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MODELLING ^{226}Ra , ^{222}Rn , AND ^{210}Pb MIGRATION IN A PROPOSED SURFACE REPOSITORY OF VERY LOW-LEVEL LONG-LIVED RADIOACTIVE WASTE

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ABSTRACT

A generic concept for disposal of very low-level long-lived radioactive waste is currently being evaluated for radium bearing wastes that originated from a historical radium extraction plant at Olen, Belgium. A total volume of approximately 217 000 m³ of waste with an average radium content of 7 Bq/g has to be disposed of. Upon request by the Belgian National Agency for Management of Radioactive Waste and Nuclear Fuels, NIRAS/ONDRAF, a generic disposal concept was evaluated for the purpose of identifying the minimum disposal concept which guarantees long term safety. Such an analysis would provide useful input to the final design of the disposal concept, as the contribution of the different engineered barriers to the overall safety will have been assessed. The analysis focussed on the migration of ^{226}Ra , ^{222}Rn , and ^{210}Pb to groundwater owing to infiltration of rainwater and the diffusive radon transport via the gas phase to the atmosphere. The generic design considered a waste dump with the contaminated material completely enclosed by a clay barrier. To protect the low-permeability clay from degradation by water and wind erosion, frost and desiccation, burrowing animals, and plant roots, among others, a multi-layer cap was designed to meet these requirements. In the performance assessment calculations, various cases were considered in which one component of the disposal concept was changed at the time. Cases considered included absence of clay layers, effects of hot spots, lower adsorption capacity of various materials, and the effect of separating the radium contaminated material from nearly uncontaminated material. Unsaturated flow calculations were done first to estimate the steady-state water content profile. Knowledge on the degree of water saturation is of paramount importance for radon transport through the gas phase. Based on the steady-state water content profile, advective dispersive transport calculations were done considering the decay chain reaction of ^{226}Ra . In addition to the radon gas flux to the atmosphere, fluxes of ^{226}Ra , ^{222}Rn , and

^{210}Pb to groundwater were also produced. Groundwater flow and transport calculations yielded radionuclide concentrations in a hypothetical well nearby the planned disposal site, whereas biosphere modelling provided the annual doses to the public considering the groundwater pathway and direct inhalation of radon in case of the atmospheric pathway. On the basis of the calculated radionuclide fluxes and doses the importance of the various model parameters and concept components will be evaluated and discussed.

INTRODUCTION

From the early 1920's to the mid 1970's the industrial activities of Metallurgie Hoboken-Overpelt (MHO) at Olen, now Umicore, were known world wide for the radium and uranium production [1]. During several decades, MHO was one of the most important radium production plants and contributed significantly to the widespread use of radium in medical and industrial applications. These industrial activities resulted in a variety of waste products, including mill tailings, dismantling and decontamination materials, contaminated soil, residues from radium extraction, etc. An impact assessment analysis for the most radioactive radium bearing wastes that are presently stored in a heavily engineered facility was reported by [2]. In the present paper we discuss the preliminary impact assessment for a proposed surface disposal facility for very low-level radium bearing waste. The impact assessment will focus on two major exposure pathways, i.e., the groundwater pathway and the atmospheric pathway. Human intrusion is not discussed here. The objective of the paper is to assess the long-term radionuclide behaviour in response to changes in disposal concept, and transport related parameters. For this purpose a series of scoping calculations were carried out, focussing on the radionuclide fluxes to groundwater and atmosphere. Such calculations may help define the minimum disposal concept which guarantees long term safety.

MATERIALS AND METHODS

Site Characteristics

An extensive hydrogeological characterization has been carried out in the vicinity of the proposed disposal site. The uppermost layer (0-8 m) is composed of well sorted sands, known as Kasterlee Sands. Between 3 and 5 m depth the sand layer has a typical green color owing to the presence of a small amount of glauconite. At the eastern boundary of the site the sand layer contains a fairly thin clay layer, known as Kasterlee Clay. At some locations the latter is only present as a series of fairly thin clay lenses. A second sand layer, known as Diest Sands, extends from 8 to approximately 90 m depth and contains about 25-30 weight % glauconite. The groundwater at the site is very close to the soil surface. The hydraulic conductivity varies between approximately 10^{-6} m/s for the upper and lower sands and about 10^{-7} m/s for the clay-rich layer. Further details can be found in [2].

Disposal Characteristics

A preliminary concept for surface disposal of approximately 217 000 m³ of very low-level radioactive waste was recently designed. The waste was mainly composed of contaminated soil, ironhydroxides and calcium sulphate as major residues from cobalt production, and residues from radium extraction [3]. Waste is composed of 135 000 m³ of radium bearing material at an average concentration of 7 Bq/g ²²⁶Ra, whereas approximately 82 000 m³ of material has a negligible radium content. The total radium inventory is estimated at 2×10^{12} Bq.

