

Assessment of radionuclide migration and radiological human exposure at the closed near-surface radioactive waste repository

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Abstract. The near-surface “RADON” type radioactive waste repository, installed in 1963 and designed to store radioactive waste formed in industry, medicine and scientific investigations, was closed in 1989 because it did not meet the requirements imposed on the radioactive waste disposal. A comparatively small amount of radioactive waste is stored in this repository, but the inventory comprises various kinds of waste: short-lived low-level radioactive waste, short-lived low and intermediate radioactive waste, long-lived intermediate and high-level radioactive waste. The possible site-specific radionuclide migration through the groundwater pathway as well as the human exposure are considered by the computer program RESRAD-OFFSITE in this paper. The analysis of the obtained data shows that out of all stored radionuclides only H-3, C-14 and Cl-36 exceeding the dose constraint of 0.2 mSv can be considered as dangerous. The monitoring carried out in the repository environment has shown the contamination of groundwater with radioactive tritium and a significant reduction of contamination after construction of additional protective engineering barriers. For the assessment of the contribution of separate site-specific parameters of the model taking into account uncertainties of the model and parameters to the annual effective dose, the computer code RESRAD-OFFSITE provides the possibility of applying the regression analysis. It has been determined that the aquifer lateral dispersion as well as the hydraulic gradient, the radionuclide activity concentration in the repository together with the rate of penetration into the environment and the precipitation amount have the largest influence on the assessment accuracy of annual effective doses.

Key words: near-surface repository • engineering barriers • radionuclide migration • groundwater pathway • radiation protection

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Introduction

The storage of radioactive waste formed in industry, medicine and scientific investigations in a special radioactive waste storage facility was started in 1963 [5]. Waste was transported not only from Lithuania but also from neighboring Kaliningrad and Gardin regions till 1988 [13]. The ferroconcrete vault partly filled with waste was sealed up and sanded in 1989, and the disposal of newly-generated radioactive waste was directed at the repositories of the Ignalina Nuclear Power Plant.

Though a comparatively small amount of waste (only about 120 m³) is stored, but it has not been sorted out: the long-lived high-level radioactive waste is placed together with the short-lived low-level radioactive waste (Table 1).

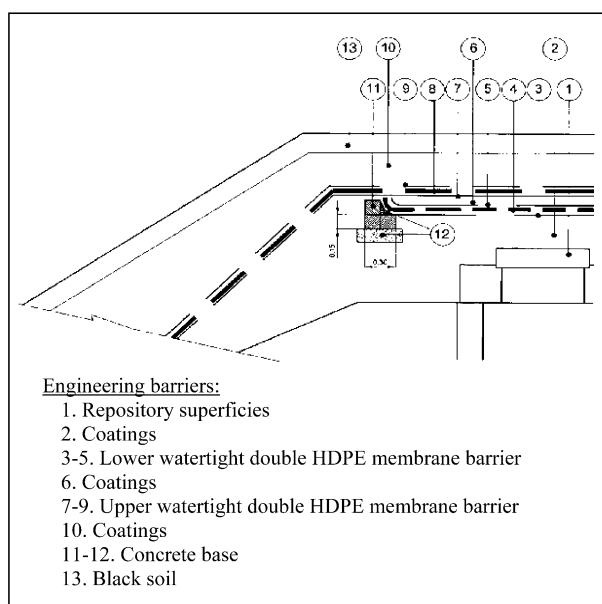
In 2002, this storage facility was released to the Radioactive Waste Management Agency (RATA). The environmental monitoring is carried out by the Institute

Table 1. Near-surface Maisiagala repository estimated inventory [13] and conformity to threshold of the in detail analyzed radionuclides at closing time (01.01.1989)

Radionuclide	Half-life (y)	Total activity	
		(Bq)	(Bq/g)
H-3	12.34	2.77E+14	1.85E+06
C-14	5.7E+03	1.77E+11	1.18E+03
Cl-36	3.0E+05	1.20E+09	8.00E+00
Co-60	5.27	7.33E+12	4.90E+04
Se-75 (container)	0.33	2.44E+10	1.63E+02
Sr-90	28.81	6.42E+11	4.29E+03
Cs-137	30.06	5.57E+13	2.39E+05
Eu-152	13.53	5.38E+10	3.60E+02
Tm-170 (container)	0.36	4.39E+12	2.93E+04
Ir-192 (container)	0.20	6.57E+11	4.39E+03
Ra-226	1.6E+03	1.11E+11	7.42E+02
Pu-239	2.4E+04	3.16E+11	2.11E+03
U-238	4.5E+09	4.31E+07	2.88E+02
U-234	2.5E+05	1.45E+03	9.69E-06
Ni-63	1.0E+02	4.14E+10	2.77E+02
Bi-207	31.78	6.72E+05	4.49E+00
Kr-85	10.76	2.19E+09	–
Ba-133	10.55	4.10E+06	2.74E+01

of Physics, groundwater samples from 10 wells and boreholes are systematically collected and their radiological assessment is performed [19].

