Migration in the formations surrounding the host rock

PA model 'Aquifer'

OPERA-PU-GRS7222

Radioactive substances and ionizing radiation are used in medicine, industry, agriculture, research, education and electricity production. This generates radioactive waste. In the Netherlands, this waste is collected, treated and stored by COVRA (Centrale Organisatie Voor Radioactief Afval). After interim storage for a period of at least 100 years radioactive waste is intended for disposal. There is a world-wide scientific and technical consensus that geological disposal represents the safest long-term option for radioactive waste.

Geological disposal is emplacement of radioactive waste in deep underground formations. The goal of geological disposal is long-term isolation of radioactive waste from our living environment in order to avoid exposure of future generations to ionising radiation from the waste. OPERA (OnderzoeksProgramma Eindberging Radioactief Afval) is the Dutch research programme on geological disposal of radioactive waste.

Within OPERA, researchers of different organisations in different areas of expertise will cooperate on the initial, conditional Safety Cases for the host rocks Boom Clay and Zechstein rock salt. As the radioactive waste disposal process in the Netherlands is at an early, conceptual phase and the previous research programme has ended more than a decade ago, in OPERA a first preliminary or initial safety case will be developed to structure the research necessary for the eventual development of a repository in the Netherlands. The safety case is conditional since only the long-term safety of a generic repository will be assessed. OPERA is financed by the Dutch Ministry of Economic Affairs and the public limited liability company Electriciteits-Produktiemaatschappij Zuid-Nederland (EPZ) and coordinated by COVRA. Further details on OPERA and its outcomes can be accessed at www.covra.nl.

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Contents

Summary

Theme of the present report is the transport of radionuclides from the host rock of the repository system through the aquifer system in the formations surrounding the host rock to the biosphere. The host rock surrounding rock formations are the third compartment succeeding the source (waste container and EBS) and the host rock (Boom Clay). It consists of several hydrogeological layers with various flow characteristics.

A detailed discussion of the geological structure in the Netherlands yields several potential evolutions of a site in Boom Clay. These potential evolutions can be described by scenarios, which are applicable for performance assessments. In OPERA-PU-DLT621, for each of these scenarios transport times have been determined for the transport of dissolved species from the top and bottom of the host rock to the surface water.

This allows a relative simple PA model for the transport through the aquifers in the host rock surrounding rock formations: a model in which all parameters only vary along the flow paths as described in OPERA-PU-DLT621, but where all parameters are constant for all directions perpendicular to the flow path. The PA model includes necessarily radionuclide decay and ingrowth. The PA model can also take into account the effect of linear absorption on the transport times of radionuclides.

The numerical implementation has been tested for an aquifer model with homogeneous properties (porosity, retardation). The calculations have been performed by the ORCHESTRA code. By benchmarking with the code POSA it has been shown that ORCHESTRA is a suitable tool.

Samenvatting

Het onderwerp van dit rapport is het transport van radionucliden vanuit het gastgesteente door de omliggende geologische formaties naar de biosfeer. De om het gastgesteente liggende geologische formaties vormen het derde compartiment na de bronterm (afvalcontainer en EBS) en het gastgesteente zelf (Boomse Klei). De omliggende formaties bestaan uit meerdere hydrogeologische lagen met verschillende stroomeigenschappen.

Een gedetailleerde analyse van de geologische structuur van Nederland laat verschillende mogelijke evolutiepaden zien van een eindbergingssite in de Boomse klei. Deze mogelijke evoluties worden beschreven door scenario's, die gebruikt worden bij de performance analyse. In OPERA-PU-DLT621 zijn voor elk van deze scenario’s transporttijden voor opgeloste stoffen bepaald vanaf de boven- en onderkant van het gastgesteente naar het oppervlaktewater.

Hierdoor kan het transport worden gemodelleerd met een relatief eenvoudig mathematisch 1-dimensionaal model. Uiteraard bevat dit model radioactief verval en ingroei. Ook kan het effect van lineaire absorptie op de transporttijden met dit model worden verrekend.

Het rekenmodel is getest voor een enkel laags systeem met homogene eigenschappen (porositeit, retardatie). Berekeningen zijn uitgevoerd met ORCHESTRA. Benchmarking met de code POSA laat zien dat ORCHESTRA een geschikte tool is.

1. Introduction

1.1. Background

The five-year research programme for the geological disposal of radioactive waste – OPERA - started on 7 July 2011 with an open invitation for research proposals. In those proposals, research was proposed for the tasks described in the OPERA Research Plan.
This report describes the execution and results of the research proposed for task 7.2.2 with the following title in the Research Plan: *PA model for radionuclide migration in the rock formations surrounding the host rock*. The main objective of this task is to develop a code for the calculation of the migration of radionuclides from the host rock (i.e. Boom Clay) to the biosphere. The proposed modelling and calculation approach is based on the findings of WP 6.2.2 (*Modelling approach for radionuclide migration in the surrounding rock formations*) and will serve as direct input to Task 7.2.4 (*Integrated modelling environment for the safety assessment*) that will be used for the sensitivity analyses and safety assessment calculations (WP 7.3.3).

1.2. Objectives

The conceptual PA model applied in OPERA represents the region between the disposed waste and potential receptors (i.e. humans) in our environment. This region is conceptually divided into compartments. In the OPERA research program these compartments are summarized for a hypothetical repository in clay and include the Waste-EBS compartment, the Host Rock (Boom Clay) compartment, the Rock Formations surrounding the host rock, and Biosphere (including potential receptors).

Nomenclature: Overburden
For the remainder of this report, *the Rock Formations surrounding the host rock* are addressed as the *Overburden*, but note that the rock formations below the host rocks are also included in this compartment

The basic premise of the Performance Assessment is that the radionuclides have to move from the waste through these compartments to reach the receptors in the biosphere. The various scenarios differ in the processes that drive the radionuclide migration through each of the compartments and/or the pathways available for radionuclide transport through the compartments.
For the Normal Evolution and most Altered Evolution scenarios, radionuclide transport through the overburden takes place in the aquifer[1] system. The complete PA model consists of the models "Waste" (including the EBS), "Clay", "Aquifer" and "Biosphere".

Nomenclature: PA model Aquifer
The implementation of the PA model for the transport through the aquifer system in the overburden is referred to as the *PA model "Aquifer"*.

1.3. Realization

This report has been compiled by NRG, TNO and GRS. TNO has compiled information on the geological structure of the overburden. The model for the calculations has been developed on the basis of results from groundwater model calculations performed by Deltares. The

[1] Aquifer: underground, typically saturated layers of water-bearing permeable rock, rock fractures or unconsolidated materials from which water can be extracted using a water well. The aquifer may contain impermeable layers like clay which alternate with the permeable layers.

calculations by the computer codes ORCHESTRA and POSA have been performed by NRG and GRS, respectively.

1.4. Explanation of contents

The OPERA safety assessment methodology and the conceptual model for the performance assessment (PA) are summarised in Chapter 2, which also includes a description of the interfaces of the overburden with adjacent compartments (host rock and biosphere). Chapter 3 provides the mathematical model, while in Chapter 4 the appropriate computer codes ORCHESTRA and POSA for the numerical simulations are described. In Chapter 4 the results of the calculations applying ORCHESTRA and POSA are compared, and in Chapter 5 conclusions and recommendations are given.