The waste material will be isolated from the geosphere by means of clay layers that enclose the waste at the bottom and at the top of the disposal dump. A schematic view of the various layers that comprise the repository, including a multi-layer protective cap, is given in Fig.1. Protection of the 1-m-thick clay layer (clay-1 and clay-2) against burrowing animals, drying, freeze-thaw cycles, etc. is secured by means of a 2.8-m-thick layer composed of several materials, including a loamy top soil, sand, and gravel. Total maximum depth of radioactive waste material is 14.5 m. At the bottom of the repository, a 0.5-m-thick clay layer (clay-3) together with a High Density Polyethylene (HDPE) foil separates the waste from the underlying groundwater.

Numerical Models

Calculation of the migration of radium and its daughter nuclides to geosphere and biosphere requires several models. Each of the models used here will be briefly discussed.

Flow model for multi-layer cap

Prior to the modelling of radium leaching to groundwater, the water flux through the waste material has to be known. The magnitude of this flux is determined by the capacity of the multi-layer cap to divert the infiltrating rainwater. The two-dimensional variably saturated flow and transport code HYDRUS-2D [4] was used for this purpose. The 2D flow model included the first six layers of the profile shown in Fig. 1, i.e., from top soil to the clay-2 layer. Appropriate values for

the van Genuchten-Mualem soil hydraulic functions [5] were estimated for each soil layer [6]. The steady-state water redistribution together with the appropriate initial and boundary conditions were imposed to model steady-state water redistribution in the multi-layer cap. A long term average net rainfall of 10^{-8} m/s was used as top boundary.

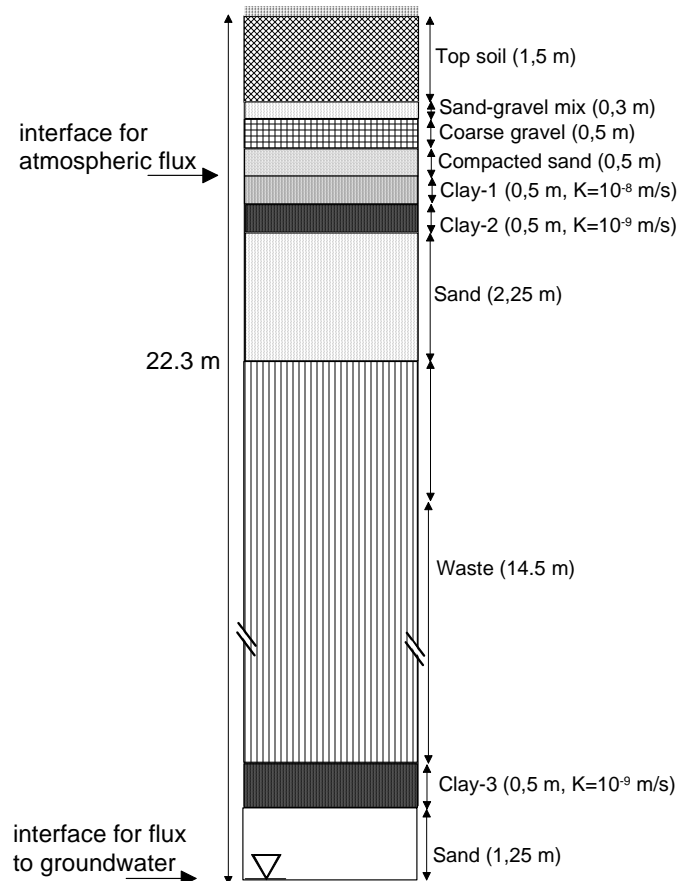


Figure 1 Vertical profile of waste material and protective layers.

Source term model

Leaching of ²²⁶Ra and its most important decay products ²²²Rn and ²¹⁰Pb was calculated using the one-dimensional variably-saturated flow and transport code HYDRUS-1D [7]. A conceptual model for the migration of the three radionuclides considered here is given in Fig. 2. The source term model calculates fluxes of dissolved radionuclides at the bottom of the repository, in addition to the flux of radon gas at the interface of the clay-1 layer and the overlying sand layer (see Fig. 1). The 1D computational domain excludes the first six soil layers, as they were treated in a separate 2D calculation. The calculated water flux at the bottom of the clay-2 layer will be used as top boundary for the 1D model.

Not including the first six soil layers implies that the effect of the first 2.8 m soil is not accounted for in calculating radon migration. This assumption is conservative as long as these protective layers are intact (i.e., during the institutional control period). However, the assumed condition becomes realistic when long term assessment is of concern. In such case, various

degradation processes may have lead to a partial removal of those layers.

