

RESULTS OF TRITIUM MONITORING IN THE ENVIRONMENT OF THE MAIŠIAGALA RADIOACTIVE WASTE REPOSITORY

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ABSTRACT. A shallow-land radioactive waste storage facility was commissioned in 1963 in an ecologically sensitive locality ~30 km from Vilnius, Lithuania. To ensure radiation safety, monitoring activities have been performed that take into account the migration of tritium. Tritium analysis in groundwater, soil moisture, and plants indicates obvious leakage from the repository. Liquid scintillation counting (LSC) involving an ultra low-level device, Quantulus™ (Wallac), is used for tritium measurement. By 2005, there were 8 monitoring boreholes installed in different directions and distances from the vault. In January 2005, 2 new boreholes were installed 0.5 m from the wall of the vault. Soil samples were taken every 0.5 m to a depth of 13.5 m, and tritium was determined in all samples from both boreholes. Maximum activity concentration (~100,000 Bq/L) was measured at a depth of 8–8.5 m. Analyses of some LSC spectra revealed possible contamination of groundwater by ¹³⁷Cs, and this has been confirmed by low-level gamma spectrometry. The storage facility requires continuous monitoring and management in the future.

INTRODUCTION

The Maišiagala storage facility was built according to the project TP-4891. The storage facility is a radon-type repository providing a 200-m³ disposal capacity for solid radioactive waste, which was placed in a concrete vault with the dimensions 15 × 5 × 3 m (International Atomic Energy Agency 1998). Radioactive waste disposed of at the repository originated chiefly in hospitals, industrial sites, and research centers. The waste is composed of gamma-ray sources of high activity, charge neutralizers, chemical compounds with radioactive tracers, smoke detectors, and similar waste. The repository closing was performed by the Institute of Physics. The filled compartments were sealed with concrete plates, and the surface was covered by a ~100-mm-thick equalizing concrete layer capped with 2 layers of hot bitumen and 50-mm-thick asphalt, and finally, with a 1.2-m-thick sandy soil layer. At the time of its closure in 1989, the total amount of radioactive substances disposed was estimated to be 1 PBq (Lithuanian Energy Institute 2004). Table 1 indicates the known inventories of the radionuclides with half-lives greater than 1 yr recalculated to February 2005.

Radiological surveillance and control of the site on a regular basis has been carried out since 1993 (Institute of Physics 1994, 1997, 2003, 2004, 2005). The radiological program at the repository site comprises the following items:

- Dose rate measurements;
- Monitoring tritium activity concentration in groundwater;
- Monitoring tritium activity concentration in soil moisture and plants growing at the site;
- Gamma spectrometry of groundwater, soil, and plant samples;
- Gamma spectrometry in situ utilizing a portable HPGe detector;
- Total beta activity determination in groundwater samples.

In 1994, monitoring of tritium activity concentrations in environmental samples confirmed tritium leakage from the vault. First, the enhanced tritium level was determined in water from the surrounding bog and in the grass growing near the vault.

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Table 1 Inventory and decrease of activity of some radionuclides since the closure of the repository in 1989. Half-life values taken from Magill (2003).

Nuclide	Half-life (yr)	Inventory (kBq)	
		February 1989	February 2005
³ H	12.33	2.51E11	1.02E11
¹³⁷ Cs	30.07	5.57E10	3.85E10
⁶⁰ Co	5.27	7.21E9	8.79E8
²⁰⁴ Tl	3.78	6.77E9	3.60E8
²³⁹ Pu	24,110	9.15E8	9.15E8
⁹⁰ Sr	28.84	6.45E8	4.39E8
¹⁴ C	5730	1.77E8	1.77E8
²²⁶ Ra	1600	1.11E8	1.10E8
¹⁵² Eu	13.54	5.35E7	2.36E7
⁶³ Ni	100.1	4.14E7	3.71E7
¹⁴⁷ Pm	2.62	1.07E7	1.55E5
⁸⁵ Kr	10.76	2.20E6	7.85E5
³⁶ Cl	3.01E5	1.20E6	1.20E6
¹²⁵ Sb	2.76	6.14E5	1.10E4
¹⁰⁹ Cd	1.27	2.26E5	3.64E1
⁵⁵ Fe	2.73	1.85E5	3.18E3
²² Na	2.60	1.38E5	1.94E3
²³⁸ U	4.47E9	4.31E4	4.31E4
¹⁰⁶ Ru	1.02	2.19E4	4.15E-1
¹³³ Ba	10.51	4.10E3	1.43E3
¹³⁴ Cs	2.06	2.83E3	1.30E1
²⁰⁷ Bi	31.55	6.72E2	4.73E2
Total		3.23E11	1.44E11