2. Conceptual model for the PA model 'Aquifer'

2.1. OPERA Safety assessment methodology

The present report describes the formulation and implementation of the assessment model for the overburden (host rock surrounding rock formations) as defined in step four of the recommended safety assessment methodology for the OPERA project (see Grupa, 2014, p.11, p.14)). Step 4, formulation and implementation of Assessment Models requires:

1. A conceptual model. The conceptual model provides a description of the components of the system and the interactions between these components.
2. A mathematical model, which is a mathematical representation of the features and processes included in the conceptual model.
3. A computer code, which is a software implementation of the mathematical model that facilitates performance of the assessment calculations.

The present chapter provides the description of the conceptual model for radionuclide transport through the aquifer system in the overburden. Chapter 3 describes the mathematical model chosen to represent this transport. The computer code and the testing of the correct implementation of the mathematical model are presented in Chapter 4.

2.2. Compartment Overburden

The conceptual model applied in OPERA represents the region between the disposed waste and potential receptors (i.e. humans) in our environment. The region between the waste and the receptor is conceptually divided into compartments, which are in line with the multi-barrier system approach described in [Verhoef, 2014; p.3].

The following compartments are defined in the PA model for OPERA:

- The Waste-EBS compartment, consisting of the waste form, the waste package and the repository building & affected materials (or enclosing engineered barrier system);
- The Host Rock (Boom Clay);
- The Overburden (note that the rock formations below the host rock are included in this compartment, too)
- The Biosphere.

Figure 2-1 shows an overview of these compartments, where the repository (Waste-EBS) is shown in red, host rock clay in green, overburden in brown, aquifers in blue lines, and the biosphere in light blue.

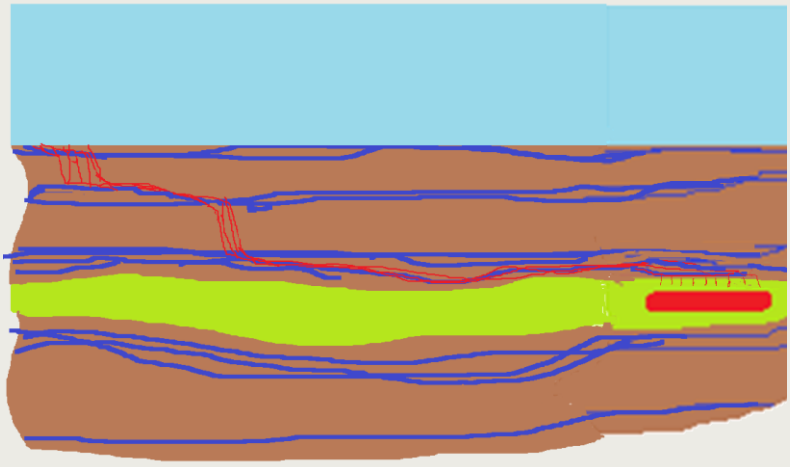

Figure 2-1 Schematic overview of the compartments

It should be noted that OPERA-PU-DLT621 also treats the flow paths that start at the bottom of the host rock. That path is not indicated in Figure 2-1, and generally these flow paths will not surface in the same location as the flow paths that start at the top of the host rock formation.

In the context of the OPERA Programme the overburden is defined as the subsurface compartment above the host rock (Rupel Clay Member or informally the Boom clay) up to the earth's surface and excluding the soil and surface water bodies like rivers, canals and lakes; these are part of the biosphere compartment.

In OPERA-PU-DLT621 [Valstar & Goorden, 2016] an extensive study has been performed to determine streamlines and transport times (or residence times) for dissolved material released from the Boom clay layer at all areas in The Netherlands where Boom clay is sufficiently thick and lies sufficiently deep.

Figure 2-1 gives an artist's impression of the streamlines (shown as red lines) for a potential site for the disposal facility, and shows the transport through the aquifers and the cross overs between the aquifers at the places where the vertical transport occurs. The radionuclide transport follows the streamlines determined in OPERA-PU-DLT621, and is presented by the red lines.

Following the geohydrological studies in OPERA-PU-DLT621, the transport through the overburden can be characterized as follows:

- The transport can be approximated by advective transport (fully driven by the groundwater flow through the aquifer system).
- The transport times for streamlines starting at a Boom clay layer at at least 500 m depth vary from 2,000 years to more than 10 million years, but are mostly in the range from 100,000 years to 2 million years.
- The length of the streamlines is in the range of 1.2 km to 135 km, and the streamlines cross various geological layers.
- The residence times in the deeper aquifers (i.e. below NHI[2]: deeper than 350 m for most of The Netherlands) is more than 2,000 years, and this minimum is generally insensitive to the surface conditions (see OPERA-PU-DLT621).

[2] Nationaal Hydrologisch Instrumentarium – NHI' [Hoogewoud et al., 2010].

- The residence times in the upper aquifers (not deeper than 300 m) ranges from a few hundreds to few 10,000 years for present day climate, and this residence time is very sensitive to surface climate and the surface morphology.

The long residence times in the deep aquifers is amongst others caused by the fact that the streamlines have to cross some low-permeable layers which are characterized by a relatively large clay content. Besides low permeability, the clay also has adsorbing capacities. Although in the past most national programs included this adsorption (see e.g. PAGIS and EVEREST for an overview), more recently some important safety reports (SAFIR-2, Dossier Argile) ignored absorption in these layers. Dossier Argile explains this as follows: "no retention property linked to geochemistry has been assigned to these formations, as a cautious approach, although it is probable that these formations or part of these have such properties."

In the PA-tool 'Aquifer' geochemical retention is optional. Depending on the goal of the assessment, e.g. a robust conservative deterministic calculation or an exploring more realistic estimate, the geochemical retention can be included or not.

2.3. Spatial geometry of the transport through the aquifer system

The shape of the nuclide transport pathway is determined by the cross width size of the repository or a section of the repository, the length of the transport path in the aquifer system and the amount of transverse dispersion (perpendicular to the flow). Typical dimensions of the nuclide bearing part of the aquifer system are: path length of 20 to 50 km, cross width of 2 to 3 km, and transverse extension of 10 to 100 m. Because of the relatively small transverse extension, the transport pattern has the form of a "sheet". An impression of this pattern is given in Figure 2-2.

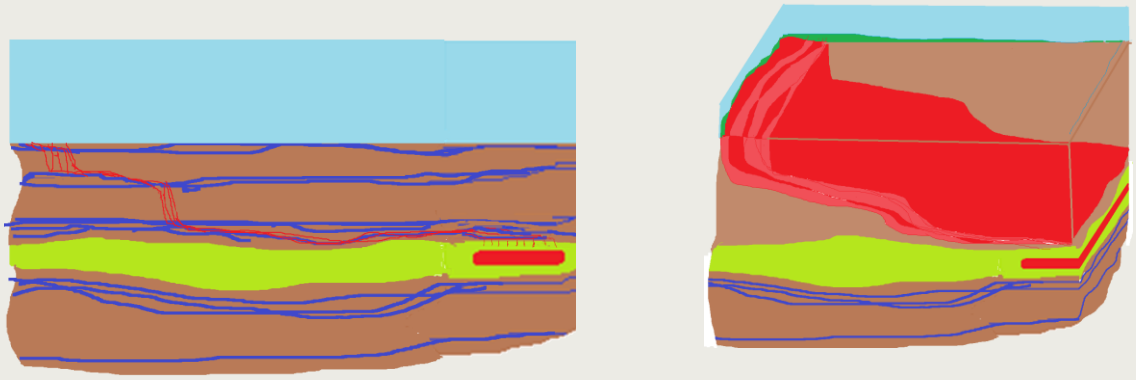

Figure 2-2 Impression of the flow transport pattern: 3D on the right side

The length and width of the nuclide bearing flow sheet are determined by the repository layout and the geological setting. The transversal width is a result of dispersion and diffusion. The results of the streamline analysis in OPERA-PU-DLT621 [Valstar & Goorden, 2016] suggest that there is little transverse dispersion, although this aspect has not been analyzed in great depth. The analyses of three pathlines (Section 3.4 of OPERA-PU-DLT621) show concentration reductions to 28%, 1.3% and 0.9% compared to the initial values.