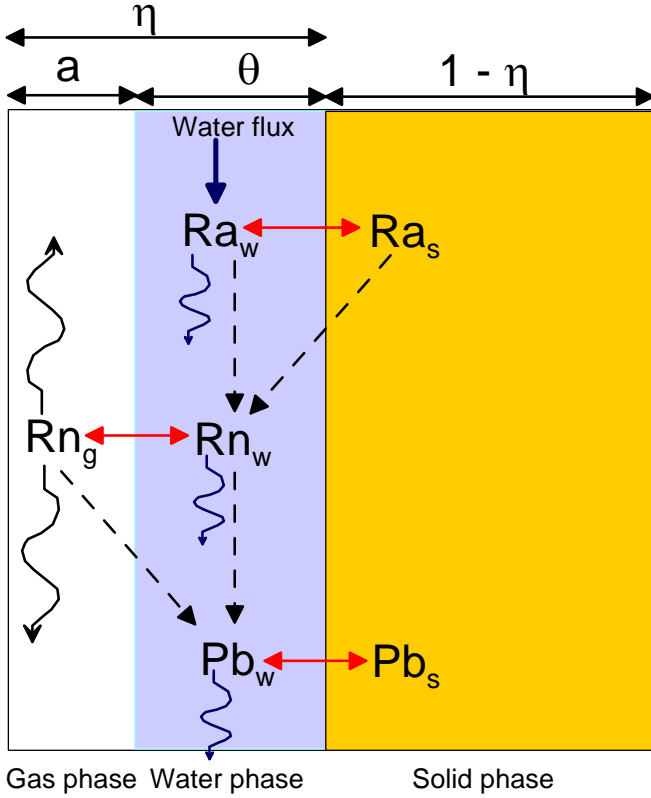


Figure 2 Partitioning of ^{226}Ra , ^{222}Rn , and ^{210}Pb between three phases of the porous medium. Soil parameters are: η = total porosity, θ = volumetric water content, $a = \eta - \theta$ = volumetric air content.

The presence of the radon gas requires that three phases are considered in the modelling, i.e. solid, liquid, and gaseous phase. If we conservatively assume radon (as a gas or dissolved) is not sorbed on the solid phase, partitioning between solid and liquid phase applies to radium and lead, whereas partitioning between liquid and gaseous phase applies to radon. The solid-liquid partitioning relationship is [8]:

$$C_s = k_d \times C_w \quad (1)$$

where C_s is solid phase concentration (Bq/kg soil), C_w is liquid phase concentration (Bq/m³), K_d is solid-liquid distribution coefficient (m³/kg soil). Partitioning between liquid and gaseous phase is defined in a similar way [8]:

$$C_w = k \times C_g \quad (2)$$

where C_g is concentration in the gas phase (Bq/m³ soil air), and k is Ostwald constant (-). The latter parameter may be calculated from the Henry constant, K_H [9]:

$$k = K_H \times R \times T \quad (3)$$

where R is universal gas constant and T absolute temperature (° Kelvin). The product $R \times T$ is 0.0244 atm×m³/mol. At 10 °C, $K_H = 14.6$ mol/atm×m³, hence $k = 0.356$. At 20 °C, $K_H = 10.7$ and $k = 0.26$ [10]. We assumed $k = 0.26$.

Total concentration of the porous medium, C_T (Bq/m³ porous medium), of each radionuclide is calculated from [8]:

$$C_T = \theta \times C_w + \rho_b \times C_s + a \times C_g \quad (4)$$

where ρ_b = dry bulk density (kg/m³), and all other parameters are as defined previously.

The partial differential equations governing one-dimensional transport of radionuclides involved in a sequential first-order decay chain during steady-state water flow in a variably saturated porous medium were solved using HYDRUS-1D. We assume radionuclides are transported by convection and dispersion in the liquid phase and by diffusion in the gaseous phase. Additional parameters for advective-dispersive transport in the liquid phase are pore-water diffusion coefficient, D_p (m²/d), and dispersivity, λ (m). Best estimate parameter values for D_p are given in Table 1. For λ a uniform value of 0.03 m was considered for the entire soil profile.

Transport in the gas phase requires estimates of the diffusion coefficient for the soil gas phase, $D_{p,g}$ (m²/d) [8]:

$$D_{p,g} = \xi_g \times D_{o,g} \quad (5)$$

where $D_{o,g}$ is free air diffusion coefficient (m²/d), and ξ_g is tortuosity factor for the gas phase, defined as $\xi_g = a^{7/3}/\theta_s^2$ [11]. A value of 0.864 m²/d was considered for $D_{o,g}$ and was taken from [12].

Modelling of the flux to the atmosphere requires a different type of soil boundary condition for volatile chemicals. A third-type (Cauchy) boundary condition was invoked, with an additional term that accounts for gas diffusion through a stagnant boundary layer of thickness d . The gas flux is proportional to the difference in gas concentrations below and above the boundary layer [13]. A value of 0.5 cm for d for bare soil was suggested by [13].