Under the EU PHARE project, the French companies THALES and ANDRA having much experience in radioactive waste management together with the Lithuanian Energy Institute and the Institute of Physics determined the technical state of the repository, analyzed the monitoring results and evaluated its safety [4]. One of the most significant results of the project is construction of additional protective engineering barriers. According to the project [12], the system of two layers of very low permeability high-density polyethylene (HDPE) membranes (Fig. 1), the durability of which is not shorter than 30 years, was constructed on the ferroconcrete vault of the radioactive waste. The main

**Fig. 1.** Additional protective engineering barriers of the closed radioactive waste repository: the system of two watertight double HDPE membrane barrier.

conclusions of the performed safety evaluation were the following [20]: though the radionuclide migration from the vault is not dangerous to the environment and people, but in the long-term perspective the safety requirements are not satisfied as there is no assurance that the stored sealed sources will not be taken out and cause human exposure.

The total activity concentration of long-lived alpha radionuclides (Pu and Ra isotopes) reaches 3 kBq/g and exceeds the set limit [6, 7]. However, the facility can be regarded as a secure repository of radioactive waste. On this basis, in 2006 the radioactive waste storage facility was designated a repository and the State Nuclear Power Safety Inspectorate (VATESI) has given a license entitling RATA the right to operate this radioactive waste repository.

The aim of this work is to assess the changes of radionuclide migration from the radioactive waste repository through the water pathway and human exposure due to the reconstruction of protective engineering barriers by applying the deterministic and stochastic approximations taking into account the radiation protection criterion established in the Republic of Lithuania, the annual effective dose constraint for population in operating and decommissioning the nuclear power objects is 0.2 mSv [3]. In the cases when there is no population living near such objects, for the assessment of the impact a hypothetical critical group which potentially could live in that territory should be formed [17].

Assessment of the radiological protection of the closed radioactive waste repository by applying a conservative drinking water scenario

Due to high mobility of hydrogen atoms, tritium is the first radionuclide released from the radioactive waste repository. This is also confirmed by the monitoring data of the environment of this radioactive waste repository. The largest tritium migration from the repository to the

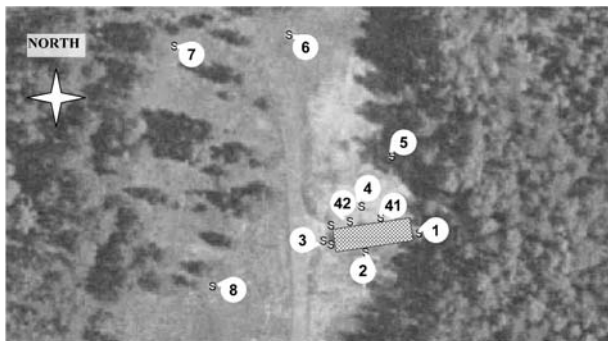


Fig. 2. Near-surface radioactive waste repository and environmental monitoring stations: borehole no. 4 water used for annual effective dose assessment in the case of very conservative scenario when water would be used for drinking and farming.

groundwater, when the activity concentration of tritium exceeded 10 kBq/l, was determined in the region of boreholes 4, 41 and 42 (Fig. 2).

The data of monitoring carried out by the Institute of Physics [19] show that after construction of new protective engineering barriers of the repository the activity concentration of tritium in the environment of the repository and in the maximal contamination borehole 4 decreased significantly in 2008 (Fig. 3). Referring to the presented experimental data the annual effective exposure dose of adult and children was evaluated by applying the most conservative scenario of groundwater from borehole 4, when water is used for drinking and farming, as well as the approach described in [2]. In the

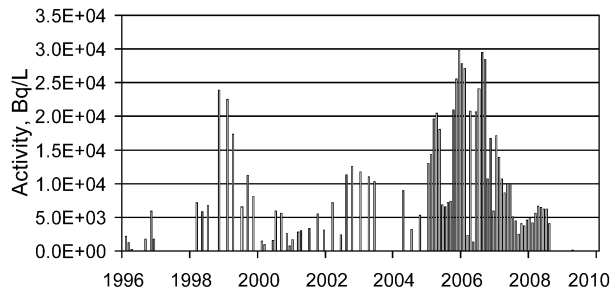


Fig. 3. Tritium activity concentration time dependent fluctuations in borehole 4 water.

exposure dose assessment special attention was paid to the parameters of annual site-specific main food products and drinking-water consumption (Fig. 4) by applying the statistical methods [1, 11].

