Radioprotection, vol. **44**, n° 5 (2009) 701–706

DOI: 10.1051/radiopro/20095128

Prediction of concentration and model validation

Key issues for the consequence analysis of radioactive waste repositories

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Abstract. In this paper we examine some critical aspects concerning the justification of simplified radioecological models used in safety assessments for geological repositories. We propose a modelling approach for regulatory review of dose and risk calculations for a nuclear waste repository. The SSI modelling tools are applied to explore uncertainties in the size of the contaminant area due to leakage of radionuclides from a damaged nuclear waste canister. We demonstrate that an improved representation of geosphere transport processes will also enhance our understanding of radionuclide migration in the biosphere and provide a better basis for evaluating radiological consequences in the safety assessment.

1. INTRODUCTION

Post-closure safety assessments for nuclear waste repositories involve radioecological modelling for an underground source term. In this paper we discuss critical aspects concerning process understanding and justification of simplified radioecological models used for such safety assessments. This study is part of the Swedish Radiation Protection Authority's (SSI) work on reviewing the Swedish Nuclear Fuel and Waste Management Co's (SKB) most recent safety assessment, SR-Can.

One of the most challenging tasks in assessments of environmental doses and risk from an underground repository is to estimate radionuclide activity concentrations in various geologic strata in the future. For example, little is known about transport pathways through the quaternary deposits to the discharge points in surface waters and other recipients in the biosphere. Traditionally simplified compartmental models are used in safety assessment to describe the fate of radionuclides in surface environment. The possibility to test such models against more detailed process models and site specific data is of key importance for confidence in the safety assessment.

As part of SSI's review of SR-Can, alternative modelling approaches were developed to explore the importance of transport process descriptions in the assessment models. The modelling results were compared with the Landscape Dose Factors (LDFs) derived by SKB in SR-Can. LDF is a new methodology adopted by SKB in SR-Can. The LDFs are defined in the units of Sv/y per Bq/y and express all the radiological information about individual repository sites and ecosystems as a single, radionuclide-specific, number that relates geosphere releases to radiological dose. Further, we suggest a method for assessing model parameters using data from field tracer tests.

2. ALTERNATIVE APPROACHES

2.1 Estimation of contaminated area through transport path 1 and 2

We propose a schematic analysis to explore transport processes in the near surface. Based on models for groundwater and surface water flows, one can derive distributions of residence time for flow parcels from their origin at a leaking canister to a defined control section that would enclose the central discharge point in a watershed, as shown schematically in Figure 1. The stream tube associated with the passive

Article published by EDP Sciences and available at http://www.radioprotection.org
or http://dx.doi.org/10.1051/radiopro/20095128

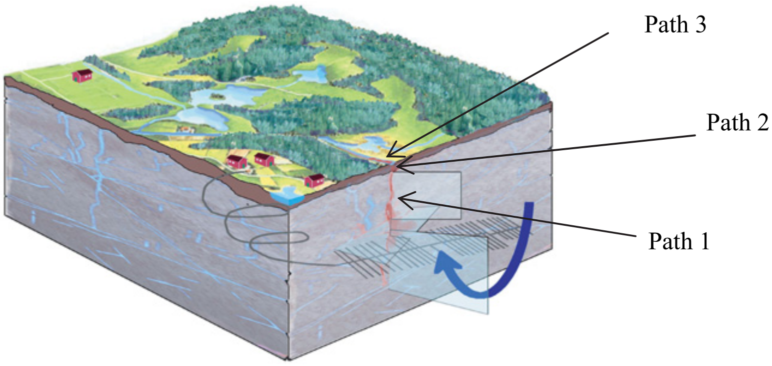

Figure 1. Schematic of the separation of pathway segments identified in the modelling approach. Path 1 – crystalline bedrock, Path 2 – Quaternary deposits, Path 3 – surface drainage system.

travel of solute elements will theoretically be very narrow, in the order of the same size as the mixing cross sectional area of the stream tube at the repository depth. A stream tube is defined as the surface created by the streamlines going through a closed contour and streamlines are the curves defined by tangency to the velocity field. However, diffusion and dispersion due to fracture intersections along the whole stream tube spreads radionuclides over a larger area, A, that can be estimated if we know the residence time. The spatial variance of the solute concentration in transverse components can be expressed according to Fischer et al. [1]:

$$\sigma_T = \sqrt{2D_T\tau} \tag{1}$$

where D_T are combined diffusion plus fracture dispersion coefficients in the transverse (T) directions, and τ is the water residence time, which is expressed as $\tau = L/u$ in which L is the mean path length in the stream tube and u is the velocity.