Table 1 Radionuclide specific parameter values.

Material	D_p (m ² /d)	K_d (m ³ /kg)
Ra		
Clay	6.2×10^{-5}	0.014
Waste	6.2×10^{-5}	2.7
Sand	6.2×10^{-5}	0/0.65
Rn		
Clay	8.65×10^{-5}	0
Waste	8.65×10^{-5}	0
Sand	8.65×10^{-5}	0
Pb		
Clay	6.2×10^{-5}	20
Waste	6.2×10^{-5}	0.27
Sand	6.2×10^{-5}	0

In the scoping calculations, several source term parameters will be given alternative values such that the effect of each parameter on the radionuclide behaviour can be assessed. Variations in the following parameters will be discussed:

- Clay layers: in the reference scenario, all three layers are present. Calculations will also be made with all three clay layers being degraded. Such a model presumably is more representative for the very long term, when natural processes and/or human actions have resulted in a deterioration of the protective clay layers.
- Presence of hot spots: several hot spots have been detected in the waste material. A field survey revealed radium concentrations in excess of 40 Bq/g in 15% of the samples [14]. Therefore, calculations with the source term model were carried out assuming a uniform concentration in the 14.5-m-thick waste zone of 40 Bq/g ^{226}Ra rather than 7 Bq/g. Since the 1D calculations provide fluxes per m^2 of surface area, the results can be extrapolated for any quantity (i.e. surface area) of hot spots (from 0 to 100 %). In the following discussion we assume 100% hot spots, i.e. the whole waste dump has 40 Bq/g radium as initial concentration.
- Reduced sorption capacity of waste material. Given the large variety in waste materials, a high level of sorption heterogeneity may be expected. Therefore, a K_d value $10\times$ smaller than the best estimate given in Table 1 was used for all materials.

In addition to the above alternative cases, several other parameters have been considered. These include degree of water saturation of the waste material at the time of waste emplacement (i.e., very dry or very wet), less permeable waste material, etc. [6]. Discussion of these cases was beyond the scope of the present paper. Therefore, only six cases will be discussed. A summary of calculation cases is shown in Table 2. Best estimate parameter values were used for the reference case (case 1) and case 4, but the effect of degraded clay layers was also investigated for the latter parameter set. For each alternative parameter set, calculations considered both intact and degraded clay layers.

Table 2 Cases considered in scoping calculations

Parameter	Intact clay layers	Degraded clay layers
Reference	1	4
Hot spots	2	5
Reduced sorption	3	6

Hydrogeological model

Radionuclide transport through groundwater is an important pathway for human exposure. In our assessment we conservatively assume the presence of a small well in the vicinity of the repository, say at 100 from the disposal facility. The well capacity is assumed small enough to have little or no effect on groundwater flow.

All groundwater flow calculations were done using the GMS-Modflow 96 software package [15]. Concentrations of

^{226}Ra , ^{222}Rn , and ^{210}Pb in the hypothetical well were calculated using a dilution factor. The latter was obtained by assuming transport of a non-adsorbing and non-decaying chemical from the bottom of the repository to the well. In this way, only dilution and dispersion in the aquifer is accounted for, while sorption has been neglected. As a result, calculated concentrations are conservative. Transport calculations were done using the MT3DMS software package [16].

Biosphere model

In the last step the annual dose for each radionuclide for a water well pathway is obtained by applying a biosphere model. The well pathway assumes that a self-sustaining farmer community uses water from the well as drinking water, for irrigation of crops, etc. The biosphere model accounts for all major exposure pathways, including ingestion of contaminated food or water, direct irradiation by contaminated soil or sediment, etc. [17].

Exposure to radon gas escaping through the degraded protective cap may also contribute to the total annual dose. The methodology for calculating the annual dose (Sv/y) for an individual that is staying for a given time on top of the repository is based on the approach described by [18]:

$$\text{Dose} = \text{exhalation rate} \times \text{dose factor} \times \text{radon concentration in air} \quad (6)$$

where exhalation rate is total radon flux density to atmosphere ($\text{atoms}/\text{cm}^2/\text{s}$), dose factor is $3.1 (\text{nSv}/\text{h})/(\text{Bq}/\text{m}^3)$, and radon concentration in air per unit of exhalation rate ($\text{Bq}/\text{m}^3)/(\text{atoms}/\text{cm}^2/\text{s})$ at 1.5 m above soil surface is depending on the stability of the atmospheric layers close to the soil surface. For weak mixing and normal mixing a value of 20, respectively 4 Bq/m^3 per unit of exhalation was proposed [18]. We assumed an intermediate value of 8 Bq/m^3 per unit of exhalation rate. The average exposure time for an individual from the reference group, i.e., a self-sustaining farmer community, was taken as 100 h/y. For example, an exhalation rate of $4.6 \times 10^6 \text{ Bq}/\text{m}^2/\text{y} = 6.95 \text{ atoms}/\text{cm}^2/\text{s}$ would result in an annual dose of approximately 17 μSv .

