Experimental research on the effects of radioactive waste repository upon groundwaters

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Abstract. The study was performed in Maišiagala radioactive waste repository where tritium (³H) spreading into the environment via groundwaters and the resulting pollution caused a major problem. To solve the pollution problem the repository underwent an upgrade in 2006. In the article studies of volume activity of ³H in the groundwater performed in March and October 2009 are presented. The obtained results are compared to the outcome of the research performed in the repository the previous year, i.e. before upgrading. Both in March and October 2009 borehole 4 happened to be dry, which prevented a direct comparison between the measured tritium volumetric activity in the respective borehole water and the values measured earlier. In 2005, prior to upgrading, the highest value of two measurements performed the same year for ³H volumetric activity of ³H in boreholes 4.1 and 4.2 were determined as $650 \pm Bq \cdot I^{-1}$ and $1570 \pm Bq \cdot I^{-1}$, respectively. The volumetric activity of ³H in borehole 4.1 established by experimental studies in March was equal to $154.9 \pm 4.5 Bq \cdot I^{-1}$, which is 17 times greater the value measured in the same borehole in October. The measured ³H volumetric activity in borehole 4.2 decreased from March to October 11 times, namely from $58.6 \pm 3.2 Bq \cdot I^{-1}$ down to $5.0 \pm 2.3 Bq \cdot I^{-1}$. Such a striking change might be attributed to a significant reduction of tritium leak to groundwaters.

Key words: radioactive waste repository • tritium (3H) • groundwater • RADON type burial-grounds

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Introduction

A safe burial of radioactive waste is a topical and complex issue. Radioactive waste shall be managed in a way minimising the consequences for future generations to the level the current generation regards acceptable for themselves. Thus, it is necessary to ensure that safety criteria for radioactive waste burial-grounds are met not only over the next few decades, but in dozens, or even thousands of years [6].

The burial of radioactive waste is the last step in the chain of radioactive waste disposal. When burried improperly radionuclides are more likely to get to the environment, thereby the risk of damaging nature and harming human beings arises. In geophysical, geochemical and biological environmental processes radionuclides participate in the exchange of chemical elements (via metabolism) which is characterized by two opposite, though related, to each other tendencies of material movement, i.e. dispersal of radionuclides in some components and their accumulation in other components of the geosystem. The potential influence of ionizing radiation, which starts upon closing burial--grounds, can rise due to processes that take part gradually, such as erosion of barriers, or some discrete events that can deteriorate the waste isolation [5].

At present, the assessment of radionuclide migration from RADON type burial-grounds is particularly relevant. RADON type burial-grounds are typical for the former Soviet Union and East European countries. Built in the 70ties and 80ties of the past century, they are standard burial-grounds, where radioactive waste was put into reservoirs, namely concrete basements or pipes [12].

Burial-grounds of that type were built in Belarus, Czech Republic, Bulgaria, Latvia, Russia and Ukraine. They were designed and built with none or little attention paid to long-term safety, so all such tasks have arisen now. The main problems concern radionuclides spread by groundwater and related soil pollution. One of the problems of RADON type repositories is their deteriorated sealing (tightness) as time progress and other climate and wear-related factors, such as rain and construction wear, act. For such reasons radioisotopes ¹⁴C, ³⁶Cl, ⁶⁰Co, ³H, ²³⁹Pu, ²²⁶Ra, ⁹⁰Sr and ²³⁸U penetrated into groundwaters at the RADON type Ekores radioactive waste repository of Belarus. They might have penetrated into drinking water as well. Long-life radionuclides such as ²³⁹Pu, ²²⁶Ra, ²⁴¹Am and ²³²Th not only pose danger to future generations, but also may contaminate the surrounding strata [17].

The research has been aimed at determining experimentally the dispersal of tritium from the repository heap to the groundwater, analyzing the obtained results and comparing them to the results recorded prior to and after the repository upgrade.