Rank (1992) mentioned several applications of tritium in hydrology. One of them, estimating possible pollutant transfer, can be successfully developed while evaluating the safety of radioactive waste disposal facilities. Baltrūnas et al. (2001) used experimental data and modeling to predict tritium activity concentration in groundwater from the Maišiagalas site with a time scale of a few hundred years. In Mažeika et al. (2003), the main processes of radionuclide migration in unsaturated and saturated zones have been characterized in connection with the aquatic system of the Maišiagalas storage facility. In another study (Butkus et al. 2003), an attempt was made to assess the possible leakage of some important artificial nuclides. However, no leakage of other nuclides, except tritium, was clearly seen by 2005. In this work, results of tritium determination in groundwater and soil moisture samples are presented with special emphasis on recent monitoring data.

METHODS

To detect a possible migration of radionuclides from the vault via the groundwater, observation boreholes were installed around the vault (see Figure 1). Borehole #1 was installed in 1989; #2, #3, and #4 in 1993; #5–8 in 1999; and #41 and #42 in January 2005.

Environmental samples, including groundwater, were gathered on a quarterly basis until 2005. It was known that activity concentration of ¹³⁷Cs in groundwater varied from 0.001–0.06 Bq/L (But-

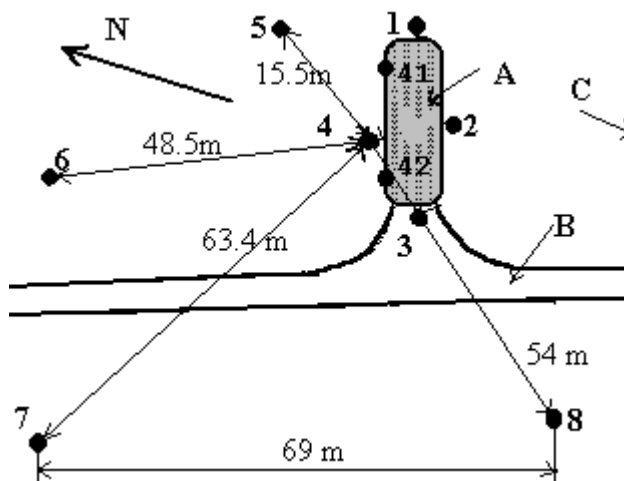


Figure 1 Scheme depicting boreholes around the vault: A—the vault, B—asphalt path, C—metallic fence; 1–8, 41, 42—the boreholes.

kus et al. 2003), hence preconcentration procedures were necessary to measure this radionuclide. Since January 2005, the sampling has been performed every month.

The new boreholes #41 and #42 were installed very close to the wall to detect possible leakage of nuclides other than tritium. Drilling was performed until the impermeable clay layer was reached. In both boreholes, the impermeable layer was found at the depth of 13.5 m. Two pipes were installed in each of the boreholes to monitor tritium content in water samples from the aquifer's upper and lower layers. The water samples collected from the upper layer at the boreholes #41 and #42 were labeled #41p and #42p, respectively. The soil samples were taken from newly made cores and analyzed in the laboratory for tritium content in soil moisture as well. Gamma spectrometry, followed by the analysis of ^{90}Sr and $^{239+240}\text{Pu}$ in selected samples, was also applied.

Liquid scintillation counting (LSC) with the ultra low-level device Quantulus™ (Wallac) was used for quantitative determination of tritium. Prior to measurement, water was extracted from the original soil by the distillation procedure. The setup for soil distillation is shown in Figure 2.

Extracted water was mixed with the commercial cocktail OptiPhase TriSafe. The mixing ratio applied was 4 mL of water to 16 mL of scintillation cocktail. For efficiency calibration, the tritiated water reference standard solution provided by the IAEA was used. While measuring samples, the counting efficiency was corrected for quenching effect by the external standard technique. Ground-water samples were also mixed with the commercial cocktail OptiPhase TriSafe. Two different ratios (12+8 and 16+4) were applied. To minimize the risk of contaminating the measuring device, the ratio 16+4 was applied for presumably active samples (#41p and #42p), while the ratio 12+8 was used for low-level samples, due to having a better minimum detectable activity (MDA) value. The MDA (in Bq/L) was calculated according to Currie's (1968) approach:

$$(2.71 + 4.65\sqrt{N_B}) / (t \epsilon V)$$

where N_B is the background counts, t is the counting time (s), ϵ is the counting efficiency (cpm/dpm), and V is the sample volume (L). The MDA is 0.8 Bq/L for a 2800-min counting time, and the cocktail-to-sample ratio is 12+8.