The assumption was made as to the equilibrium of the tritium activity concentration in all environmental components, i.e. the tritium activity concentration in water of plants (C_{pw}^{HTO}) and in organic compounds (C_{pw}^{OBT}), in water of live stock (C_{an}^{HTO}) and in organic compounds (C_{an}^{OBT}) agrees with the average of the tritium activity concentration in water of borehole 4 (C_w , Bq/l). In this case:

$$(1) \quad C_{pw}^{HTO} = C_w \times WC_p$$

$$(2) \quad C_{pw}^{OBT} = C_w \times (1 - WC_p) WEQ_p$$

where WC_p is the water fraction; WEQ_p is the water equivalent factor, i.e. the total hydrogen amount present

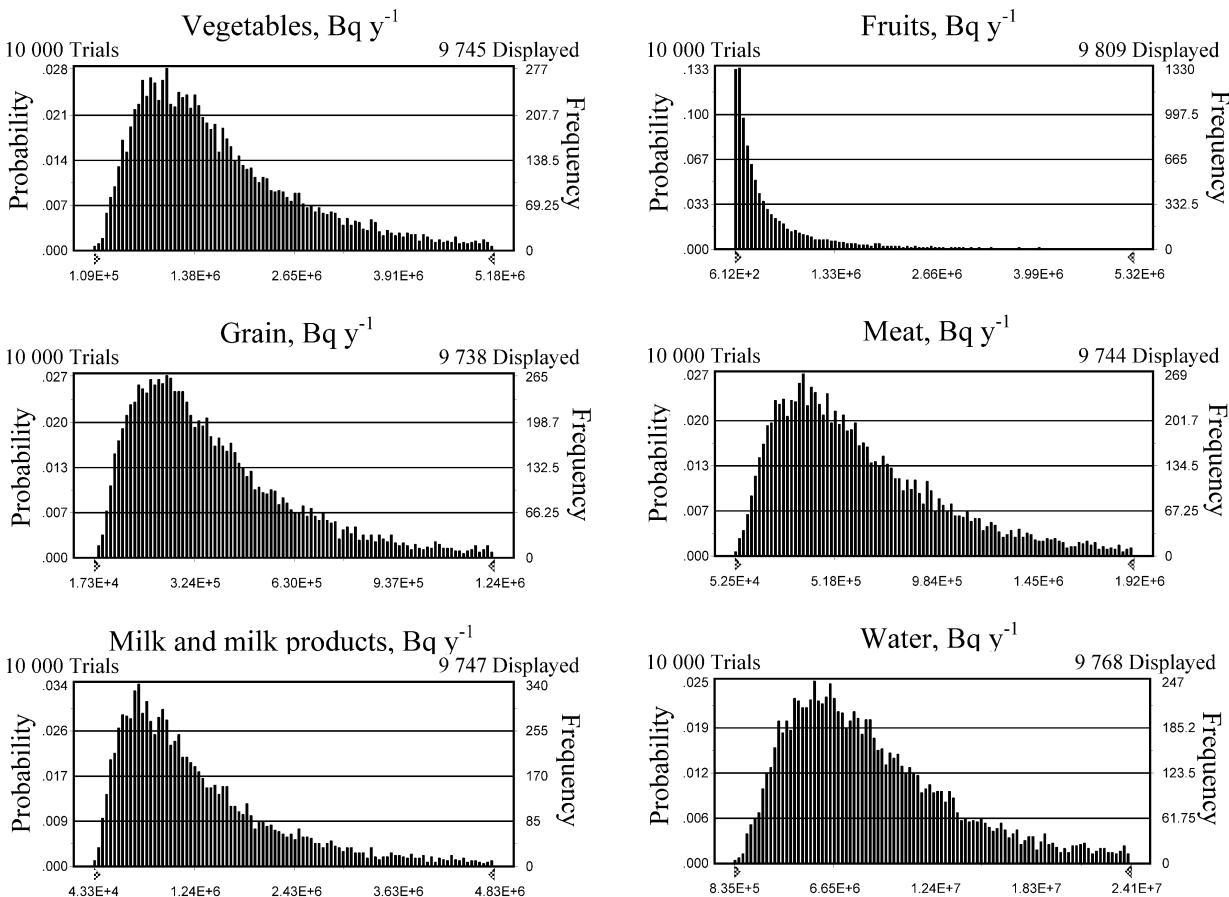


Fig. 4. Site-specific distributions of annual tritium contaminated food products and drinking-water consumption.