The time history of the total annual dose will be based on the contribution from the groundwater pathway and the atmospheric pathway. Only the maximum value and the time it occurs will be reported here.

RESULTS AND DISCUSSION

The 1D calculations using the source term model resulted in three types of output, notably (1) depth profiles of radionuclide concentrations in the relevant phases (water and solid phase for radium and lead and water and gas phase for radon) at selected time intervals, (2) radionuclide fluxes at the interface repository/groundwater, and (3) radon flux at the interface between clay-1 and the overlying sand (Fig. 1).

Depth profiles for various cases are given in Fig. 3. Only concentrations in water or gas phase are discussed. Concentrations on the solid phase may be easily calculated using Eq. (1). A comparison of ^{226}Ra concentrations for case 1, 3, 4, and 6 reveals that reduced sorption (case 3 and 6) results

in a higher pore water concentration in the waste zone and below and a faster leaching compared to the reference parameter set. Hence, fluxes for case 3 and 6 will be significantly higher than those for the other cases (see further). The effect of degraded clay layers, i.e. compare cases 1 with 4 and 3 with 6, has a much smaller effect on the downward movement of radium. Therefore, regarding radium fluxes to groundwater, the effect of reduced sorption (based on the assumptions used here) is expected to be much more important than the effect of degraded clay layers (see further).

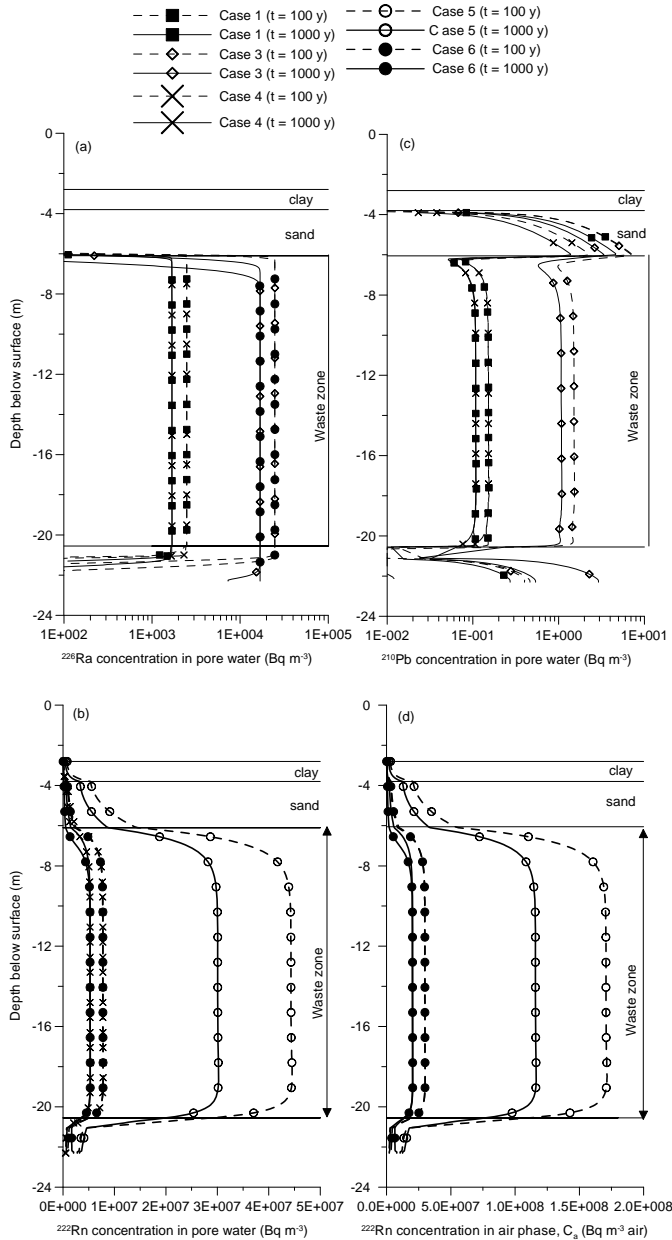


Figure 3 Calculated depth-profiles for (a) dissolved ^{226}Ra concentrations, (b) dissolved ^{222}Rn concentrations, (c) dissolved ^{210}Pb concentrations, and (d) ^{222}Rn concentrations in gas phase. For explanation of cases, see main text.