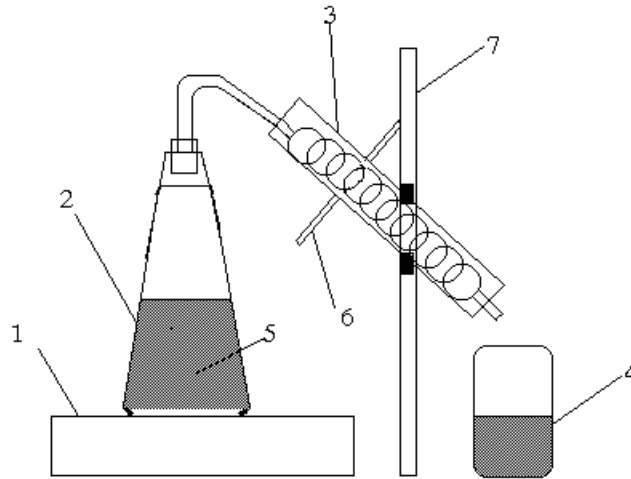


Figure 2 Scheme for water extraction from the soil: 1-hot plate; 2-reactor; 3-cooling device; 4-glass for collecting water; 5-soil sample; 6-water for cooling; 7-support.

In order to establish a quench curve, CCl_4 was used as the quenching agent. The quench curve is shown in Figure 3.

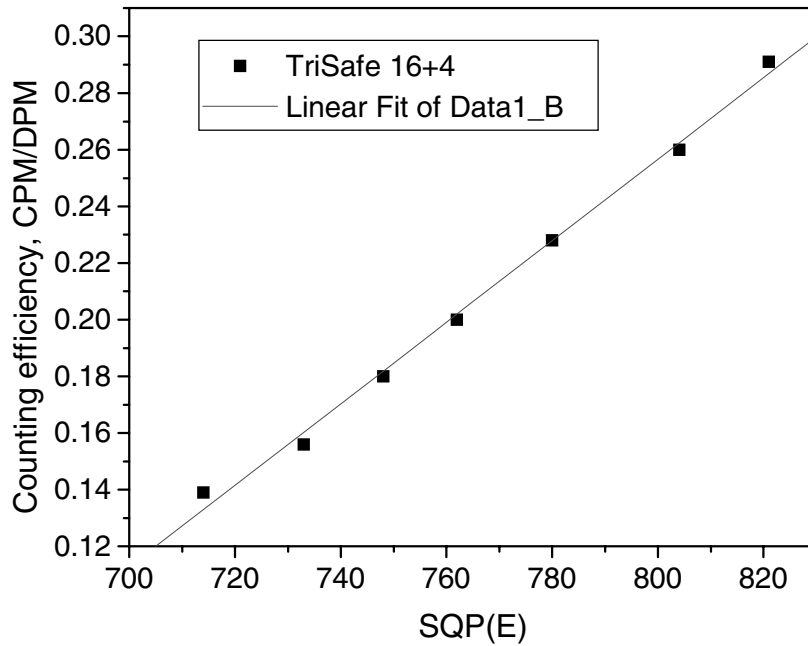


Figure 3 Quenching curve for tritium determination using the OptiPhase TriSafe cocktail

The ^{137}Cs activity concentration in water from the samples #41p and #42p was determined via a gamma spectrometer with a HPGe well-type detector. The spectrometer could accommodate samples of up to 4 cm^3 volume; the counting efficiency for the 661.7-keV line was 13.6% (Gudelis et al.

2000). Original water samples, not preconcentrated, were measured. The background gamma spectrum is shown in Figure 4. Radiation of the natural radionuclides ^{40}K , ^{212}Pb , ^{214}Pb , ^{214}Bi , ^{226}Ra , ^{228}Ac , ^{208}Tl , as well as an annihilation line at 511 keV, can be found in the spectrum. There are no traces of ^{137}Cs present in the background.