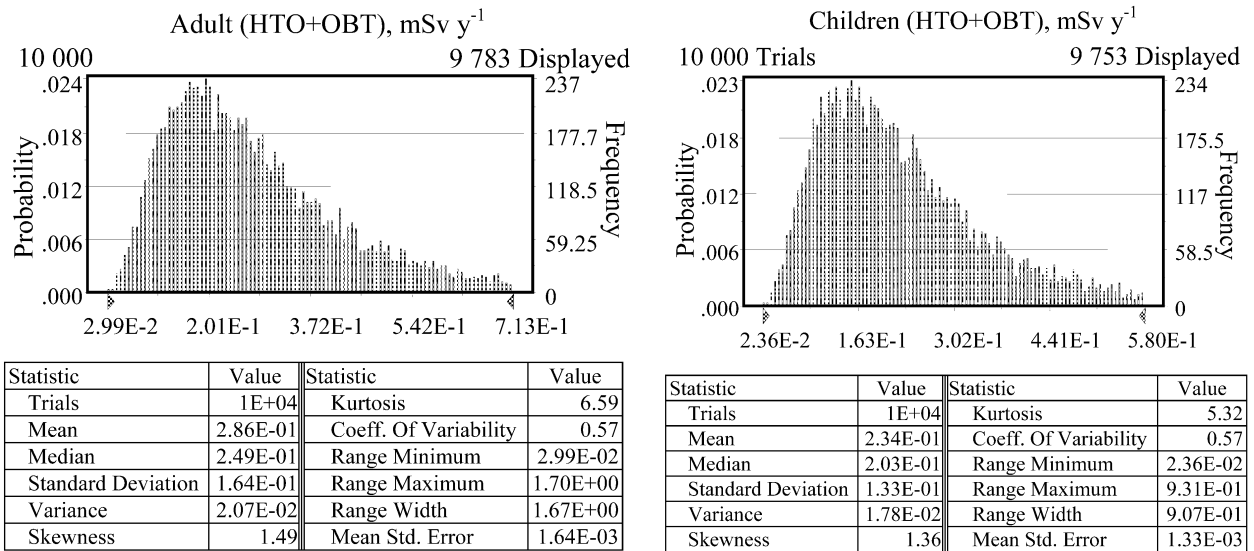


Fig. 5. Distributions of the annual effective internal exposure dose of adults (> 17 years) and children (7–12 years) due to tritium in food products and drinking water together with statistical parameters in the case of a very conservative (till the reconstruction of the repository engineering barriers) scenario when borehole 4 water would be used for drinking and farming.

Table 2. Annual average effective dose of internal exposure of adults and children due to inorganic and organic tritium fractions in food products in the case of a very conservative scenario (till the reconstruction of repository engineering barriers) when borehole 4 water used for drinking and forming

Tritium fractions	$e(g)_{int}$ (Sv·Bq ⁻¹)	Effective dose (mSv·year ⁻¹)			
		Average	Standard deviation	Min	Max
Adults (> 17 years)					
H-3 (HTO)	1.80E-11	2.43E-01	1.41E-01	2.41E-02	1.31E+00
H-3 (OBT)	4.20E-11	4.31E-02	2.65E-02	3.60E-03	3.96E-01
Total		2.86E-01	1.64E-01	2.99E-02	1.70E+00
Children (7–12 years)					
H-3 (HTO)	2.30E-11	1.97E-01	1.13E+01	1.95E-02	1.06E+00
H-3 (OBT)	5.70E-11	3.71E-02	2.25E-02	3.10E-03	3.41E-01
Total		2.34E-01	1.33E-01	2.26E-02	1.40E+00

in food product water and in organic compounds (proteins – $P_{protein}$, fat – P_{fat} and carbohydrates – $P_{carbohydrate}$) is estimated as follows:

$$(3) WEQ_p = 8.94 [0.07 \cdot P_{protein} + 0.12 \cdot P_{fat} + 0.062 \cdot P_{carbohydrate}]$$

where 0.07 is the hydrogen fraction in proteins; 0.12 is the hydrogen fraction in fat; 0.062 is the hydrogen fraction in carbohydrates; 8.94 is the ratio of water and hydrogen molecular weights.

The internal effective dose due to food product contamination with tritium $E_{H-3,p}$, Sv, after consuming p type food product was assessed using the following dependencies:

$$(4) E_{H-3,p} = E_{HTO5,p} + E_{OBT,p}$$

$$(5) E_{HTO,p} = C_{pw}^{HTO} \cdot Q_{wp} \cdot e(g)_{HTO,ing} \cdot T_{ing}$$

$$(6) E_{OBT,p} = C_{pw}^{OBT} \cdot Q_{wp} \cdot e(g)_{OBT,ing} \cdot T_{ing}$$

Here T_{ing} is the internal human exposure consuming food products contaminated with tritium with the assessment period $T_{ing} = 1$ year; Q_{wp} is the amount of

p type food product (kg) consumed by a man per year; $e(g)_{HTO,ing}$, $e(g)_{OBT,ing}$ are factors of tritium HTO and OBT form effective internal exposure doses due to food product and drinking-water contamination with tritium, Sv/Bq.

Distributions of annual effective internal exposure doses of adult (> 17 y) and 7–12 year old children due to food product and drinking-water contamination with tritium are presented in Fig. 5 and the exposure part falling on the inorganic and organic tritium fractions in food products is indicated in Table 2. The change of exposure of adult and children during the period of 2005–2008 in case of a hypothetical conservative water consumption scenario from borehole 4 is shown in Fig. 6.