For an analysis of radon concentrations in pore water, also the radon concentrations in the gas phase has to be considered. In the waste zone, dissolved radon is determined by the concentrations of its parent, ^{226}Ra , whereas gaseous radon is determined by the partitioning law between water and gas phase (Eq. (2)). Above the waste zone, presence of gaseous radon is the result of upward diffusive transport via the air phase. When the clay layers are intact, gas transport is limited to the unsaturated sand, as no gas pathway exists in the saturated clay. When the clay has degraded to some more permeable and therefore unsaturated material, radon transport also takes place through the latter layer. We conservatively assumed the top of this layer coincides with the interface between soil and atmosphere (Fig. 1), hence exhalation of radon gas occurs.

Based on theoretical considerations, the diffusion length in unsaturated media is at most 2 m [12]. Our calculations indicate a diffusion length that is nearly twice as large. This is most probably due to the large concentration gradient above the waste zone.

Concentration profiles for ^{222}Rn are shown for cases 4, 5, and 6, i.e. the only cases for which upward transport through the gas phase may occur. Cases 4 and 6 display a very similar concentration profile, as a result of the assumption that radon is not adsorbed, and hence K_d parameters for radon are identical for case 4 and 6. Intuitively, one would expect ^{222}Rn to have $10\times$ higher pore water concentrations, as does its parent ^{226}Ra . However, ^{222}Rn is generated by decay of both dissolved and sorbed ^{226}Ra , and total radium activity has not changed. Hence, with no differences in K_d between case 4 and 6, radon concentration profiles are nearly identical. Evidently, when total initial radium increases by a factor of $40/7$ (case 5), decay products will increase the same amount.

A fairly uniform ^{210}Pb concentration profile is observed within the waste zone, where it is generated due to decay of ^{222}Rn in both water and gas phase. In the sand layer above the waste zone, the presence of ^{210}Pb is due entirely to decay of ^{222}Rn in the gas phase. Because we assumed lead is not sorbed on the sand (see Table 1), higher concentrations exist in the sand compared to the underlying waste material, where sorption is considered important. Although radon gas diffuses through the degraded clay layer, lead concentrations in the degraded clay are very small owing to the high sorption onto the clay. Note that we assume that the clay is only physically degraded, but not chemically. In other words, its hydraulic conductivity has increased considerably, but its sorption capacity is still at its initial value.

Analysis of concentration profiles for ^{210}Pb based on cases 1, 3, and 4 demonstrate a higher sensitivity with respect to the sorption parameter than to absence of clay layers. In the sand layer above the waste zone, lowest concentrations exist for case 4, whereas cases 1 and 3 have similar values. Lower concentrations for case 4 (degraded clay) are due to the lower accumulation of its parent ^{222}Rn in the sand layer, as a considerable amount of the radon gas will diffuse through the degraded layer to the atmosphere.

An analysis of the effect of variations in model parameters on radionuclide fluxes is based on Fig. 4. When all clay layers are intact, i.e. cases 1-2-3, largest effect is due to a reduced sorption (case 3). For the latter case, maximum ^{226}Ra flux is nearly three orders of magnitude larger than case 1 (also see

Table 3). Also note that the maximum flux for case 3 occurs considerably earlier than that for case 1 and 2, i.e., between 1000 and 2000 y for the former and 10000 y for the latter two. Fluxes for case 2 are only 40/7 times larger than case 1, because of the linearity between flux and source concentration. In other words, when the source concentration is increased by a factor of 40/7, fluxes increase likewise. When clay layers have degraded, fluxes increase by a factor of approximately 50 for case 4 and 5 compared to case 1 and 2, and nearly a factor of 10 for case 6 (compared to case 3). The effect of degraded clay layers is thus larger when best estimate sorption parameters are used than when a ten times smaller sorption is used. In the latter case, transport time smaller than half-life of ^{226}Ra ($T_{1/2} = 1600$ y), hence decay of ^{226}Ra is not yet influencing the flux values, whereas for the former case transport time is much longer and hence significantly affected by decay.

The flux history for ^{222}Rn under the condition of intact clay layers is determined in part by that of its parent ^{226}Ra and in part by the faster transport velocity of radon compared to radium. Owing to the assumed zero sorption of dissolved radon, its mobility in the water phase will be orders of magnitude higher than that of radium, especially in the lower clay-3 which isolates the waste at the bottom from groundwater. The peak flux appears around 2000 years, whereas that for radium it appeared around 10000 years (Fig. 4). When all clay layers have degraded, the radon flux is observed very soon after repository closure. This behaviour is due to the presence of a free gas phase between the bottom of the waste material and the groundwater table, in the absence of a low permeable clay layer. In other words, diffusion of radon through the gas phase is now determining the flux history, rather than diffusive transport through the water phase. There is one exception, however. Fig. 4 shows that case 6 has a maximum flux at a later time than that of case 4 and 5, although the initial flux for case 4 and 6 is identical, illustrating the same initial transport process, i.e., diffusion through the gas phase. However, since radium is 10 times less retarded compared to case 4 and 5, the contribution of the radon flux in the water phase to the total flux is not negligible anymore.