The area where the radionuclides reach the surface can be as small as a few kilometers wide by 100 m long. In the biosphere model description [Grupa et al., 2017] it is pointed out that the reference group (or critical group) is a small agricultural community living in the area where the radionuclides surface. When there is little dispersion, this area is small enough to be 'covered' by one community, i.e. all nuclides that reach the surface enter the biosphere of one community.

Although this concept, that all radionuclides enter the biosphere of a small reference group is conservative, it is still realistic. For the conceptual model this leads to a simplification: it is sufficient to be able to estimate the nuclide flux (in Bq/year) that reaches the surface. The concentration of the radionuclides in the aquifer water is less relevant (as long as there are no strong non-linear effects in the adsorption), i.e. there is an uncertainty in the amount of water in the aquifer in which the nuclides are transported (for a larger amount of water the concentration is lower), but this does not affect the nuclide flux.

Although the concentration in the aquifer water is of less importance, it should be noted that:

- the concentration in the surface waters are important for the exposure, as pointed out in the following section (interface aquifer system - biosphere)
- for the well scenario it should be taken into account that the concentration in the aquifer water is inaccurate, and - in a conservative calculation scheme - too high.

In combination with some mathematical operations described in Chapter 3, the transport of radionuclide-bearing species through the aquifer system can be modelled for the PA by a mathematical 1-dimensional advective transport model characterized by a transport path length and a groundwater flow speed, with an emphasis on predicting the radionuclide flux into the surface water. This PA model for the aquifer was also used for the PA of the Belgium disposal concept in Boom clay (at 200 m depth) in the SAFIR study [Preter et al., 2001] and the Dutch studies carried out in the OPLA research program for disposal in rock salt [OPLA, 1989] and [Prij et al, 1993].

2.4.Longitudinal dispersion

Longitudinal dispersion can be defined as the spreading of dissolved species along the flow's longitudinal axis. This process will lead to a change in characteristics of an initial state of high concentration and low spatial variance, to a downstream state of lower concentration and higher spatial variance.

In isotropic media, the longitudinal dispersion is of the same intensity as the transverse dispersion. But because the aquifers are non-isotropic (vertical permeability is low), the longitudinal dispersion in the aquifer system will be much larger than the vertical transverse dispersion.

In principle, longitudinal dispersion could be important for the PA model. However, dispersive transport depends on the second derivate of the spatial concentration profile (in more than 1 dimension: the divergence of the gradient of the concentration: ΔC). For a typical geometry (streamline length of many kilometers) and a typical release profile from the clay to the aquifer (at least several 10000 years) the second derivate is small and advective transport will dominate over dispersive transport. In general, ignoring longitudinal dispersion will lead to slightly conservative[3] maximum values of the nuclide flux to the surface waters in the PA.

[3] Some nuclides, of which the half-life is shorter than the transport time, will mathematically show a higher peak value, since a fraction of the nuclides will reach the surface faster due to longitudinal dispersion, and these nuclides will incur less decay. However, since the transport time in the preceding clay compartment is much longer, such nuclides will have decayed before reaching the aquifer.

2.5. *Interfaces of the overburden with the adjacent compartments*

The compartment 'Overburden' interfaces with the adjacent compartments Boom Clay and Biosphere. This section describes the conceptual models for these interfaces.

Clay-Aquifer interface

Radionuclide bearing species from the repository migrate through the clay and enter the aquifer at about 500 m depth, and these species reach the surface through the aquifer system, where they can enter the biosphere. These interfaces need to be addressed in the conceptual and mathematical models, as well as in the computer code.

On a conceptual level of the model, the interface between the clay and the aquifer at 500 m depth is regarded as a sharp transition from almost impermeable clay to a permeable, sandy aquifer. Actually, the transition is gradual: at the top and the bottom of the clay layer more and more sand sheets are observed [ONDRAF / NIRAS, 2012], and the character of the soil gradually changes from low permeable, highly adsorbing clay to permeable, low adsorbing sandy soils.

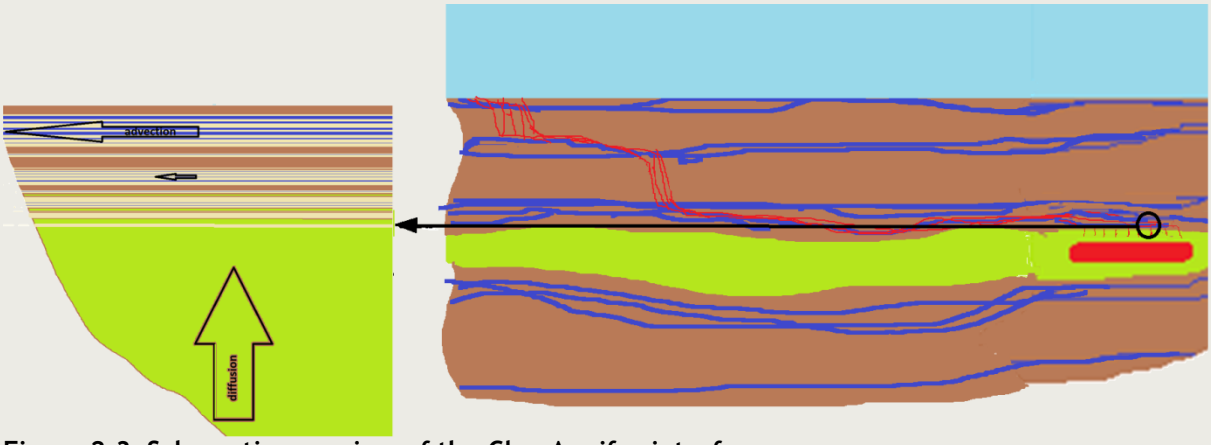

Figure 2-3 Schematic overview of the Clay-Aquifer interface

The radionuclide diffusion in clay is driven by the concentration gradient. The aquifer acts as a sink at the interface: due to the flow of water in the sandy layers in the aquifer, the water in the aquifer is continuously refreshed, and radionuclides are removed from the interface, causing a relatively low concentration in the aquifer water at the interface. Sandy layers with a low water flow velocity are inefficient as a sink compared to layers with a higher water velocity. Sandy layers with an extremely low water velocity (deeper in the clay, not connected to a percolating network) don't act as a sink at all.

The diffusive migration through the clay can be approached by assuming that the radionuclide concentration in the aquifer water is zero, although in the numerical implementation of the PA model the actual concentration in the aquifer is used. This interface allows the clay model to calculate the rate at which the radionuclides transfer from the clay to the aquifer.

The average concentration of radionuclides in the aquifer water can be calculated by dividing the radionuclides transfer rate by the water yield of the aquifer.

Aquifer-Biosphere interface

Living organisms take up nutrients that are dissolved in the water. In this process, however, they also take up other materials, such as radionuclides, from the water. In effect, the concentration of radionuclides in a biotope is in equilibrium with the concentration of

radionuclides in the water that the biotope is in contact with. Biosphere models provide the relation between the concentration of radionuclides in the water and the distribution of radionuclides over the various organisms in the biotope, among which humans.

A biotope feeds on one or more of the following water bodies: **river, well, pond, lake and wetland (Dutch: polder)**, providing radionuclide transport **pathways** from the overburden to the biosphere. A biotope can also feed on the sea, but the radionuclide transfer is minor compared to the other pathways because of the enormous dilution in the large amount of water in the sea.