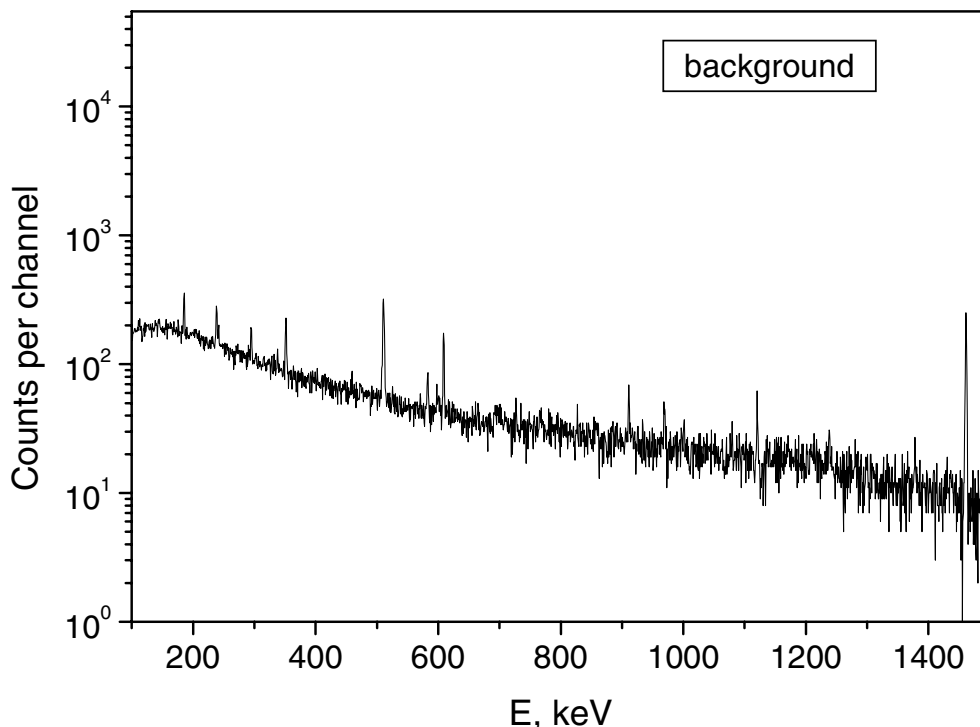


Figure 4 Background spectrum of an HPGe well-type detector. The acquisition time was 50,000 s.

RESULTS AND DISCUSSION

The uncertainty of the results in all figures and Table 2 is reported with a $1-\sigma$ error. Variation of tritium content in soil moisture in 2 cores is shown in Figures 5 and 6.

The results confirm the leakage of tritium from the vault to the surrounding environment. They are consistent in magnitude (10^5 Bq/L) with previous investigations (Institute of Physics 1997; Mažeika et al. 2003). Generally, the distribution of tritium with depth has 2 maximums; the first maximum corresponds to the bottom of the vault. The second maximum at 8–8.5 m occurs, evidently, at the start of a saturated zone matching the upper groundwater layer (Figure 7).

Enhanced tritium activity concentration in groundwater was detected in borehole #4. According to constant water-level measurements, borehole #4 is located in a downstream groundwater flow (Butkus et al. 2003; Mažeika et al. 2003) and consistently exhibits the highest tritium concentration. The maximum annual values since 1996 to 2004 are shown in Figure 8. In 2005, the activity concentration varied from 6590 Bq/L in July to 20,500 Bq/L in April. The concentration decreases in summer-time most probably due to dilution by infiltration recharge following rain precipitation.

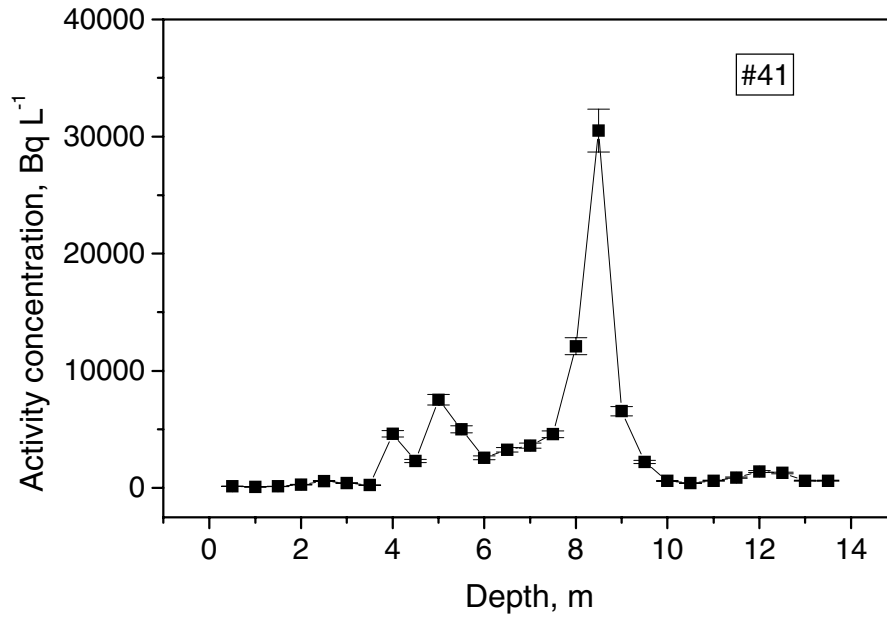


Figure 5 The course of tritium activity concentration in soil moisture from core #41