Flux history for ^{210}Pb is very similar to that of ^{222}Rn , both in the presence and absence of clay layers (Fig. 4). When transport in the water phase dominates the mobility of lead and its parent radon, i.e. in the presence of the clay layers, both radionuclides migrate at the same rate because we assumed zero sorption for both in the lowest sand layer (see Table 1). In the absence of clay layers, lead history is determined by that of its volatile parent radon.

A summary of maximum fluxes and doses is given in Table 3. Total dose for groundwater pathway includes contributions from all three radionuclides considered. However, in all cases the contribution of ^{226}Ra is far more important than that of ^{222}Rn and ^{210}Pb .

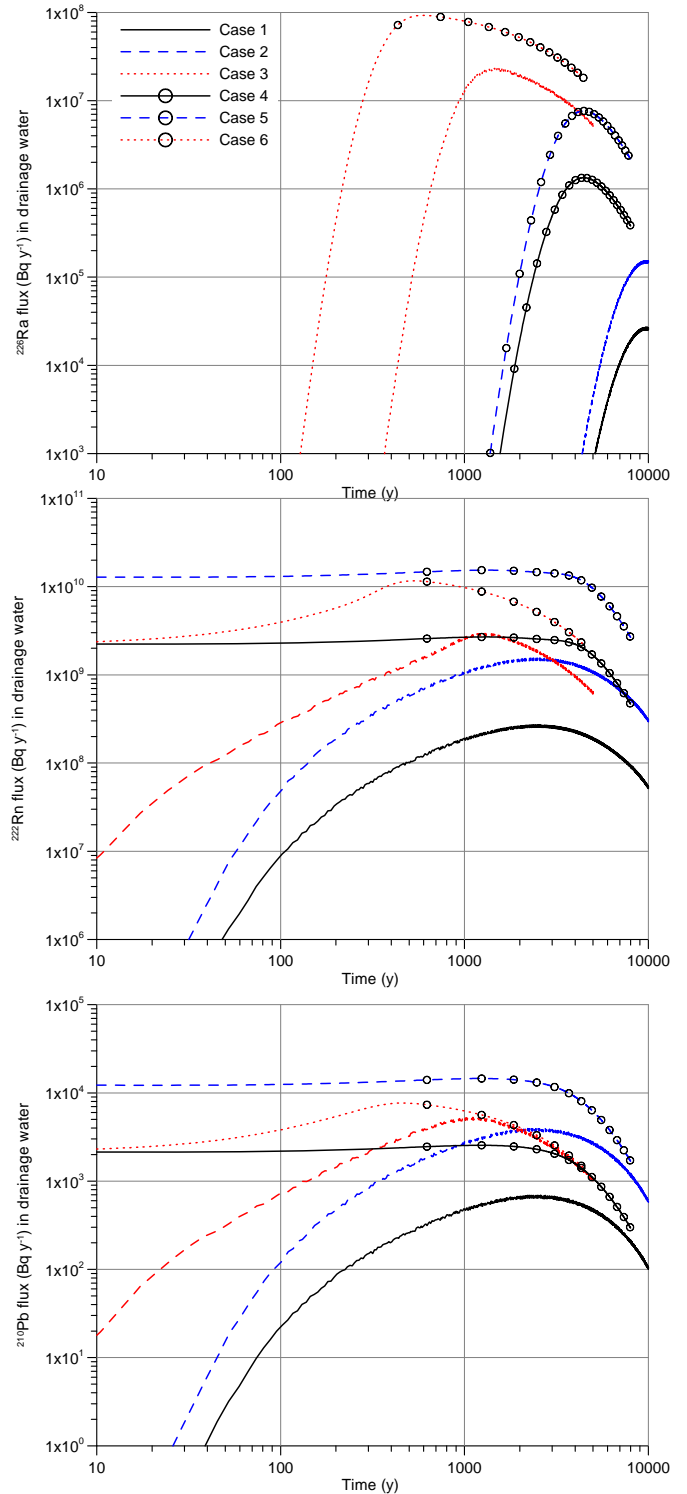


Figure 4 Calculated total radionuclide fluxes at the bottom of the repository. For explanation of cases, see main text.

Table 3 Maximum fluxes (F_{\max}) and dose (H_{\max}) for groundwater and atmospheric pathway, and combined total dose (H_{tot}) from both pathways.