The object and methods of the study

Taking into consideration the risk caused by burialgrounds of RADON type, a burial-ground built in Bartkuskis grove near Maišiagala was chosen for the study. Maišiagala radioactive waste repository is a RADON type repository where radioactive waste is buried without containers. The repository is located at the distance of 7 km northwest of Maišiagala town in Širvintos district. The repository territory belongs to the Baltic hills, formed by the last glaciation, and lies in the river basin of the Juoda stream [8]. It leans against a swampy lowland, having the status of Bartkuskis Telmological Natural Reserve (Fig. 1).

Radioactive waste of industrial, medical and scientific origin was transported to Maišiagala repository



Fig. 1. The location of Maišiagala radioactive waste repository (scale 1:200).



Fig. 2. 200 m³ basement of RADON type burial-ground.

from Lithuania, Kaliningrad and Grodno regions and was accumulated there until 1989. Since the waste was not sorted there, it meant storing both short-term and long-term waste. According to the present regulations, the waste, regardless the type, must be buried separately in special packages and at suitably equipped burial-grounds. At the closing time about a volume of 120 m³ of waste was accumulated there, i.e. the repository has been filled up to 60% of its total capability. The remaining space has been filled with sand, concrete, covered with bitumen, asphalt and topped with a thick earth layer. A number of radioactive waste carrying both technogenic and natural nuclides, namely ³H, ¹³⁷Cs, ⁶⁰Co, ²⁰⁴TI, ²³⁹Pu, ⁹⁰Sr, ¹⁴C, ²²⁶Ra, ¹⁵²Eu, ⁶³Ni, ¹⁴⁷Pm, ⁸⁵Kr, ³⁶Cl, was buried there. Activity during conservation exceeded 1 GBq.

The hard radioactive waste repository is shaped as a monolithic reinforced concrete reservoir of $5 \times 15 \times$ 3 m (see Fig. 2). The repository has a practical capacity of 200 m³. It is located underground, with the bottom level at 3 m below the surface.

Radionuclides are found everywhere, both around and within us [11]. The mobility and fate of radionuclides in the environment depends to a great extent on the speciation and transformation they undergo [9]. Physical and chemical properties of radioactive substances and components of the environment determine the environmental behaviour of radionuclides [18]. Bartkuskis burial-ground is built in a geologically sensitive area, at a swampy site. The main hydrogeological characteristic of Maišiagala repository, affecting radionuclides transfer from the burial-ground heap of radioactive waste, is a considerable thickness of the aeration area and a small thickness of groundwater horizon (2.0–2.5 m). In such cases processes occurring in the aeration zone are crucial. To study these processes we measured the main direction of groundwater flow with relation to the groundwater levels and their changeability in boreholes [10].

Ionizing radiation is one of harmful impacts affecting mankind [3]. Great attention shall then be paid to tritium isotope (³H) [14]. The major sources of technogenic tritium are nuclear facilities, along with manufactures of luminous consumer products, as well as their disposal [16]. This radionuclide, as well as carbon-14, krypton-85, iodine-129, belongs to the risk group of radionuclides [1]. Tritium is of environmental importance because it is released from nuclear facilities in relatively large quantities and because of its half-life of 12.26 y [2]. Due to tritium small energy, it is attached to radionuclides that are difficult to measure. When ³H and hydrogen and oxygen atoms combine into a molecule, water marked by tritium is formed. ³H can enter a human by inhaling, swallowing, or through the skin. The tritium water is then distributed with blood



Fig. 3. Newly-installed barrier system of the repository (scale 1:100).

into the fluids all over the body, the same way as simple water is. While decaying tritium causes soft tissues to be irradiated. Thus, the presence of ³H in human increases the effective irradiation dose value. The effective average half-life activity of tritium is only 10 days, still, it takes months until it is completely eliminated from the body. Tritium water forms are well assimilated by organisms [14].