A rough estimation of the transverse dispersion of radionuclides along the length of the fracture beneath the Quaternary deposits (QD) and in the QD can be expressed as $4\sigma_T$, which means a contaminated area at discharge points might be expressed as:

$$A = \left(4\sigma_{T\_path1}\right)\left(4\sigma_{T\_path2}\right). \tag{2}$$

2.2 Transport models for path 2 and 3

Once the discharge area is determined, concentrations of radionuclide activity in media can be estimated from the rate of inflow of radionuclides into the area/strata and by accounting for accumulation processes. A modelling study has previously shown that discharge points are predominantly located in low-lying areas of the catchments where layers of QD often are relatively deep [2]. Such areas include riparian zones, wetlands and lakes. To demonstrate the concept of our approach the system described in section 2.1 is further simplified to the one shown in Figure 2 including a QD representing path 2 and a stream representing path 3 as well as an agricultural land to be irrigated by the water from the stream as a case study. Reactions along the transport pathways are assumed to be constant and that the flow field is steady. These are crude assumptions since, for instance, we do not take into account the changes in redox potential at the geosphere-biosphere interface, which can affect solid-liquid distribution coefficients (K_d) [3] and transient flow due to land rise. However, the latter effect was approximated by calculating the latent dose rates, i.e. the transformation of former QD and river sediments into agricultural land.

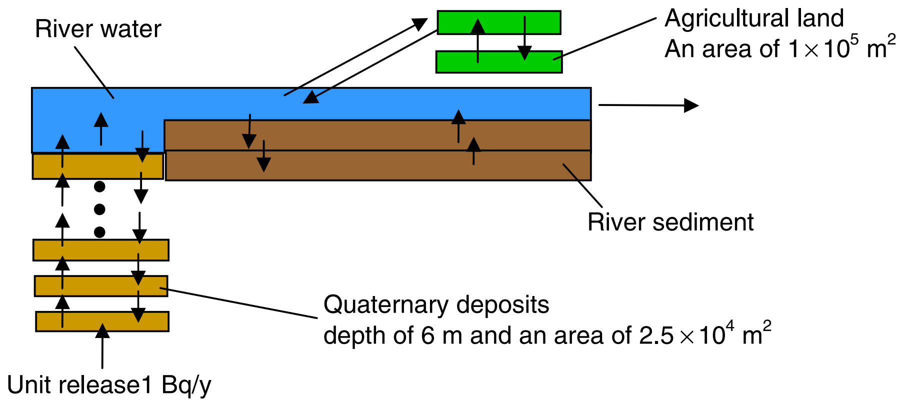

Figure 2. System of compartment models to describe transport through QD, river and irrigated agricultural land with unit release 1 Bq/y.

The radionuclide activity concentration along Path 2 is estimated by an advection-dispersion model [4] assuming constant moisture content and one-dimensional flow:

$$\frac{\partial C(x,t)}{\partial t} = \frac{D}{R_{QD}} \frac{\partial^2 C(x,t)}{\partial x^2} - \frac{v}{R_{QD}} \frac{\partial C(x,t)}{\partial x} - \lambda C(x,t) \tag{3}$$

where $C(x,t)$ is activity concentration of radionuclide in pore water [Bq/m^3], D is the dispersion coefficient [m^2/y], v is the mean pore water velocity [m/y], x is co-ordinate [m], t is time [y], R_{QD} is the retardation factor which quantifies the effect of radionuclide sorption on transport velocity, defined as

$$R_{QD} = 1 + K_d \frac{\rho}{\varepsilon} \tag{4}$$

where ρ is the bulk density of the QD [kg/m^3] and ε is porosity [-] and K_d is the solid-liquid distribution coefficient [m^3/kg].