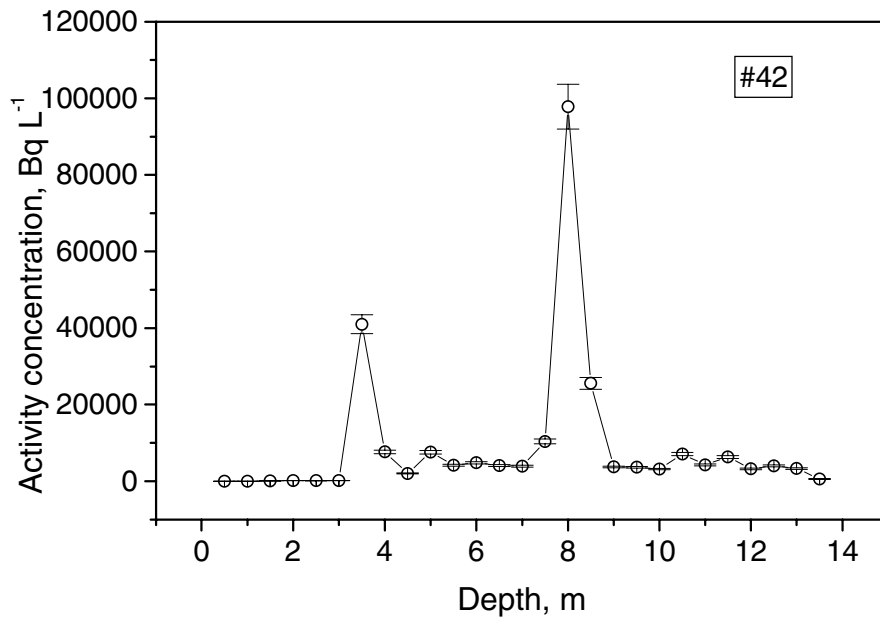


Figure 6 The course of tritium activity concentration in soil moisture from core #42

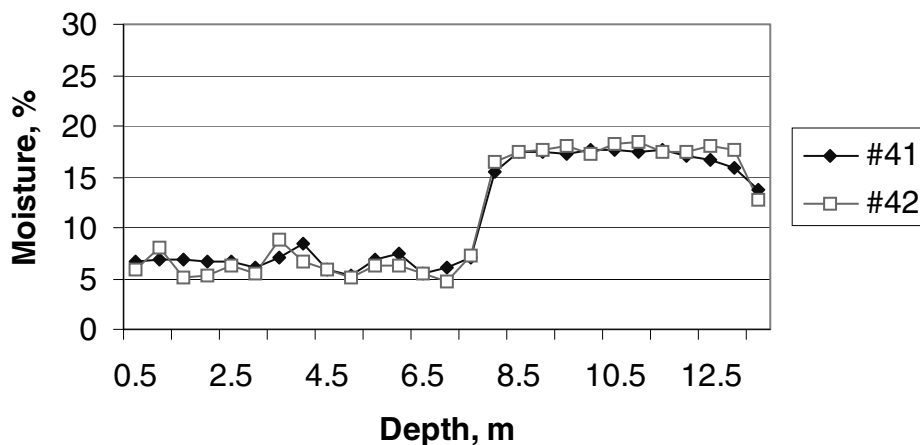


Figure 7 Moisture distribution in the cores

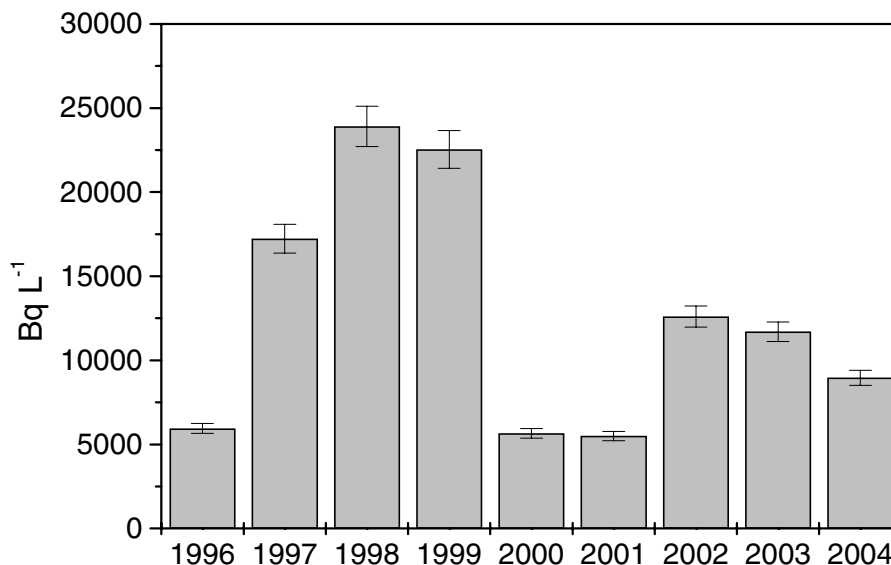


Figure 8 Maximum annual tritium activity concentration in groundwater from borehole #4