Precipitation waters when infiltrated into the repository provide a mobile agent to radionuclides, hence water infiltration shall be prevented. French companies created a model of engineering barriers, the basis of which consist in two water-tight membranes laid upon the top of the repository that prevents water from infiltrating (Fig. 3).

A water retention function is performed by the upper membrane, stretching 5 m side-ways from the edges of the repository. Another controlling membrane is designed to assess the condition of the upper one. It drains the water penetrating the upper membrane into a special container. The performance of the upper membrane can be determined by the amount of water accumulated there [15].

The main agents that transfer radionuclides are ground moisture and groundwaters [4]. To control a likely migration of radionuclides from the heap, 8 monitoring boreholes were drilled around the repository to take groundwater samples. Later, two more boreholes were drilled, each at a 0.5 m distance from the heap wall; those were marked as 4.1 and 4.2. Borehole 4.1 lies between the old boreholes assigned as 1 and 2, while borehole 4.2 is located between old boreholes marked as 4 and 3 (Fig. 4).



Fig. 4. Repository and groundwater boreholes. A – repository; B – asphalt road; C – fence; D – controlled area; E – wells for controlling observation; 1, 2,... 8; 4.1, 4.2 – boreholes numbers.

As groundwater level tends to drop down in autumn and rise in spring, it was decided to carry out the studies in March and October. In order to determine volumetric activity of tritium groundwater, samples were taken in March and October 2009. The groundwater samples were taken from special boreholes drilled outside the heap (Fig. 4). A special vessel lowered down the borehole by rope, has been used for scooping groundwater. The collected water was poured into two 20³ ml glass vessels, that were labelled with a water-resistant slate-pencil as the number of the sampled borehole. The samples were taken to the laboratory, where radiological tests were carried out.

A fluid scintillation method is generally used to determine the volumetric tritium activity in groundwater samples. Trisulphate is put into the water sample, then alkaline solution is added and the whole is distilled. Equal parts of the distilled matter and the scintillated solution are mixed in a counter vessel. In the obtained mixture (mostly emulsion) the kinetic energy of beta particles of tritium partly turns into photons. These photons are considered as impulses and the counting rate provides the measure of tritium volumetric activity.

Tritium splits into helium emitting beta particles of the greatest energy of 18.6 keV. It takes tritium 4540 days (12 y and 43 d) to reach half-life.

The volumetric activity of 3 H in the sample is calculated using the Eq. (1):

(1)
$$C = \left[\frac{\overline{R} - \overline{R_0}}{\varepsilon \cdot V_2} + c_0(t)\right] \cdot e^{\lambda \Delta t}$$

where: C – stands for tritium volumetric activity in the sample in Bq/m³ at sample collection; \overline{R} – stands for the average value of calculation frequency impulses per second when two uniform samples are taken. No other standard solution is added; R_0 – stands for the average calculation frequency value impulses per second of the two neighbouring background water samples; ε – efficiency of calculation; V_2 – the volume of the sample or of the background water sample in a vessel (m³); λ – the splitting constant in years⁻¹ (λ = 0.05576); Δt – a yearly interval between the sample collection and calculation; $c_0(t)$ – tritium volumetric activity in Bq/m³ in the background water at the measurement time t.

When tritium volumetric activity in the background water is low in comparison to the tritium volumetric activity in the sample, it is not necessary to correct the result for splitting of tritium volumetric activity in the background water. If $\Delta t < 0.5$ a year, the last part of the equation may be neglected. The error is denoted by a \pm sign, next to numbers, and it refers to a single measurement error.

Results and discussions

The chapter presents the results of the studies on the volume activity of ³H in groundwater conducted in March and October 2009. The results are compared with the results of studies performed a year earlier i.e. prior to upgrading Maišiagala repository. In March and October 2009 borehole 4 was dry, which prevented a direct comparison of the volumetric activity of tritium in the borehole water to the values measured earlier.