Radionuclide transport in streams can be described in terms of advection, dispersion, exchange with hyporheic zones and adsorption to sediments. Here we use the Advective-Storage-Path (ASP) model [5] to describe radionuclide transport in a stream.

$$\frac{\partial C}{\partial t} + \frac{1}{A_T} \frac{\partial (AUC)}{\partial x} - D \frac{\partial^2 C}{\partial x^2} = J_S \tag{5}$$

where C is the dissolved activity concentration of radionuclide in the stream water [Bq/m^3], A_T [m^2] is the cross-sectional area of the main stream including side pockets, A is the cross-sectional area of the main stream excluding side pockets [m^2], U is the flow velocity in the main stream [m/s], and D is the main stream dispersion coefficient [m^2/s].

The net radionuclide flux [Bq/m^{3}s] in the dissolved phase in the stream water can be written as integrating over the distribution of transport pathways:

$$J_S = \frac{1}{2} \int_0^{\infty} f(T) \frac{P}{A} \xi \left(-V_Z(\tau,T)|_{\tau=0}\, c_d + (V_Z(\tau,T))|_{\tau=T}\, g_d \right) dT. \tag{6}$$

where g_d is solute mass per unit volume of water in the hyporheic zone [Bq/m^3], V_z is the infiltration velocity [m/s] into the bed in the direction of the streamlines denoted by $V_Z(\tau,T)|_{\tau=0}$ and exfiltration velocity out of the bed in the direction of the streamlines $V_Z(\tau,T)|_{\tau=T}$, $f(T)$ is the probability density function (PDF) of T weighted by the velocity component normal to the bed surface, V_n, T is the total residence time from inlet to exit of hyporheic flow path [s], τ is the exfiltration residence time [s]

($0 < \tau <$ T), P is the wetted perimeter [m], A is the cross-sectional area of the stream [m^2], and ξ is an area reduction factor equal to V_n/V_Z that accounts for the fact that the streamlines are not always perpendicular to the bed surface.

2.3 Implementation of the models

Compartmental models are often used to analyse the migration of contaminants in both abiotic and biotic parts in ecosystems due to the variety of geo- and biochemical processes. For instance the irrigation model [6] used in this analysis is a typical compartmental model. Because a fundamental assumption behind compartmental models is instantaneous mixing of solute in each compartment, the spatial distribution of contaminants in the system is not always well represented. Caution should be given for the number of compartments chosen to represent solute transport processes on ecological modelling in different media. Xu et al. [7] proposed the criteria for resolution-scales and parameterisation of compartmental models of hydrological and ecological mass flows.

There are similarities between a finite difference approximation of the advection-dispersion (A/D) type of equation and a compartmental model. Furthermore, when certain criteria are fulfilled compartmental models can provide identical solutions to those of the A/D equations [7]. Here, a compartmental model is used to solve the two transport problems of the system and an irrigation model shown in Figure 2 since this allows radionuclide chain decay to be handled conveniently and a consistent model structure to be used for the whole system. The models are implemented in Ecolego Toolbox [8].

In order to compare our results with SKB's LDFs values we adopt a unit release to the biosphere (1 Bq/y), i.e. our analysis starts from Path 2 using a unit continuous radionuclide release as the boundary condition and we use the same parameter values as in the SR-Can as much as possible. However, not all the parameter values used in our calculations are available from the SR-Can data base. In the next chapter we will show how the parameter value such as the advective velocity used in the stream model is derived from the tracer experiment, while the dispersion coefficients used in the estimation of contaminated area are adopted from the literature, Marsily [9]. Nevertheless, the highest values from the parameter interval given in Marsily [9] are used to give a largest possible size of the contaminated area from the estimation.