In the boreholes #41 and #42, one can observe how tritium migrates from the upper part of the saturated zone to the deeper groundwater layer (Figures 9–10). Tritium activity concentration in both #41 and #42 was below 10 Bq/L at the start of observation (January–April for #41; January–March for #42). Later on, tritium reached the deeper groundwater layer, and its concentration suddenly increased by ~230 and 400 times, respectively, in bores #41 and #42. It should be noted that an increase occurred at a different time in the bores #41 and #42.

Pronounced ¹³⁷Cs activity in environmental water close to the repository was determined by LSC. One cannot expect to detect ¹³⁷Cs in an ordinary 8-mL water sample. The usual procedure for ¹³⁷Cs determination in water involves evaporating 10 L of water followed by analysis of the precipitation by high-resolution gamma spectrometry. However, while measuring tritium in water taken from the upper layer of the saturated zone at newly established boreholes, an increase in the counting rate was

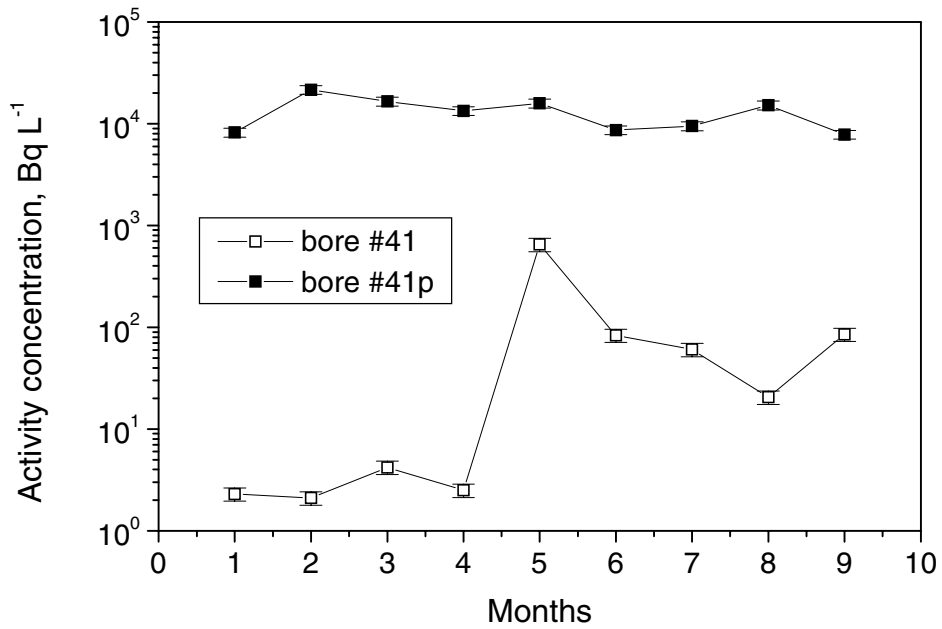


Figure 9 The variation of tritium activity concentration in borehole #41 in 2005

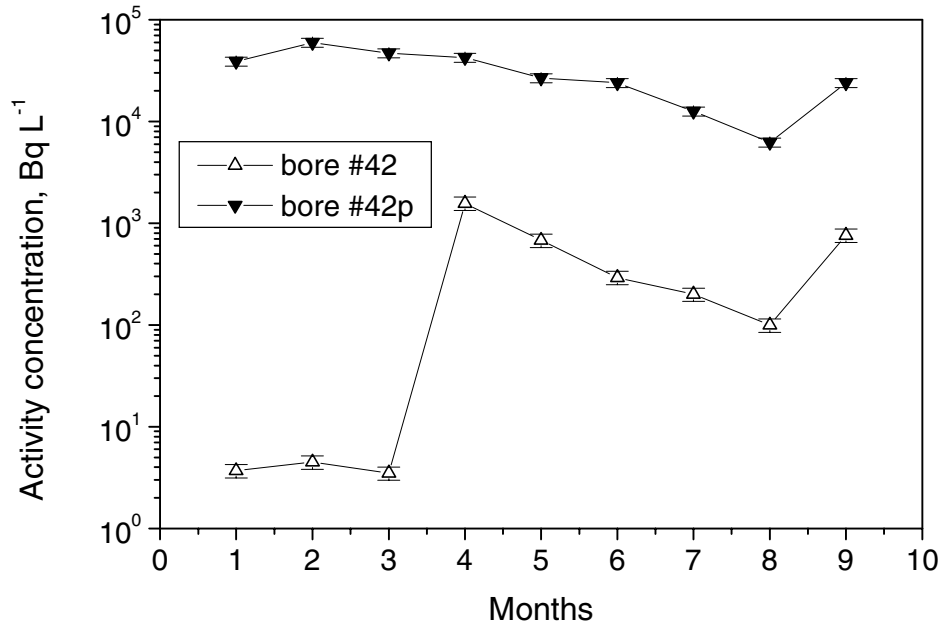