Case	Groundwater pathway				Atmospheric pathway		Total dose H_{tot} (Sv/y)
	F_{max} (Bq/y)			H_{max} (Sv/y)	F_{max} (Bq/y)	H_{max} (Sv/y)	
	^{226}Ra	^{222}Rn	^{210}Pb				
1	2.7E4	2.7E8	6.9E2	1.1E-6	-	-	1.1E-6
2	1.5E5	1.5E9	3.9E3	6.0E-6	-	-	6.0E-6
3	1.3E7	2.7E9	5.5E3	6.5E-4	-	-	6.5E-4
4	1.3E6	2.7E9	2.5E3	4.8E-5	6.9E10	1.9E-5	4.8E-5
5	7.6E6	1.5E10	1.5E4	2.7E-4	3.9E11	1.1E-4	2.7E-4
6	9.3E7	1.2E10	7.7E3	2.3E-3	6.9E10	1.9E-5	2.3E-3

The maximum total dose was calculated as the sum of the doses due to the groundwater and atmospheric pathway. It is important to note that the time of the maximum flux, and hence also the dose, does not occur at the same time for both pathways. As is demonstrated in Fig. 4, the maximum ^{226}Ra flux to groundwater occurs between 4000 and 5000 y (case 4 and 5). The maximum dose due to the atmospheric pathway occurs during the first 100 years after repository closure. Between 100 and 1000 y, the dose drops by almost a factor of 2. Between 1000 and 5000 y, the dose further drops by at least one order of magnitude. Case 6 is an exception, with the maximum ^{226}Ra flux around 500 y. This means that the maximum dose for both pathways occurs almost at the same time. However, the maximum dose due to the atmospheric pathway is two orders of magnitude smaller than that of the groundwater pathway.

Compared to the background dose of 2-3 mSv/y, nearly all calculated doses are smaller. The only exception is case 6, for which the dose has the same order of magnitude as the background value. All other cases have a maximum dose that is one to three orders of magnitude smaller than the annual background dose.

The results clearly demonstrate that the dose contribution via the groundwater pathway is more important than that via the atmospheric pathway. Although the gaseous radon concentrations in the waste zone are quite high, the thickness of the soil layers considered in the calculations (3.25 m) was large enough to reduce the fluxes at the assumed interface soil/atmosphere to acceptable levels.

CONCLUSIONS

Long-term safety was investigated for a proposed surface disposal facility for very low-level radium bearing waste. Exposure pathways investigated were leaching to groundwater and exhalation to the atmosphere. For this purpose, a source term model, a groundwater flow and transport model, and a biosphere model were adopted. These models were run for a generic disposal concept and a set of best estimate model parameters. To assess the sensitivity of the model outcome with respect to some key parameters, alternative runs were carried out which included the following modifications: (1) the initial radium concentration in the waste zone was increased from 7 (best estimate) to 40 Bq/g, assumed uniformly distributed over the entire repository depth, to address the issue of waste heterogeneity (2) the sorption of radium and its daughter

nuclides was decreased by a factor of 10 to address the issue of sorption heterogeneity, and (3) waste confining clay layers were assumed to be degraded, in order to address the issue of increased leaching and radon exhalation. Parameters for the groundwater flow and transport model and biosphere model were kept constant.

Results show that the proposed design using the best estimate parameter set leads to an annual dose of nearly 10^{-6} Sv, which is approximately three orders of magnitude smaller than the background dose. When early degradation of the clay layers is accounted for, i.e. immediately after repository closure, the maximum dose using best estimate parameters increases about 40 times. However, the dose is still about two orders of magnitude below the background.

The effect of a higher initial radium concentration will depend on the fraction of the waste that contains hot spots. As an extreme case, we assumed 100% hot spots with a uniform concentration of 40 Bq/g. As expected, the increase in dose from 1.1×10^{-6} to 6×10^{-6} Sv was proportional to the increase in concentration. For smaller fractions of hot spots resulting doses can be easily derived.

The results further showed that reducing the sorption ten times increases the dose nearly 500 times. Such a dose would still be smaller than the background dose if clay layers are intact.

An important conclusion of this study is related to the significance of the atmospheric pathway. Radon exhalation happens only if clay layers have degraded. Under such conditions, doses are negligible compared to the doses due to leaching of radium and its daughter nuclides to groundwater.

The issue of degraded engineered barriers is an important one. In reality, as soon as institutional control has ended, the soil cover will degrade gradually to a condition where the least permeable clay layer becomes permeable also for radon gas. Because it is difficult if not impossible to predict when this will occur, we conservatively assumed it happens very soon after repository closure. Our calculations demonstrate that even under such highly unlikely situation doses due to radon exhalation are negligible. Furthermore, the accelerated leaching of radium and its daughter nuclides to groundwater owing to increased infiltration when the clay has degraded, does not lead to a dose that is higher than the annual background dose of 2-3 mSv.

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