3. RESULTS AND DISCUSSIONS

Reviewing the discharge area data used in SR-Can, we found that the smallest discharge area during the entire simulation period was about 2.5 km^2 and the radiological consequences of canister leakage into this area was then analysed by SKB. Using equation (2) by accounting for both the spreading-length in the fracture and the QD we obtain a much smaller contaminated area of 25,000 m^2 [10] although, as mentioned previously, the highest values from the parameter interval in the literature [9] are used. It should be noted that the gaussian dispersion model used here may not capture the transport characteristics in heterogeneous fractured rock. Nevertheless this simple analysis strongly suggests that the discharge area may be significantly smaller than that assumed in SKB's SR-Can safety assessment.

Once the size of the discharge area is determined the activity concentration in the Quaternary deposits can be calculated using equation (3) and after that, the activity concentration in the water and sediment of the stream can be calculated using equation (5). Dose rates for different ecosystems can then be calculated using Aggregated Transfer Factors [11], the same procedure as SKB used in SR-Can. The details and the parameter values used in the calculation can be found in Xu et al. [10]. Dose rates at equilibrium obtained from the calculation are shown in Table 1. The results agree with our earlier study [12], which showed that Quaternary deposits serve the function of both retarding and accumulating radionuclides. As can been seen, the latent dose rates are 2 to 4 orders of magnitude higher than those estimated using the LDF-concept when the Quaternary deposits are transformed to agricultural land. ^{226}Ra is the exception since it has a short half life. Calculated results show that dose rates from river water are comparable to SKB's LDFs. The lowest dose rates for all radionuclides, except for ^{36}Cl, are obtained for agricultural land irrigated by river water. Latent dose rates from river sediments are not negligible compared with LDFs and they depend very much on the K_d values used in the calculation. This means that river sediments are a potential exposure source to individual humans.

As mentioned previously, a compartmental model was used to mimic A/D type transport model in our calculations. A large number of compartments are needed for the QD model because, for a moderately sorbing nuclide, the transport residence time through 6 metres of QD is about 150,000 years. When 60 compartments are used in the QD compartment model, the simulated fluxes are close to those obtained using a semi-analytical solution for the same problem [8].

There are no site-specific data available for parameters such as the advective velocity into sediment and the hydraulic radius for the river model. Therefore, we employ data obtained from a tracer experiment performed in Säva Brook in Uppland County [5] as a typical characterisation of an agricultural stream in a landscape type that is likely to exist if leakage occurs from the repository after a considerable land rise has occurred.

Table 1. Calculated dose rates [Sv/y per Bq/y] for various media comparing with the LDF [Sv/y per Bq/y].

Nuclides	QD latent	River water	Agricultural land	River latent	LDF
^{226}Ra	4.9×10^{-26}	2.5×10^{-29}	1.9×10^{-33}	2.2×10^{-29}	4.7×10^{-11}
^{36}Cl	3.0×10^{-10}	8.6×10^{-14}	6.5×10^{-14}	2.8×10^{-13}	8.1×10^{-15}
^{135}Cs	1.8×10^{-10}	3.7×10^{-11}	2.6×10^{-15}	1.7×10^{-13}	2.3×10^{-12}
^{129}I	1.5×10^{-9}	4.3×10^{-11}	2.9×10^{-13}	1.5×10^{-12}	1.6×10^{-11}
^{59}Ni	7.6×10^{-12}	4.5×10^{-15}	2.2×10^{-17}	7.6×10^{-15}	4.4×10^{-15}

The lumped parameter, the transfer rate for the water to the sediment, was obtained by fitting the simulated breakthrough curves with experimental data using 250 compartments to represent the water course in the model [8]. The calibrated lumped parameter value was then used in the assessment model and one compartment was used to represent the water course in the river model for assessment purposes. This is because the transport residence time in rivers is rather short and the up-stream boundary condition, with a continuous inflow, dominates the model behaviour.

4. CONCLUSIONS

This study shows that a good understanding of the size of discharge areas and dilution in the biosphere is critical for the justification of simplified compartmental dose models used in safety assessments for nuclear waste repositories. Further, it is important to verify the models used in the assessments against observations or field data with a view to demonstrate process understanding and the predictive capability The impact of discretisation of compartmental models may have significant effects on the results and should therefore be analysed when the models are implemented in the safety assessment.

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