- Well: contaminated groundwater withdrawn from aquifers is used as irrigation water and drinking water for human and farm animals.
- River or lakes: contaminated water is used for irrigation and drinking purposes. Both arable land and pasture are irrigated. Fish in large rivers and lakes are caught for human consumption.
- Wetland (polder): soil water in the topsoil is directly contaminated by the upward movement of groundwater when the water table is high. Plants may grow on the soil and be used for food or animal feed.

Dose conversion coefficients (DCC's) for the radionuclides which may be released in the biosphere are calculated in OPERA-PU-NRG7231 [Grupa et al., 2015b] for release in a river, well, pond/lake and wetland.

The conceptual model of the aquifer-biosphere interface is based on the volume of water in a relevant water body, the inflow of radionuclides into this body and the in- and outflow of the water. The mathematical model is essentially the balancing equation for the radionuclides in the water, and the key parameter in which this information is summarized is the *dilution factor* for the water body.

Since the water uptake by the biotope is usually small compared to the other flows, the concentration in the water is not determined by biological factors at all. Therefore, the calculation of the concentration in the surface water bodies is *technically* considered to be part of the hydrological model. Interestingly, some of the dilution factors are driven by human activities, e.g. how much water is drawn from a well. However, these human actions are not regarded as part of the biosphere.

The water body dilution factors, human behaviour and biosphere dose conversion factors depend on the climate, and will therefore be derived for various climates.

2.6.Initial state and future evolution of the overburden

The initial and future evolution of the geosphere was extensively assessed in WP4.1 *Geology and geohydrological behavior of the geosphere* and reported in [Ten Veen et al., 2015]. That study focused on tectonic and climatic processes, with human activities being regarded as modulator of these processes. The following future climate scenarios for the Normal Evolution Scenario were identified and described in [Ten Veen et al., 2015]:

- The present-day temperate maritime climate
- The Mediterranean climate
- The Boreal climate
- The Periglacial climate

Glacial conditions were also assessed although they are considered only as an alternative evolution scenario in [Ten Veen et al., 2015].

Possible future states of the disposal system that may affect the performance of the disposal system were evaluated in Task 7.1.1 *Scenario development*. In that task scenarios relevant for the assessment of the long-term safety of the repository in Boom Clay were evaluated and altered evolutions of these scenarios were defined (see [Grupa & Wildenborg, 2016] for more detail).

3. Mathematical PA model for transport in aquifer system

This chapter describes the mathematical model for the transport of radionuclides in the aquifer system. The mathematical model is based on the conceptual model and the work done in OPERA-PU-DLT621, and is implemented or adequately approached by the numerical model in the codes described in Chapter 4.

3.1. The migration model

For the PA model it is assumed that the geometry of the nuclide bearing flow sheet through the aquifer flow system can be simplified to a sheet of about 2 to 135 km long (length of the flow path), about 0.2 to 5 kilometres wide (width of the repository) and 1 to 10 m high (thickness of the flow sheet). The water velocity and absorption in this sheet are constant, the velocity ranging from 0.1 to 1 m/year.

In the PA model, the geometry of the flow sheet through the aquifer system is approximated by a 1-dimensional geometry, i.e. all parameters only change in the flow direction (X-axis), but not along the Y- and Z-axes perpendicular to the flow direction. In the Y- and Z- direction, at the boundaries of the nuclide bearing flow sheet, the dispersion in the Y- and Z- direction is ignored, which is slightly conservative. Mathematically this translates to a hypothetical aquifer that is bounded by an impermeable wall.

The flux J_i of radionuclide i through a cross section of the aquifer at position x is:

$$J_i(x,t) = vC_i(x,t)A(x)\eta(x)$$

$J_i(x)$ flux (Bq/year) of radionuclide i at position x along the nuclide flow sheet,
t time (s),
v effective flow velocity (in the X-direction) of the water in the aquifer (m/s),
$C_i(x)$ concentration (Bq/m^3) of radionuclide i dissolved in the aquifer water,
A cross section of the part of the aquifer where nuclide transport takes place (m^2),
η porosity of the aquifer.

The following sections show how the parameters in this equations relate to the travel times (also termed "residence times") determined in OPERA-PU-DLT621.

3.1.1. Transport through a uniform aquifer

A uniform aquifer is uniform along its flow path, i.e. the parameters v, A, η, and adsorption are constant along the flow path.

For this system the generally known advection equation can be used to get directly to the resulting differential equation. However, in order to incorporate radioactive decay and ingrowth, and to obtain unique definitions of all parameters in the equation, the derivation of this equation is given for the 1-dimensional case.

The change of the amount of nuclide i in the spatial volume between position x and x+∂x in the aquifer can be calculated from the following mass balance, where M_i (Bq) is the amount of radionuclide i in volume between x and x+∂x and ΔM_i (Bq) is the increase of this amount between time t and t+Δt:

Inflow at x between time t and Δt: $\Delta M_i = vC_i(x,t)A\eta\Delta t$
Outflow at x+∂x between time t and Δt: $\Delta M_i = -vC_i(x+\partial x,t)A\eta\Delta t$
Decay: $\Delta M_i = -\lambda_i M_i \Delta t$
Ingrowth from parent-nuclide j: $\Delta M_i = \lambda_i M_j \Delta t$

So:

$$\Delta M_{i=} vC_i(x,t)A\eta\Delta t - vC_i(x+\partial x,t)A\eta\Delta t - \lambda_i M_i\Delta t + \lambda_i M_j\Delta t$$

The total concentration $C_{total,i}$ (Bq/m^3) in the volume of length ∂x is $C_{total,i} = M_i/(\partial x A)$. Substituting this in the equation above gives:

$$\Delta C_{total,i} = -v\eta[(C_i(x+\partial x,t) - C_i(x,t))/\partial x]\Delta t - \lambda_i C_{total,i}\Delta t + \lambda_i C_{total,j}\Delta t$$

For ∂x and Δt approaching zero, this gives the common 1D-advective transport equation including radioactive decay and ingrowth:

$$\frac{\partial C_{total,i}(x,t)}{\partial t} = -v\eta\frac{\partial C_i(x,t)}{\partial x} - \lambda_i C_{total,i}(x,t) + \lambda_i C_{total,j}(x,t)$$

Also the Darcy velocity or Darcy flux v_D can be used: $v_D = v\eta$, note that:

1. the discharge q (m^3/s) of this aquifer is: $q = vA\eta = v_D A$
2. the transport time t (s) over length L (m) in the aquifer is: $t = \frac{L}{v} = \frac{L\eta}{v_D}$
3. the solid material may absorb a fraction of the nuclides

Optional adsorption is included in the mathematical model as follows. Adsorption is characterized by the amount of nuclides adsorbed on a given mass of solid material and the concentration of the radionuclide in the water surrounding this mass of solid material.

Considering a volume V of saturated porous material,

the mass of the solid is:	$M_s = \rho V$
the amount of nuclide i adsorbed is:	$M_{i,adsorbed} = M_{i,adsorbed}(M_s)$
the volume of the water is:	$V_{water} = \eta V$
the amount of nuclide i dissolved is:	$M_{i,dissolved} = C_i\eta V$

So the total concentration (dissolved and adsorbed) $C_{total,i}$ is:

$$C_{total,i} = \frac{M_{i,adsorbed}(M_s)}{V} + C_i\eta$$

Often $M_{i,\ adsorbed}$ is proportional with M_s and C_i for a broad range of chemical conditions. In that case, using a proportionality constant K_d (m^3/kg):

$$M_{i,adsorbed}(M_s) = K_d M_s C_i$$

So, the total concentration of nuclide i in a volume V is:

$$C_{total,i} = \frac{K_d M_s C_i}{V} + C_i\eta = \frac{K_d\rho V C_i}{V} + C_i\eta = C_i(K_d\rho + \eta) = C_i\eta R_i$$

where R_i is the retention factor or retardation factor for nuclide i.