It is evident that the reconstruction of protective engineering barriers of the closed radioactive waste repository in summer 2006 has significantly reduced the exposure of adult and children, which was assessed by applying a very conservative scenario of water consumption from the largest pollution well. Based on the monitoring data of 2007–2008 and the exposure assessment, it can be stated that at present the effective exposure dose is significantly lower compared with the dose constraint established in the Republic of Lithuania [3].

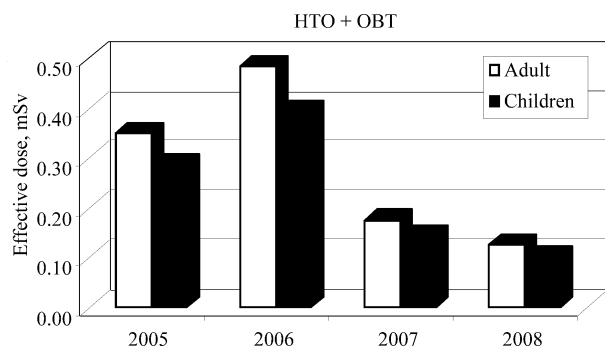


Fig. 6. Time-dependent adult (> 17 years) and children (7–12 years) annual effective internal exposure doses fluctuation due to HTO and OBT fractions of tritium in food products and drinking water (2005–2008 data).

Assessment of radiological protection of closed radioactive waste repository applying the radionuclide migration scenarios

When assessing the radionuclide migration and forecasting the human exposure in the environment of repositories the scenarios can be created and chosen in some different ways. For the assessment of human exposure out of the scenarios presented in the IAEA document [8, 9], the scenario of the radionuclide leaching from the repository after the closure of facility, which is also called the radionuclide migration through the water pathway, is considered. The water penetrates through the engineering barriers of the repository due to precipitation infiltration. From the repository, radionuclides with water flow are transferred to the geological structure, which comprises the unsaturated zone and the aquifer. The assumption is made that geological and hydro-geological conditions are stable during the whole analyzed period of the repository development. For the assessment of the radionuclide migration from the near-surface storage facility and the annual effective dose of human exposure, the following scenarios were chosen.

- Scenario A – naturally degrading engineering barriers. Due to the natural aging of engineering barriers and their degradation the processes of radionuclide penetration into the environment from the repository through the water pathway were assessed as indicated in [15].
- Scenario B – reconstruction of protective engineering barriers. According to the project [12], additional protective barriers – the system of soil and two very low permeability membranes – were constructed over the radioactive waste vault in 2006 as shown in Fig. 1. With the constant control and renewal of the membrane system an assumption is made about the unchanging radionuclide penetration into the environment through the engineering barriers till the end of the active institutional superintendence period.

Model and computer program of radionuclide migration and human exposure assessment

The model of the computer program RESRAD-OFFSITE of the radionuclide migration via the water path-

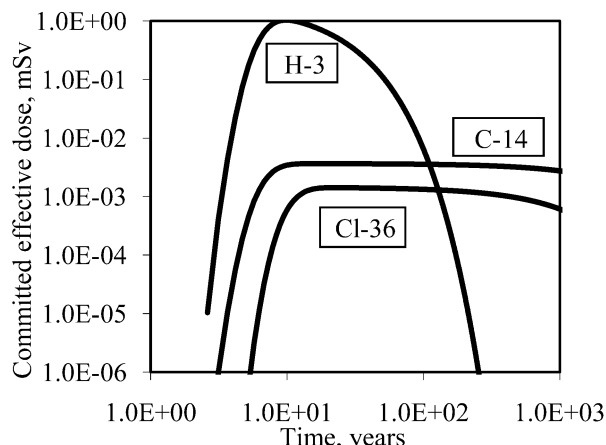


Fig. 7. RESRAD-OFFSITE code simulated variation of the annual effective dose due to radionuclide migration through the water pathway: scenario A when the water source (water used for drinking and farming) is at a distance of 500 m from the repository.

way [18] assesses the radionuclide penetration into the unsaturated zone by infiltration through the repository engineering and natural barriers as well as their further transfer to the aquifer through the water embedded in soil pores. Radionuclides migrate in the aquifer through the groundwater in the prevailing direction due to advection-dispersion processes. Eventually, radionuclides reach the biosphere, where having penetrating into the drinking water well or bore become a potential source of human exposure. In the RESRAD-OFFSITE code simulations of 109 site-specific parameter collections, in most cases and their distributions, have been used.

In the case when only single-measure (longitudinal) hydrodynamic dispersion in the unsaturated zone and the aquifer is assessed, not taking into account a possible decrease of the radionuclide activity concentration in water due to lateral and vertical dispersion, the human exposure is assessed in a conservative way. The computer program RESRAD-OFFSITE makes it possible to assess the groundwater migration in the three-dimensional space taking into account longitudinal, lateral and vertical dispersion.