Figure 10 The variation of tritium activity concentration in borehole #42 in 2005

observed in the channel window between 400 and 800 (Figures 11–12). The presence of a beta emitter was suspected in the sample. An original aliquot of 4 mL was measured via a high-efficiency gamma spectrometer with an HPGe well-type detector, and ¹³⁷Cs was easily detected (Figure 13). The leakage of an artificial nuclide other than tritium has been proven by this measurement. Moni-

toring of ^{137}Cs activity concentration in the upper groundwater layer demonstrated its variation with time (Table 2). Again, the decrease in the activity concentration of ^{137}Cs can be explained by the dilution of groundwater by rain during the summer. Longer time-scale investigations are needed to interpret the data more explicitly.

[A] 16.000 CPM/ch 1242.88 min C:\ARUG\BAR5-32\Q012700N.000 SP#12

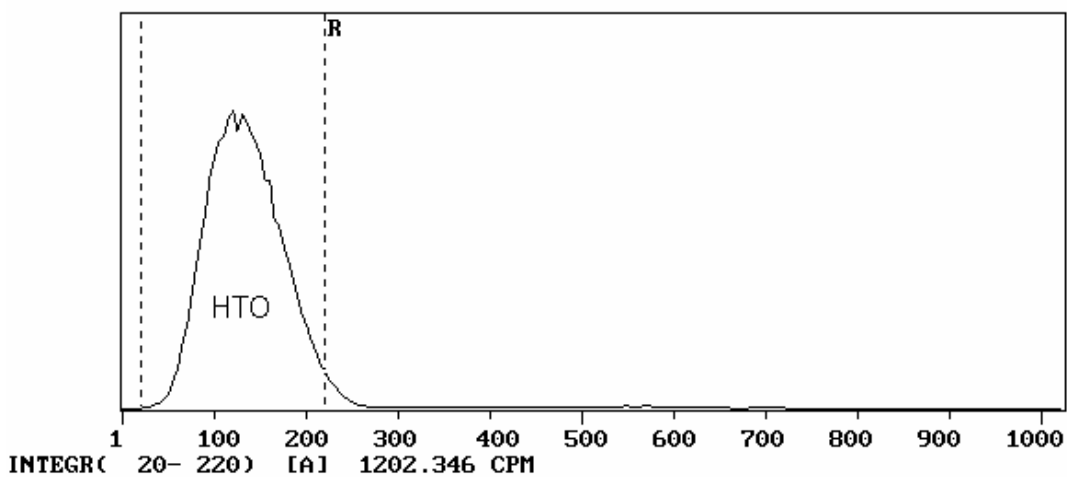


Figure 11 LSC spectrum of a water sample from borehole #41p taken 25 March 2005

[A] 0.400 CPM/ch 1242.88 min C:\ARUG\BAR5-32\Q012700N.000 SP#12

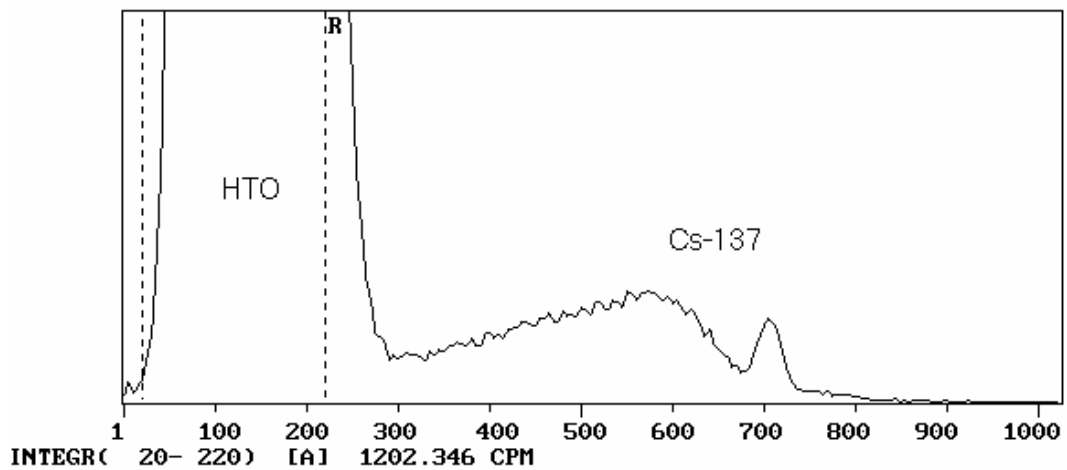


Figure 12 The same spectrum as in Figure 11 re-scaled 40 times to show the presence of ^{137}Cs