No groundwater was found in the boreholes 2, 3 and 4 (Fig. 5). In March the highest volumetric activity of tritium of the value of 6.9 ± 3.9 Bq·l⁻¹ was found in borehole 5, in October the highest activity was determined as 3.9 ± 1.4 Bq·l⁻¹ in borehole 8. The lowest ³H volumetric activity was found in samples taken from borehole 7 in March and October, and it amounted to 2.4 ± 1.1 Bq·l⁻¹ and 1.5 Bq·l⁻¹, respectively.

The results, presented in Fig. 5 show that in March tritium volumetric activity in the borehole 4.1 reached 154.9 ± 4.5 Bq·l⁻¹, which is 17 times higher than the value measured in the same borehole in October. ³H volumetric activity measured in the borehole 4.2 in March and October decreased over 11 times, i.e. it dropped from 58.6 \pm 3.2 Bq·l⁻¹ down to 5.0 \pm 2.3 Bq·l⁻¹. Such a striking difference could be explained by a significant decrease of the tritium leakage from the repository into the groundwater.

Based on the research carried out previously at Radiation Safety Centre at the Institute of Physics (in Vilnius) and the reports by the Radioactive Waste Disposal Agency (in Vilnius) it can be concluded that tritium has been spreading from the heap. The highest amount of tritium was recorded in borehole 4, therefore this borehole shall be studied more extensively. Figure 6 presents the changes of ³H volumetric activity in borehole 4 between the years 1995 and 2009.

Tritium volumetric activity in borehole 4 varied significantly (Fig. 6). From 1995 till 1998 ³H volumetric activity in groundwater increased over 10 times,



Fig. 5. ³H volumetric activity in boreholes 1–8.



Fig. 6. The highest value of two ³H volumetric activity measurements (performed the same year) for borehole 4 from 1995 till 2009.

i.e. from 2210 Bq·l⁻¹ up to 23 900 Bq·l⁻¹. In 2000 a considerable decrease was observed. The greatest ³H volumetric activity of 29 800 \pm 1500 Bq·l⁻¹ was noted in 2005. The volumetric activity of tritium in the borehole showed a steady increase and tritium presence in the groundwater proved the dispersal of tritium through the reinforced concrete cover to the environment did take place. Hence, the concrete cover intended to protect the environment and inhabitants from the radioactive pollution, proved not tight enough and thus the necessary of upgrading.

According to the data presented in Fig. 6 a gradual decrease of ³H volumetric activity in groundwater after the repository upgrading was observed. In 2007 and 2008 tritium volumetric activity, in comparison to the values measured in 2006, was found to be reduced by a factor of 1.7 and 4.5, respectively, and dropped from 29 500 Bq·l⁻¹ down to 6600 Bq·l⁻¹. This result indicates that the re-equipped barriers are efficient in blocking the tritium transfer from the repository to the environment.

Since in March and October 2009 the borehole 4 was dry (Fig. 5), it was impossible to compare directly tritium volumetric activity in the water from this borehole with the values measured earlier. Nevertheless, the assumption that the installed barriers effectively prevented atmos-



Fig. 7. Groundwater level change in borehole 4 from 2005 till 2009, measured in October.



Fig. 8. Groundwater level change in borehole 4.1 from 2005 till 2009, measured in October.



Fig. 9. Groundwater level change in borehole 4.2 from 2005 till 2009, measured in October.

pheric precipitation from infiltrating into the repository and under the ground, hence they stopped additional amounts of mobile nuclides flow into the surrounding ground, proved sustainable. On the other hand, a distinct drop in groundwater level (Figs. 7–9) could also contribute to the reduction of tritium volumetric activity. The groundwater level in 2009 fell significantly (by 0.5 m on average) in comparison to the years 2005–2008.