In this work by assessing the annual effective dose variation, when the drinking-water source is at the 100 m, 500 m and 2.5 km distance from the closed radioactive waste storage facility taking into account site-specific geological conditions [10, 14, 15], it was assumed that parameters of longitudinal, lateral and vertical dispersion are the following: $\sigma_{D,x} = 1.0$ m, $\sigma_{D,y} = 0.15$ m, $\sigma_{D,z} = 0.1$ m. The results of assessment of radionuclide migration through the water pathway by the RESRAD-OFFSITE computer code have shown that out of all stored radionuclides at present time H-3, C-14 and Cl-36 are regarded as dangerous from the radiation protection point of view. The variation of the annual effective dose due to migration of these radionuclides through the water pathway is presented in Fig. 7.

It should be noted that in cases of considered scenarios of radionuclide dispersion through the water pathway, as shown in Fig. 8, the pathway of the highest exposure in case of H-3 and C-14 is drinking water, while in the case of Cl-36 also the pathway of local origin food product contamination.

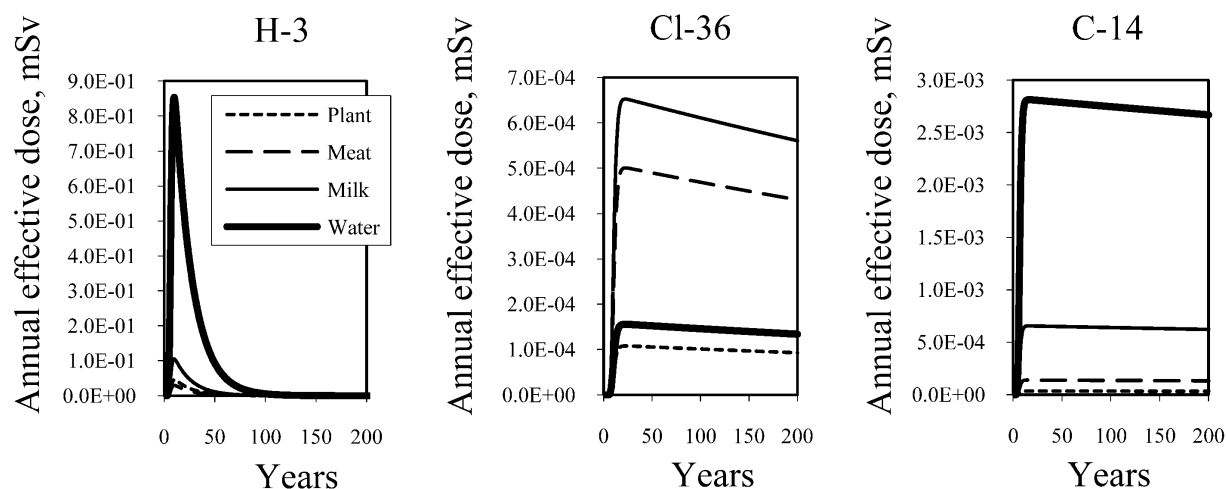


Fig. 8. The contribution of separate exposure pathways on the annual effective doses in the case of H-3, C-14 and Cl-36.

Uncertainties and sensitivity analysis

Uncertainties having influence on the assessment of exposure doses and risk factors can be of these main categories: uncertainties of scenarios, models and parameters [16, 21].

- Scenario uncertainties are uncertainties related to inaccuracies of the intended development of the radioactive waste storage system. In this work a very conservative scenario of drinking water (also used for farming) from the largest contamination well and two scenarios of possible development of the repository (scenario A – naturally degrading repository barriers and scenario B – new protective engineering barriers of the repository) were analyzed.
- Model uncertainties are related to accepted simplifications of real processes and properties during formation of conceptual models and the differences of between the software used in modeling and mathematical models.
- Uncertainties of parameters are related to the consistency of used parameter values with the investigated environment. According to the sensitivity analysis results, we can determine the more precise assessment of which model parameters would allow a significant reduction of the model result uncertainty and the further correction of which parameters makes no sense due to their insignificant influence on the results. A typical result of the sensitivity analysis is the list of

main model parameters by evaluating the importance of parameters by quantitative measures.

In order to assess the contribution of separate parameters to the exposure dose the computer program RESRAD-OFFSITE provides the possibility of investigating uncertainties and sensitivity of the model results using the regressive analysis. The contribution of more than 100 model parameters to the exposure dose was evaluated. As several statistical methods of the sensitivity analysis are known, it is useful not only to choose a proper method but also to compare the obtained results with those obtained by other methods. In this case a partial grade coefficient of correlation, the standardized grade regression coefficient, a partial correlation coefficient and the standardized regression coefficient were determined. The main determined parameters along with the values of stochastic variables in the case of H-3 are presented in Table 3.

