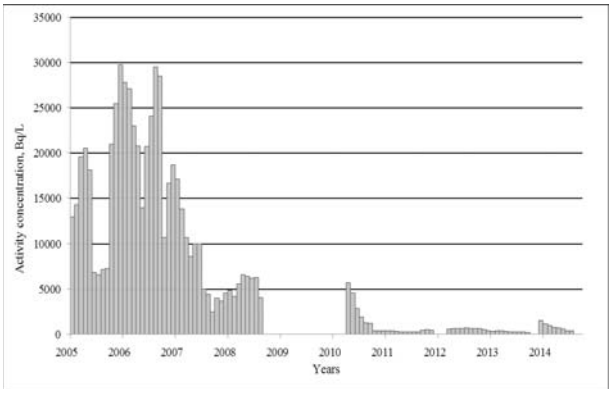

Fig. 3. Tritium long-term activity concentration in groundwater from monitoring well no. 4. Water was not found in the monitoring well in the periods September 2008 – March 2010, December 2011 – February 2012, October–November 2013 and August–September 2014.

standard technique is applied for quench correction of individual samples. Bioindicators are measured with a thin window coaxial HPGe detector while a well-type HPGe crystal is used for measurement of the residue after groundwater evaporation. The reference standards provided by the Czech Metrology Institute are used for efficiency calibration and yield determination.

Results and discussion

Tritium activity concentration in groundwater from the monitoring well no. 4 covering the period January 2005 – August 2014 is presented in Fig. 3. The highest activity concentration of (29 800 ± 1500) Bq/l was measured in the sample taken in December 2005. Long-term investigation results confirm an effectiveness of the new barriers installed in 2006. Application of low permeability membranes stopped the precipitation infiltration to the vault and resulted in decrease of radionuclide leaching and release to the environment to a considerable degree.

Groundwater level data in the monitoring well no. 5 from January 2008 to September 2014 are shown in Fig. 4. There was always water in well no. 5; not as the monitored well no. 4 during drying periods. It can be seen from Fig. 4 that the groundwater level course may have very different character through the years. Typically, the groundwater level has a maximum value in the spring, normally in April. This corresponds to a minimum distance between water found in a well and the soil surface. However, depending on the local precipitation intensity, there could be also other scenarios (e.g., years 2010 and 2012) when groundwater level continues to increase in summertime reaching in the second half the maximum values of a year. A rare scenario happens when groundwater level is very low (2009) causing drying of some wells. The changes in groundwater level can influence activity concentration of radionuclides in particular wells, when the rising groundwater came into contact with and washed soil levels contaminated by intensive leakages before the reconstruction in 2006. This effect can be noticed from tritium data in Fig. 3.

Recent results of measurements of some gamma-ray emitters in bioindicators are summarized in Table 2. The activity concentration of the fission product ^{137}Cs is typical for Lithuania forest environment and is caused by a global distribution of this artificial radionuclide. No considerable influence from the repository could be attributed to these data.

The activity concentration of ^{210}Pb in groundwater taken from wells closest to the vault is shown in Fig. 5. ^{210}Pb is the decay product of ^{238}U-series with a half-life of 22.23 years [7]. It is known from the literature that ^{210}Pb may be found in groundwater and drinking water, thus causing an extra exposure to humans. The reference activity concentration of 200 mBq/l and 100 mBq/l in drinking water for ^{210}Pb and ^{210}Po, respectively, is set by the document [8]. According to this document, if the actual concentration of ^{210}Pb exceeds this value recommended by European Commission, consideration should be