Substituting this into the advection equation and dividing by η R_i gives:

$$\frac{\partial C_i(x,t)}{\partial t} = -\left(\frac{v}{R_i}\right)\frac{\partial C_i(x,t)}{\partial x} - \lambda_i C_i(x,t) + \lambda_i \left(\frac{R_j}{R_i}\right) C_j(x,t)$$

In the case of no absorption, R_i = 1 must be chosen for all nuclides.

For radionuclides that have no radioactive parent in the system (C_j = 0) the following simplification is possible. Introduction of $C'_i(x,t)$:

$$C'_i(x,t) = C_i(x,t)\exp(\lambda_i t)$$

Substituting C'_i into the advection equation gives (if C_j = 0) the normal advection equation:

$$\frac{\partial C'_i(x,t)}{\partial t} = -\left(\frac{v}{R}\right)\frac{\partial C'_i(x,t)}{\partial x}$$

Thus, for nuclides with no parents in the system, the transport equation without decay can be solved resulting in C'_i and after that the effect of decay can be introduced by using:

$$C_i(x,t) = C'_i(x,t)e^{-\lambda_i t}.$$

There are two types of solutions for this system.

Type 1: Boundary condition at t =0

Assume that the concentration profile at t=0 is $C^0_i(x)$.

So:

$$C'_i(x,t=0) = C_i^0(x)$$

In that case, a solution of the equation is:

$$C'_i(x,t) = C_i^0\left(x - v, \frac{t}{R_i}\right)$$

and with decay:

$$C'_i(x,t) = C_i^0\left(x - v, \frac{t}{R_i}\right)e^{-\lambda_i t}$$

This means, that if a given concentration profile exists in the aquifer, the profile will translate along the aquifer over time and all concentrations will decrease by a factor $e^{-\lambda_i t}$, but its shape will not change.

Type 2: boundary condition at x =0

Alternatively, assume that the concentration over time is known at x = 0:

So:

$$C'_i(x=0,t) = C_i^{00}(t)$$

In that case, a solution of the equation is:

$$C_i'(x,t) = C_i^{00}\left(t - x\,\frac{R_i}{v}\right)$$

and with decay:

$$C_i'(x,t) = C_i^{00}\left(t - x\,\frac{R_i}{v}\right)e^{-\lambda_i t}$$

If the end of the aquifer is at x = L, we get for C_i(L,t):

$$C_i(L,t) = C_i^{00}\left(t - L\,\frac{R_i}{v}\right)e^{-\lambda_i t}$$

So, the concentration pattern at x = 0 will also occur at the end of the aquifer, however delayed by time $t_{residence} = (LR_i/v)$ and reduced by the decay factor $e^{-\lambda_i t}$.

Obviously, a short peak of non-decaying, non-adsorbed tracer material released at x = 0 and t = 0 will occur (as a short peak) at the exit of the aquifer at time $t = L/v$, which is precisely the transport time through the aquifer system (or residence time in the aquifer system).

Decay chains

For nuclides that have radioactive parents in the system (C_j > 0) analytical solutions are less straightforward. The PA-programs in which the advection equation is implemented use a numerical scheme to solve the equation, including the ingrowth.

3.1.2. Transport through a non-uniform 1-dimensional aquifer

The streamlines identified in OPERA-PU-DLT621 follow a path where water velocity, cross section, porosity and adsorption characteristic change along the path, when crossing various geological layers with different properties.

Streamlines with constant v, R_i and Rj, but varying cross section and porosity

For sections of the aquifer stream line where v is constant, the advective flux equation can be written as:

$$J_i(x,t) = vC_i(x,t)A(x)\eta(x)$$

$J_i(x)$	flux (Bq/year) of radionuclide i at position x
x	length along the streamline, where x=0 is at the clay-overburden interface
t	time (s)
v	effective flow velocity (in the X-direction) of the water in the aquifer (m/s)
$C_i(x)$	concentration (Bq/m^3) of radionuclide i dissolved in the aquifer water
$A(x)$	cross section of the nuclide bearing part of the aquifer at position x (m^2)
$\eta(x)$	porosity of the aquifer at position x (1)

Now we introduce a 1-dimensional concentration C^1:

$$C_i^1(x,t) = Ci(x,t)\,A(x)\,\eta(x)$$

This will lead to the following advection equation for sections in the streamline where v, R_i and R_j are constant:

$$\frac{\partial C_i^1(x,t)}{\partial t} = -\left(\frac{v}{R_i}\right)\frac{\partial C_i^1(x,t)}{\partial x} - \lambda_i C_i^1(x,t) + \lambda_i \frac{R_j}{R_i} C_j^1(x,t)$$

Important is to note that it is possible to calculate the outflux of this streamline section at x_{out} if the influx of radionuclides at the beginning of this streamline (at x_{in}) is given, even without knowing A(x) and η(x).

Since $C^1_i(x,t)$ can be calculated for this condition, an estimate of the concentration can be obtained from $C^1_i(x,t)$, if estimates of A(x) and η(x) are available.

Variation of v and R along the streamline

The transport along the streamlines is then approached by transport through a sequence of segments (or layers) with different characteristics. Each segment can be considered a uniform aquifer. Key is the mathematical description of the interfaces between these segments.

The mass balance at the interface between two segments requires that all nuclides that leave a segment enter the next segment

So, at the interface at x_s between segments n and n+1:

$$J_i(x_s,t) = v_n C_{n,i}(x_s,t) A_n \eta_n = v_{n+1} C_{n+1,i}(x_s,t) A_{n+1} \eta_{n+1} =$$

$$= v_n C^1_{n,i}(x_s,t) = v_{n+1} C^1_{n+1,i}(x_s,t)$$

Residence times

A short peak of non-decaying, non-adsorbed tracer material released at x = 0 and t = 0 will occur (as a short peak) at the exit of segment 1 at time $t = L_1 / v_1$ independent of the A(x) and η(x) profile along this segment.

For type 2 conditions the transport of non-adsorbing radionuclides with no radioactive parents in the system can be represented by a uniform aquifer, where the total residence time is:

$$t_{residence} = \frac{L_1}{v_1} + \frac{L_2}{v_2} + \frac{L_3}{v_3} + \cdots$$

This can be achieved by using a suitable effective flow velocity ($v_)$ in the equation:

$$v = \frac{L_{streamline}}{t_{residence}}$$

The nuclide flux (in Bq/year) to the surface water can be calculated using the total residence time, without detailed knowledge about the cross section of the aquifer, porosity and residence times in the subsequent geological layers. Concentrations in the ground water can be estimated based on estimated A, η and segment specific residence times, but these have no impact on the nuclide outflux (in Bq/year).