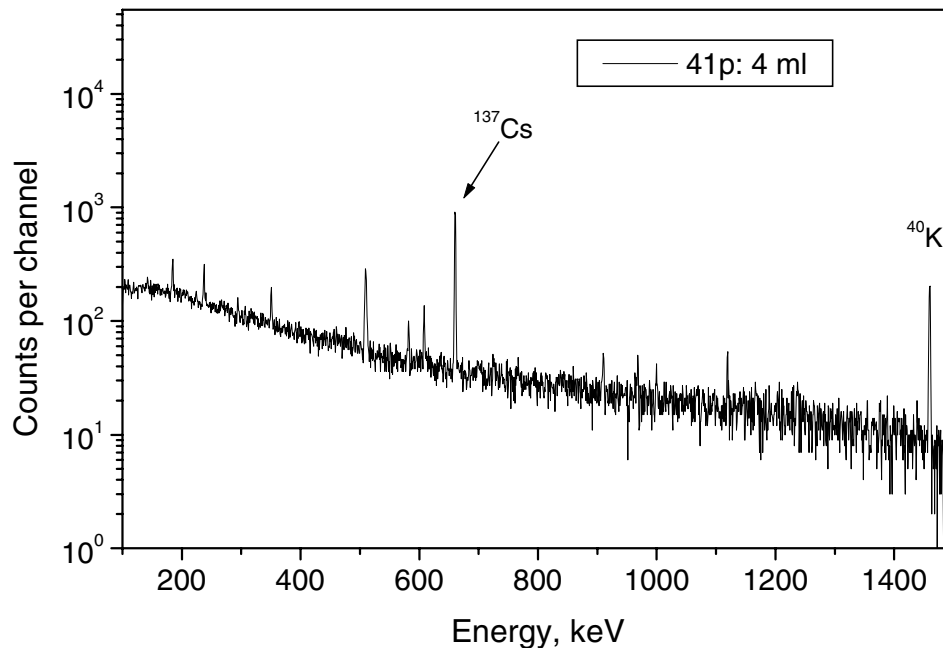


Figure 13 Gamma spectrum of water sample taken 22 April 2005. The acquisition time was 50,000 s.

Table 2 Variation of the activity concentration of ^{137}Cs in 2005.

Sampling date	Activity concentration (Bq/L)	
	Sample #41p	Sample #42p
18 February	473 ± 14	596 ± 18
25 March	123 ± 6	7.6 ± 1.3
22 April	120 ± 6	25.5 ± 1.5
27 May	93.7 ± 5.6	18.4 ± 1.5
27 June	62.7 ± 3.8	15.8 ± 1.0
8 July	4.1 ± 1.2	1.6 ± 0.6
5 August	1.8 ± 0.7	11.3 ± 1.4
12 September	8.4 ± 1.6	1.2 ± 0.6

CONCLUSIONS

The profile of the tritium activity concentration in soil from the 2 new boreholes indicates obvious leakage from the vault. The results are consistent with values reported in previous investigations. Leakage of ^{137}Cs is demonstrated by the direct measurement of an original water sample from the upper saturated zone. Monitoring carried out on a monthly basis reveals considerable variation of tritium and ^{137}Cs activity concentration with time. A decrease in the activity concentration of both radionuclides can be explained by the dilution of groundwater caused by rain during the summer. The results implicate the need for a good vault capping system to prevent an increase of water infiltration and radionuclide leaching from the vault. Measurements of other radionuclides present in the vault (e.g. ^{14}C , ^{63}Ni) can be considered to be significant as well and provide more specific information on the vault integrity and radionuclide containment functions.

ACKNOWLEDGMENTS

We are grateful to our colleagues G Kandrotas and J Podoroga for their technical assistance in sampling and preparation of samples for measurement. The new boreholes have been installed within the PHARE project No. 632.06.01 "Safety Assessment and Upgrading of Maišiagala Repository in Lithuania." Financial assistance from the Radioactive Waste Management Agency (RATA) under the contracts with the Institute of Physics in 2003–2005 is also acknowledged.

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