Examples of distributions of main site-specific parameters in the case of tritium dispersion are shown in Fig. 9.

These main regularities of parameter uncertainties describe dispersion of C-14 and Cl-36 and their contribution to the exposure. The presented values of stochastic variables show that the main parameters describing the radionuclide dispersion causing the exposure dose (in the cases of radionuclide leaching scenarios A and B) are related to the aquifer lateral dispersion, the hydraulic gradient and hydraulic permeability, the precipi-

Table 3. Values of main parameters and route probability variables determining the exposure dose in the case of H-3 (distance from well to the repository – 500 m)

Description of probabilistic variable	PRCC ^a	SRRC ^b	PCC ^c	SRC ^d
Horizontal lateral dispersivity of saturated zone to well	-0.97	-0.79	-0.61	-0.48
Leach rate of H-3	0.91	0.41	0.57	0.42
Hydraulic gradient of saturated zone to well	-0.73	-0.20	-0.32	-0.20
Concentration of H-3	0.72	0.19	0.35	0.22
Hydraulic conductivity of saturated zone	-0.59	-0.13	-0.34	-0.21
Thickness of contaminated zone	0.56	0.12	0.21	0.13
Precipitation amount	0.44	0.09	0.14	0.09

^a PRCC – partial rank correlation coefficient.

^b SRRC – standardized rank regression coefficient.

^c PCC – partial correlation coefficient.

^d SRC – standardized regression coefficient.

tation amount, the radionuclide activity concentration and the constant of penetration into the environment. The presented uncertainty data determine the limits of the annual effective dose variation.

To assess the human exposure variation due to the construction of protective engineering barriers in

the cases of scenarios A and B the migration/dispersion of H-3, C-14 and Cl-36 currently regarded as dangerous in the far zone and population consuming water from wells at a distance of 500 m and 2.5 km from the repository (also using this water for agricultural needs) and local food products has been considered. The variation