For a nuclide that is adsorbed, the (prolonged) transport time is:

$$t_{retarded} = \frac{L_1 R_{i,1}}{v_1} + \frac{L_2 R_{i,2}}{v_2} + \frac{L_3 R_{i,3}}{v_3} + \cdots$$

For type 2 conditions the transport of radionuclides with no radioactive parents in the system can be represented by a uniform aquifer where:

$$\frac{L_{streamline} R_{i,eff}}{v} = t_{retarded} = \frac{L_1 R_{i,1}}{v_1} + \frac{L_2 R_{i,2}}{v_2} + \frac{L_3 R_{i,3}}{v_3} + \cdots$$

So, it is possible to calculate the nuclide flux (in Bq/year) to the surface water for adsorbed nuclides if in each segment "n" $t_{residence,n}$ (for unadsorbed material) and $R_{i,n}$ is known. Concentrations in the ground water can be estimated based on estimated A and η, but these have no impact on the nuclide outflux (in Bq/year).

Decay chains

For nuclides that have radioactive parents in the system (C_j > 0) analytical solutions are less straightforward. The PA-programs in which the advection equation is implemented use a numerical scheme to solve the equation for the sequence of segments, including the ingrowth.

Input data

The transport model requires the following input data:
- The total residence time in the aquifer,
- Indicative length of the streamline and indicative cross section of the aquifer,

and for adsorbed species:
- The retardation factor in each streamline segment
- The residence time in each streamline segment

The length of the streamline and the segment specific residence times in the aquifer are determined by particle tracking and detailed flow calculations within task 6.2.1. Optionally, radionuclide specific retardation coefficients can be used.

The flow path starts at the top or the bottom of the clay layer and ends at the top of the overburden. The connection between the diffusion dominated region (clay) and the advection dominated region (aquifer) is realized by a diffusion connection.

3.2.Dispersion

To test whether dispersion has an impact on the radionuclide flux into the surface waters, a dispersion term has to be included in the transport equation. Mathematically, dispersion is treated similar to diffusion, where the dispersion coefficient (m^2/s) is:

$$D_{dispersion} = vL + GD_w$$

where:
v effective flow velocity (in the X-direction) of the water in the aquifer (m/s)
L geometry coefficient accounting for the pore structure driven dispersion due to the

water flow - also called dispersion length (m)

G geometry factor accounting for the pore structure (i.e. tortuosity, constrictivity)

D_w diffusion coefficient of the solute in free water [m^2/s],

Combining the advective and dispersive mass transport balance gives the dispersion-advection equation, which is given in Appendix 2.

3.3. The interface with the clay

If the advective transport in the aquifer is much faster than the diffusive transport in the clay, the concentration in the aquifer will be close to zero in comparison with the concentration in the clay.

The rate at which the radionuclides leave the clay formation depends on the (apparent) diffusion constant in the clay, the concentration gradient in the clay, and the length and width of the area where the radionuclides leave the clay and enter the aquifer (see Figure 3-1). The latter are roughly equal to the size of the repository, i.e. several kilometres wide by several kilometres long.

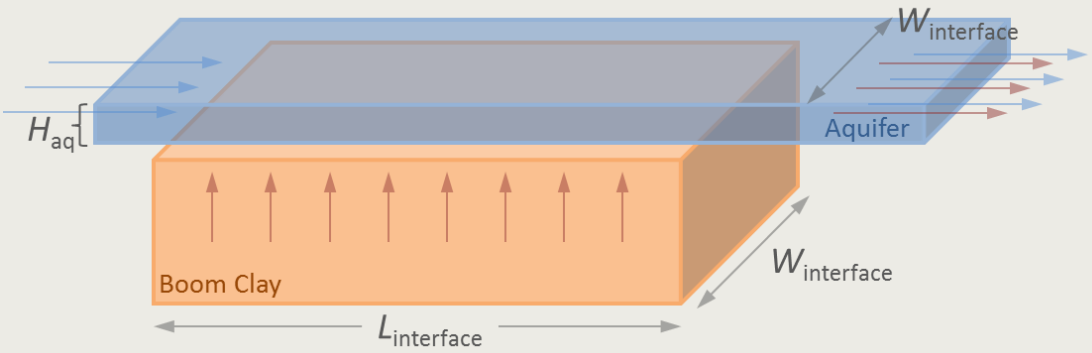

Figure 3-1 Schematic of RN transfer at Clay - Aquifer interface

The rate at which radionuclides are removed from the interface area depends on the effective velocity of the water in the aquifer (v), and the width and height of that part of aquifer through which the nuclides are transported (H_{aq}). For a width that is about equal to the width of the repository ($W_{interface}$), the flux through the interface ($J_{interface}$) becomes:

$$J_{interface} = \eta G D_w \frac{\partial C}{\partial z} L_{interface} W_{interface} = C v H_{aq} W_{interface}$$

D_w diffusion coefficient or diffusivity of the solute in free water or 'free-solution' diffusion coefficient (in absence of porous medium) [m2/s],

η total porosity [-],

G geometry factor accounting for the pore structure (i.e. tortuosity, constrictivity),

$\partial C/\partial z$ concentration gradient of the nuclide species in the clay near the interface

$L_{interface}$ length of the clay-aquifer interface where nuclides leave the clay layer, measured in the direction of the aquifer flow

$W_{interface}$ width of the clay-aquifer interface where nuclides leave the clay layer, measured horizontally and perpendicular to direction of the aquifer flow

C concentration of the nuclide-species in the aquifer water

v effective velocity of the water in the aquifer

H_{aq} height of the part of the aquifer through which nuclides are transported

A conservative (maximum) estimate of the diffusive transport through the clay can be obtained by assuming that the (boundary) concentration in the aquifer water on top of the clay is zero. In reality these concentrations can only be larger than zero, in which case dC/dz and thus the diffusive fluxes are overestimated in the model.

The order of magnitude of this overestimation is likely to be small, but can be estimated by comparing calculated concentrations in the source (repository) with those calculated in the aquifer. Because the overall diffusive flux through the clay layer will be driven by the total concentration gradient in the clay layer, a small deviation from zero at the aquifer side of the clay layer will hardly effect the fluxes through the clay layer.

3.4. The interface with the biosphere

The aquifer PA model gives a good prediction of the nuclide flux. The predicted concentration at the end of the aquifer does not consider the dilution in the biosphere water. Actually, the refreshment rate of surface waters is high in comparison to the influx of the aquifer water. Therefore, for each surface water body, a dilution factor has to be determined.

Generic mathematical model

Each water body where the biotope feeds on can be characterized by:

- the volume of water: V (m^3);
- inflows of water:
  - ✓ a 'fresh water' inflow, $Q_{in,fresh}$ (m^3/year)
  - ✓ an inflow of contaminated water from the aquifer $Q_{in,aq}$ (m^3/year) and a nuclide inflow of $J_{in,aq}$ (Bq/year)
- outflows of water:
  - ✓ water uptake by the biotope $Q_{out,bio}$ (m^3/year)
  - ✓ evaporation $Q_{out,evaporation}$ (m^3/year)
  - ✓ flow of water to the next water compartment, eventually the sea $Q_{out,flow}$ (m^3/year)

Provided that V is constant: $Q_{in,fresh} + Q_{in,aq} = Q_{out,bio} + Q_{out,evaporation} + Q_{out,flow}$

Since the nuclide inflow changes only very slowly (hundreds of years), the concentration of radionuclides in the water C (Bq/m^3) is quasi stationary:

$$C = \frac{J_{in,aq}}{Q_{in,fresh} + Q_{in,aq} - Q_{out,evaporation}} = \frac{QJ_{in,aq}}{Q_{out,bio} + Q_{out,flow}}$$

Generally:

$$Q_{in,fresh} \approx Q_{out,flow} \gg \max(Q_{in,aq},\ Q_{out,evaporation},\ Q_{out,flow})$$

So:

$$C = \frac{J_{in,aq}}{Q_{in,fresh}} = \frac{J_{in,aq}}{Q_{out,flow}}$$

where:

C	concentration of radionuclides in the biosphere water (Bq/m^3)
$J_{in,aq}$	nuclide inflow from the aquifer into the biosphere water(Bq/year)
$Q_{in,fresh}$	'fresh water' inflow, $Q_{in,fresh}$ (m^3/year)
$Q_{out,flow}$	flow of water to the next water compartment, eventually the sea (m^3/year)

Since the water uptake by the biotope is usually small compared to the other flows, the concentration in the water is not determined by biological factors at all. Therefore, this calculation is technically considered to be part of the hydrological model.