Figure 10 shows that by the time the repository was modernized the greatest values of ³H volumetric activity for the two measurements performed in 2005 in boreholes 4.1 and 4.2 were determined as 650 ± 40 Bq·l⁻¹ and 1570 ± 160 Bq·l⁻¹, respectively.

Following the repository upgrade in 2008, an evident increase of tritium volumetric activity in borehole 4.1 was observed, when compared to the data from 2007. In this borehole ³H volumetric activity increased 13 times, while quite to the contrary, the results obtained in 2008 for borehole 4.2 showed the highest levels of the measured ³H volumetric activity to be reduced from 153 Bq·l⁻¹ down to 118 Bq·l⁻¹, in comparison to the results dated 2006.

Figure 11 presents the comparison of values for ³H volumetric activity for boreholes 4.1 and 4.2 in March and October from 2006 to 2009.

Following the data shown in Fig. 11 with reference to the tests performed in March 2009, it can be noted that



Fig. 10. The highest value of two ³H volumetric activity measurements (performed the same year) for boreholes 4.1 and 4.2 over the years 2005–2008.



Fig. 11. The highest value of two ³H volumetric activity measurements (performed the same year) for boreholes 4.1 and 4.2 measured in March and October over the years 2006–2009.

the highest value of ³H volumetric activity, in comparison with the March values measured between 2006–2008, was found in borehole 4.1. The lowest ³H volumetric activity in the groundwater from borehole 4.1 was measured in March 2007 and it was equal to 2.5 Bq·l⁻¹, hence it was 62 times lower than in March 2009.

Figure 11 shows the increase of ³H volumetric activity in borehole 4.2 in March 2007 in comparison to the value measured in March 2006, however, in 2008 and 2009 a steady decrease was already noted. Figure 11 shows for borehole 4.2 a gradual increase in ³H volumetric activity, which over the years 2006–2008 went up from 1.5 up to 5 Bq·l⁻¹.

All the above observations are linked with a longterm transfer (or transportation) of tritium, thus, if efficient performance of the new barriers is to be continued, as the time progresses tritium volumetric activity in thick pipes of both boreholes 4.1 and 4.2 should reach their maximum values and then gradually stabilize.

From the experience with RADON type Russian repositories, we can see that with time many operational and natural factors heavily impacted the installed engineering barriers and damaged the repository tightness [13].

The infiltration of tritium (³H) to groundwaters was reported for Kiev and Kharkov, Ukrainian RADON type radioactive waste repositories. Inadequacy of technical standards for engineering buildings applied there, and departure from requirements regarding physical and radioactive waste storage, were identified as the main reasons for radioactive pollution. Due to improper design of the repository building, operating since 1950, water in the repositories was affected by condensation and infiltration [7].

Conclusions

The major problem of radioactive waste repositories is attributed to tritium dispersal by groundwater. The highest ³H volumetric activity in groundwater was measured for borehole 4. In 2005 it reached 29 800 \pm 1500 Bq·l⁻¹.

Following the repository upgrade a gradual reduction of tritium volumetric activity in groundwater was observed for borehole 4. This evident result made us assume the new barriers installed recently to be effective enough to block tritium transfer from the repository heap to the environment.

While analysing samples taken in March and October 2009 it was established that the problematic borehole 4 was dry. It was associated with the effectiveness of the installed engineering barriers, which stopped the infiltration of precipitation water to the repository under the ground.

The lowest ³H volumetric activity in the groundwater from borehole 4.1 was measured in March 2007 and it was equal to 2.5 Bq·l⁻¹, i.e. it reached the value 62 times lower than in March 2009. ³H volumetric activity in borehole 4.2 in March 2007 is increasing, compared to the value in March 2006, though in 2008 and 2009 a decreasing trend was observed. Such effects are linked with a long-term transfer (or transportation) of tritium, thus, if new barriers continue to operate efficiently, tritium volumetric activity in thick pipes of both boreholes 4.1 and 4.2 should reach their maximum values in due time and then gradually stabilize.

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