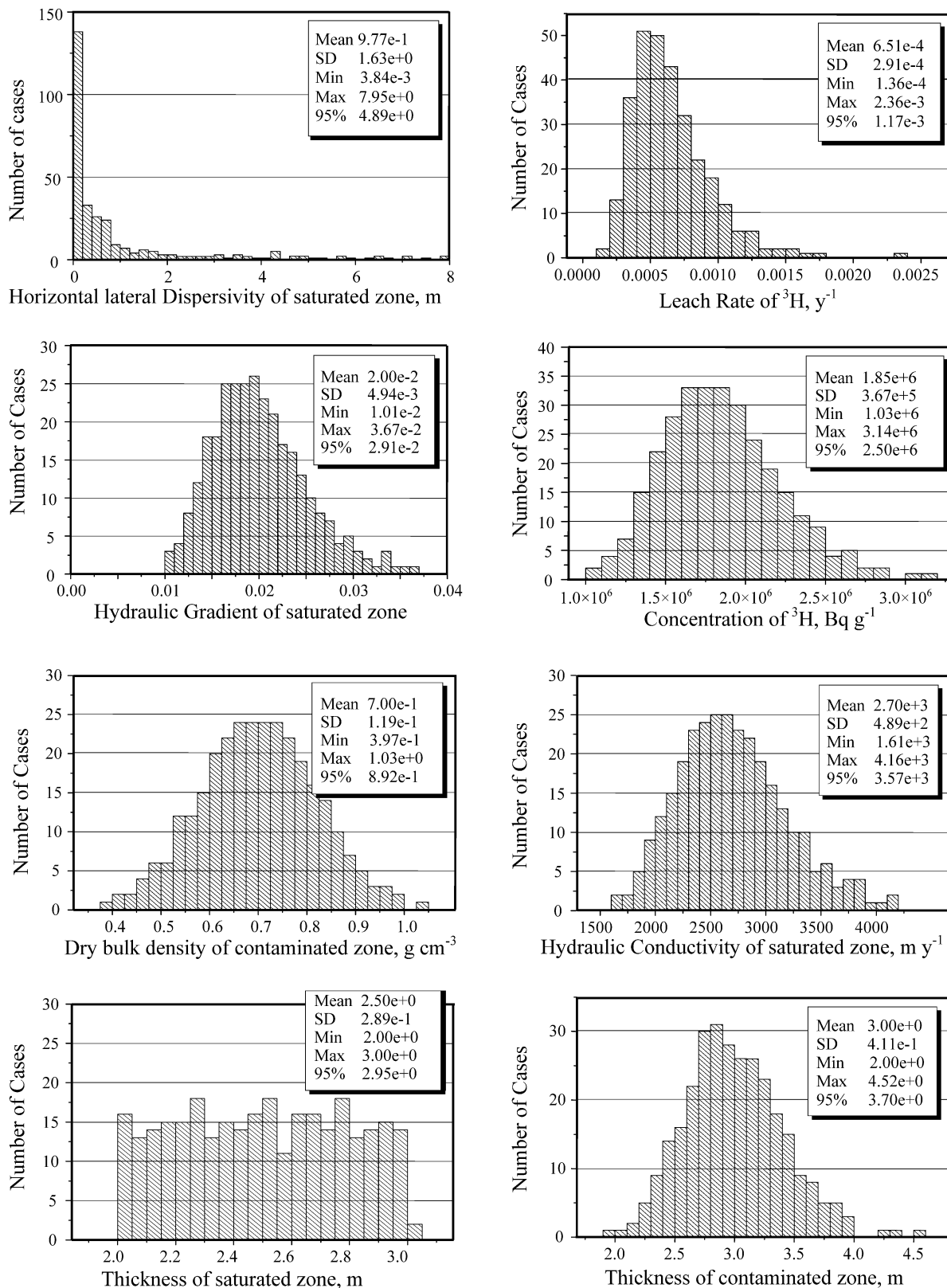


Fig. 9. RESRAD-OFFSITE code simulated distributions of the main site-specific parameters in the case of tritium migration through water pathway from the closed near-surface radioactive waste repository.

Table 4. Variation of main annual committed effective dose assessments by applying the stochastic approximation of radionuclide pathway in the environment and leaching through the repository barriers (radionuclide leaching scenario A and B) when the drinking-water wells are at a distance of 500 m

Radionuclide	Scenario A annual committed effective dose (mSv)				Scenario B annual committed effective dose (mSv)			
	Min	Max	Average	Standard deviation	Min	Max	Average	Standard deviation
50 years after the repository closure								
H-3	5.34E-03	2.22E+00	2.02E-01	2.23E-01	3.77E-03	9.02E-01	8.89E-02	9.51E-02
C-14	3.22E-04	5.03E-02	6.38E-03	6.99E-03	1.29E-04	2.02E-02	2.80E-03	2.80E-03
Cl-36	1.21E-07	2.54E-02	2.62E-03	2.80E-03	4.57E-05	1.04E-02	1.08E-03	1.15E-03
Sum	7.57E-03	2.26E+00	2.11E-01	2.30E-01	4.67E-03	9.19E-01	9.28E-02	9.77E-02
100 years after the repository closure								
H-3	5.56E-04	1.16E-01	1.22E-02	1.27E-02	2.26E-04	5.08E-02	5.17E-03	5.45E-03
C-14	3.19E-04	4.87E-02	6.85E-03	6.81E-03	1.28E-04	1.98E-02	2.76E-03	2.75E-03
Cl-36	1.08E-04	2.20E-02	2.44E-03	2.53E-03	4.50E-05	9.67E-03	1.04E-03	1.09E-03
Sum	1.67E-03	1.94E-01	2.70E-02	2.55E-02	5.68E-04	6.80E-02	8.96E-03	8.29E-03

of the effective doses and the results of the uncertainty analysis are presented in Table 4.

The obtained data show that the newly constructed protective engineering barriers restricting the radionuclide migration through groundwater, and further continuing monitoring of the repository environment, ensure the radiation protection of population meeting the requirements of hygiene standards valid in the Republic of Lithuania.

Conclusions

The analysis of the potential radionuclide migration from the closed near-surface radioactive waste repository via the water pathway has shown that out of all stored radionuclides H-3, C-14 and Cl-36 can be considered as dangerous. In the case of H-3 and C-14 the highest exposure is due to contamination of drinking water, while in the case of Cl-36 due to local origin food product contamination as well.

The reconstruction of engineering barriers of the closed radioactive waste repository in 2006 significantly reduced the exposure of adults and children, which was assessed by applying a very conservative scenario when water from the largest contamination well was used for drinking and farming. Based on the data of monitoring and the exposure assessment, it can be stated that at present the annual effective dose is significantly lower as compared with the dose constraint of 0.2 mSv in the environment of nuclear power objects as determined in the Republic of Lithuania.

By assessing the radionuclide migration in the environment of the repository and forecasting the annual effective dose as well as taking into account the uncertainties of the model and parameters having influence on the dose, it has been determined that at the distance of 500 m during the period of forthcoming 50 years in the case of scenario A (naturally degrading engineering barriers) the annual effective dose would exceed the determined 0.2 mSv dose constraint (in the range from 0.008 mSv to 2.26 mSv), and in case of scenario B (new additional engineering barriers) the average annual effective dose would be 0.1 mSv (in the range from

0.005 mSv to 0.92 mSv). In this case the upper exposure limit would meet the requirements of the main radiation protection standards.

The correlation and regression analysis of exposure parameters and routes of the radionuclide migration through the water pathway (in the case of radionuclide leaching scenarios A and B) has shown that the lateral dispersion of water migration, the aquifer parameters, the precipitation amount, the radionuclide activity concentration in the repository and the rate of their penetration into the environment have the largest impact on the accuracy of the effective dose assessment.

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