4. Computer code

4.1. The ORCHESTRA computer code

The open source reactive transport modelling framework ORCHESTRA [Meeussen, 2003] was used as the main tool for the OPERA PA calculations. ORCHESTRA stands for Objects Representing CHEmical Speciation and TRAnsport and ORCHESTRA is a versatile framework that is widely used for development of state-of-the-art mechanistic models [Filius et al., 2000], [Weng et al., 2005], [Weng et al., 2008], but also for applying these models for risk assessment of complex soil or cement-based systems [Schröder et al, 2005], [Schröder et al, 2008], [Sarkar et al, 2010]. Although the principal option for a coupling of ORCHESTRA with a suitable THM-code is considered, it is assumed to be of minor relevance for the OPERA Safety Case and no further details are given here. Unlike other programs used in PA, the open, object-oriented structure of ORCHESTRA enables the user to define new sub-models enabling bridging of the different conceptual levels of the OPERA programme in WP6.1 and WP7. I.e., instead of trying to approximate a particular outcome of OPERA WP6.1, ORCHESTRA enables us to represent each sub-model algorithm provided in WP6 exactly. Thus, the ORCHESTRA framework facilitates and simplifies the conversion of the complex scientific output of WP6.1 into a tested, validated, well-documented and more efficient PA model representation. Different from other programs, the model definitions within ORCHESTRA are not built in the source code but can be defined as input at run time, making it possible to store the model definitions in a data base that can be accessed and altered by the user. There are no restrictions on the choice of the chemical models and transport equations, new models can be implemented without adjustment of the source code.

ORCHESTRA is also very suitable for uncertainty analysis because model equations are not hard coded in source code but given as (text) input at run time. It allows any model parameter variable to be defined as input (including reaction constants etc.). Apart from modelling radionuclide migration, ORCHESTRA may also be used to address several topics related to geochemical interactions in WP5.1. ORCHESTRA therefore provides directly a consistent and correlated framework for incorporating the impact of the chemical environment in the PA.

ORCHESTRA meets the following requirements which make it very suitable for the PA calculations including uncertainty analysis:

- stochastic treatment of the relevant input parameters,
- specification of the stochastic parameter values as input at the run-time,
- computational efficiency.

The transport by diffusion and the hydrological dispersion are neglected. The aquifer compartment is divided into 50 connected calculation cells. The reactive advective transport between cells is calculated numerically in small time steps by a mixing-cell transport algorithm. The mixing-cell concept implies that transport systems can be composed of (well-mixed) cells and connections between these cells. The cells contain the information on the local physical and chemical composition, while the connections between the cells contain the mass transport equations (in this case advection). Both the connections between the cells, but also the literal equations involved in mass transport, are defined in text input files and can be modified or extended for specific purposes. Effectively, the mixing-cell concept results in a finite difference scheme, central in space and forward in time, that is solved by an explicit sequential non-iterative approach (SNIA). The chemical equilibrium in each cell is calculated by iteration after every transport step. The radioactive decay reactions are solved using the same time step as the transport processes.

4.2. The POSA computer code

POSA is a module of the computer code package RepoTREND [Reiche et al, 2011]. It was developed from a former computer code of GRS (chetlin), which was applied in many research projects in Germany, applying modern programming tools in C++ language. Details of the computer code are given in [Reiche et al, 2011].

The release of radionuclides from the near field in the first phase of OPAP has been calculated by the computer code CLAYPOS [Reiche et al, 2011]. CLAYPOS is also part of the computer code package RepoTREND [Reiche et al, 2011]. The results of the calculations are time-dependent values of flows and concentrations of all considered radionuclides at the interface of the clay and the aquifer (gsk file of CLAYPOS). These data are part of the input to POSA, see below.

The transport of radionuclides in the aquifer is modelled as one-dimensional. The path is divided into 230 cells of 100 m length. Advective flow, dispersion, and sorption are taken into account; diffusion is neglected.

4.3. Output: Safety and performance indicators

The OPERA compartment models will be integrated in *a PA integrated modelling environment* enabled to repeat calculations of predefined scenarios. This *PA integrated modelling environment* will allow the calculation of the Safety and Performance indicators [NEA, 2012].

The indicators related to the 'Overburden' compartment are summarized in Table 4-1. The output from the PA model 'Aquifer', necessary for the calculation of the indicators related to the aquifer compartment, reduces to the following three sets of data:

(1) activity concentration [Bq/m^3] of radionuclide *n* in the aquifer water in any cell of the compartment,
(2) activity [Bq] of radionuclide *n* in aquifer water in the compartment 'Overburden',
(3) activity flux [Bq/yr] of radionuclide *n* released from compartment 'Overburden'.

The PA model 'Aquifer' was set up to provide these output datasets for each of the radionuclides considered in the Performance Assessment. These output datasets are shown in the following in terms of amount (mol) instead of activity (Bq). In future calculations, these data can be transformed to activities. The data set “activity (amount) in the compartment” is not calculated in the POSA code, which is used for benchmarking.

Table 4-1 Indicators for the OPERA Safety Case related to the compartment Overburden

Indicator	*Type*	*Output from the PA model aquifer necessary for the calculation of the indicator*
Radiotoxicity concentration in aquifer water	PI	average activity concentration [Bq/m^3] of radionuclide n in the 'aquifer' water; the average activity can be determined based on either the activity concentration in each cell of the compartment or the (total) activity in the compartment "Overburden'
Radiotoxicity in the compartment 'Overburden'	PI	activity [Bq] of radionuclide in compartment 'Overburden'
Radiotoxicity flux from compartment 'Overburden'	PI, SI	activity flux [Bq/yr] of radionuclide released from compartment 'Overburden'
Time-integrated radiotoxicity flux from the compartment 'Overburden'	PI	activity flux [Bq/yr] of radionuclide released from compartment 'Overburden'
Contribution of each safety function	PI	activity [Bq] of radionuclide n in compartment 'Overburden'
Transport time through compartments	PI	activity flux [Bq/yr] of radionuclide released from compartment 'Overburden'
Performance of the integrated repository system*	PI	activity flux [Bq] of radionuclide n in compartment Overburden
Retardation due to migration through 'Overburden'*	PI	activity flux [Bq/yr] of radionuclide n released from compartment 'Overburden'

SI-safety indicator; PI-performance indicator

* Both activity and radiotoxicity based. For purpose of internal consistency checks as part of QA and comparison reasons, the use of activity-based indicator as defined by SCK·CEN [Marivoet et al., 2010] was proposed to be calculated as well in the OPERA safety assessments.

4.4. Test of the implementation of the PA model 'Aquifer'

In order to validate the implementation of the PA model 'Aquifer' in ORCHESTRA, a number of test calculations have been performed, where the results of the PA model 'Aquifer' in ORCHESTRA have been compared with the results for the same test calculations with POSA.

The radionuclide concentration maxima and the related time points at the interface with the biosphere for the ORCHESTRA calculation and POSA are given in Table 4-2.