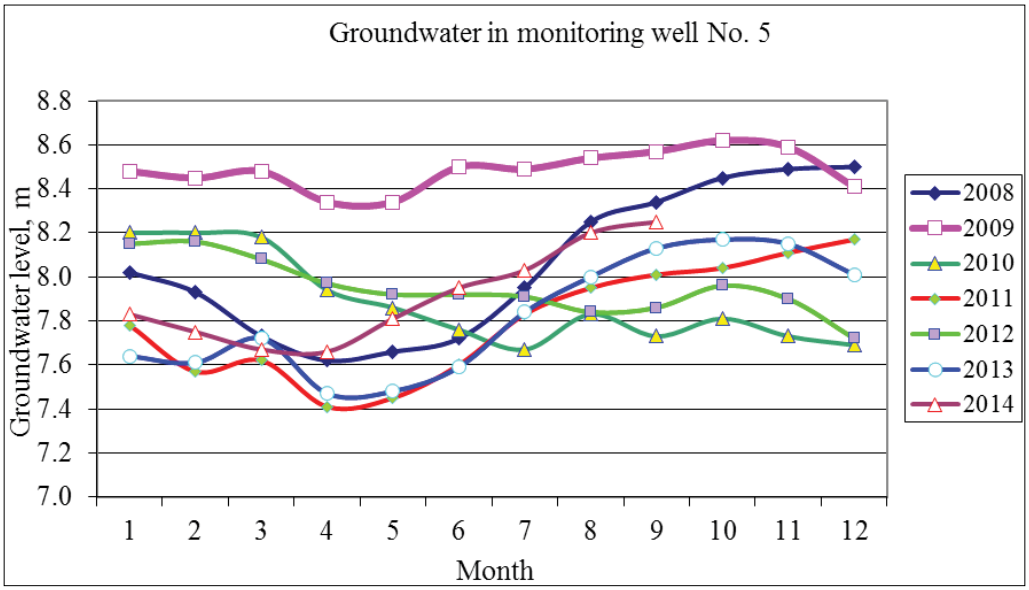

Fig. 4. Groundwater level variations in monitoring well no. 5.

Table 2. ^{40}K and ^{137}Cs activity concentrations in bioindicators [Bq/kg fresh wt]

Analyzed species	2013		2014	
	^{40}K	^{137}Cs	^{40}K	^{137}Cs
Woodland strawberries (*Fragaria vesca*)	54.7 ± 8.2	2.0 ± 0.1	73.2 ± 8.8	< 4.8
Alpine cranberries (*Vaccinium vitis-idaea*)	29.3 ± 4.7	3.3 ± 0.1	37.7 ± 4.6	4.6 ± 0.2
Bilberries (*Vaccinium myrtillus*)	–	–	11.2 ± 1.4	< 1.7
Common hazelnuts (*Corylus avellana*)	–	–	178.0 ± 18	3.8 ± 0.4
Mushrooms 1 (*Boletus edulis*)	–	–	78.4 ± 7.9	6.0 ± 0.2
Mushrooms 2 (*Boletus luteus*)	–	–	68.3 ± 6.9	8.9 ± 0.3
Mushrooms 3 (*Leccinum scabrum*)	–	–	73.4 ± 7.9	2.1 ± 0.1
Mushrooms 4 (*Russula flava*)	–	–	60.8 ± 7.3	25.8 ± 0.8

'–' means 'not measured'. The uncertainties are given with the coverage factor $k = 1$.

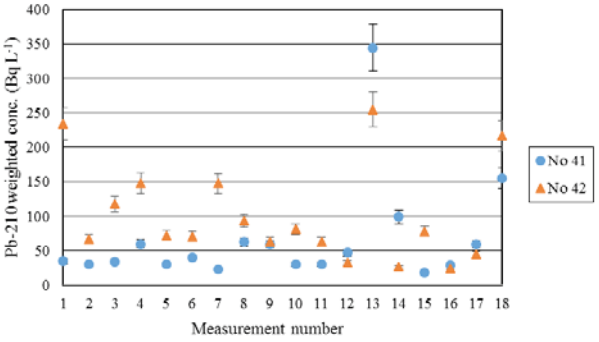

Fig. 5. Variation of ^{210}Pb activity concentration in monitoring wells no. 41 and 42.

given to whether the remedial action is needed to protect human health. The literature data demonstrate that ^{210}Pb activity concentration in water in different countries is usually far below 200 mBq/l [9–12]. Our results showed that ^{210}Pb mean activity concentration in groundwater from the monitoring wells no. 41 and 42 was 102 mBq/l and 66 mBq/l, respectively, during the research period. Moreover, the reference value of 200 mBq/l was exceeded in four cases. We can see from Fig. 5 that sharp peaks of activity concentration may be expected in the spring during groundwater rising periods (note values at no. 1 and 13 corresponding to samples collected in March 2013 and 2014, respectively). At the same time, the results obtained from samples collected in August 2014 (values at the number 18 in Fig. 5) suggest the problem that should be further monitored and carefully investigated to make any explanation. At present, it might only be supposed that enhanced ^{210}Pb activity concentrations in groundwater close to the vault are probably governed by the dispersion and radioactive decay of radon gases ^{222}Rn which come from the vault, in turn, as a decay product of ^{226}Ra.

Conclusions

Original constructions of the near-surface radioactive waste repository have been located in complicated environmental conditions for more than 50 years. Systematic monitoring data show that the stored radionuclides are yet being released into local environment at sufficiently short distances from the vault. However, a gradual degradation of the physical installation beneath the earth surface poses serious problems from the long-term perspective. The radioactive wastes of the repository should be removed to the more appropriate storage facility.

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