Table 4-2 Concentration maxima in the aquifer near the interface with the biosphere

Radionuclide	*ORCHESTRA*		*POSA*	
	Maximum total concentration [mol/m³]	*Time points of maximum concentration [yr]*	*Maximum total concentration [mol/m³]*	*Time points of maximum concentration [yr]*
Se-79	5.51E-08	9.99e+4	5.70E-08	9.67e+4
Tc-99	1.43E-10	2.90e+6	1.03E-10	3.31e+6
I-129	1.11E-06	9.99e+4	1.15E-06	9.90e+4
Th-232	5.48E-06	3.63e+7	5.66E-06	3.54e+7
Np-237*	8.92E-12	2.50e+6	8.94E-12	2.70e+6
U-238	4.93E-03	2.30e+7	5.06E-03	2.20e+7

* Total dissolved concentration, no difference is made between Np-237 from initial inventory and ingrowth

The POSA and ORCHESTRA results show a good agreement of the calculated concentrations within the time range analyzed (see Figure 4-1). The main difference between both calculations is the earlier breakthrough shown by the ORCHESTRA concentrations. The curves of Tc-99 differ slightly in the two calculations; in POSA the breakthrough is a little bit later and consequently the concentrations and the maxima are lower due to radioactive decay.

Figure 4-1 presents the calculated concentration in the aquifer water as the amount of dissolved nuclides per m^3 of water.

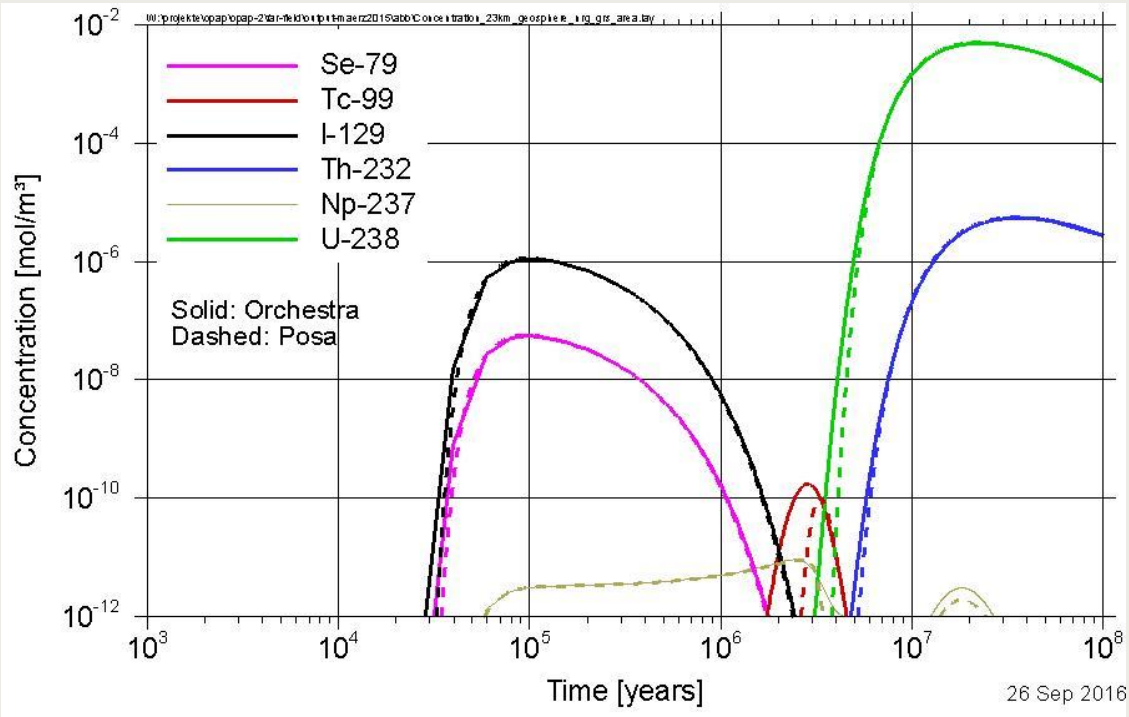

Figure 4-1 Time dependent ORCHESTRA and POSA calculated nuclide concentrations at the end of the transport path in the aquifer

In Figure 4-2 the radionuclide flow rates (or fluxes) at the end of the transport path are compared for ORCHESTRA and POSA calculations. The differences in the curves are similar to the concentrations shown in Figure 4-1.

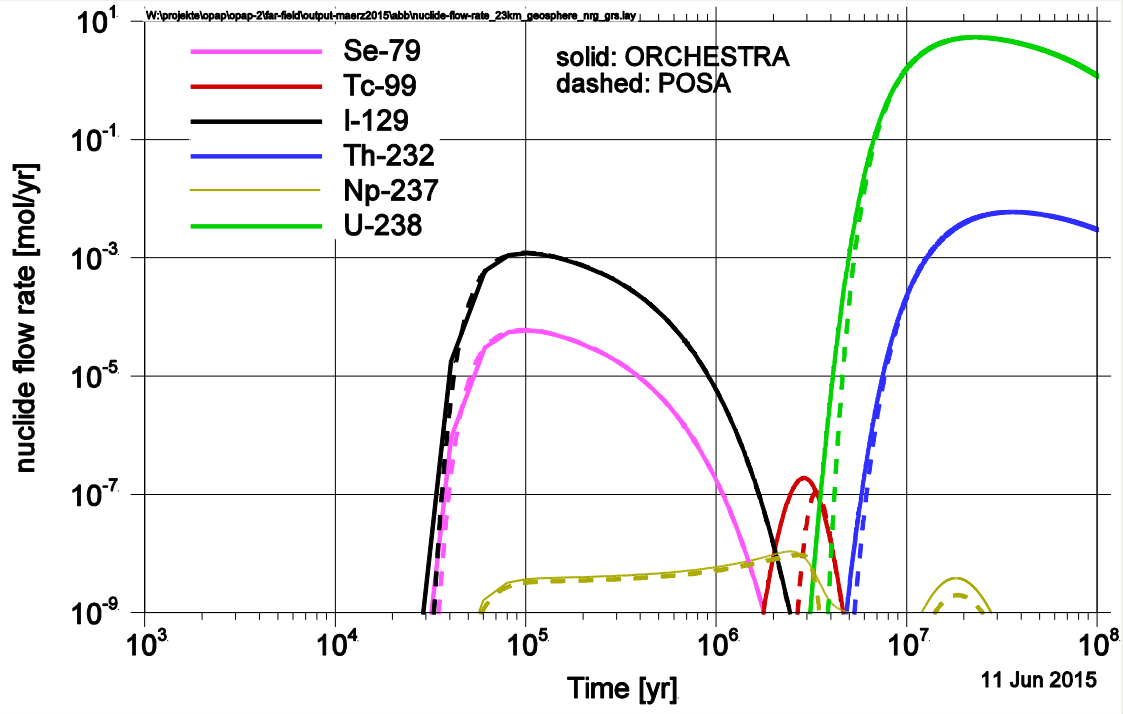

Figure 4-2 Time dependent ORCHESTRA and POSA calculated nuclide flow rates at the end of the transport path in the aquifer

Table 4-3 Maxima of radionuclide fluxes at end of the transport path in the aquifer

Radionuclide	*ORCHESTRA*		*POSA*	
	Maximum flux [mol/yr]	*Time points of maximum flux [yr]*	*Maximum flux [mol/yr]*	*Time points of maximum flux [yr]*
Se-79	6.27E-05	9.99e+04	6.33E-05	9.65E+04
Tc-99	1.99E-07	2.84E+06	1.15E-07	3.31E+06
I-129	1.26E-03	9.99e+04	1.27E-03	9.74E+04
Th-232	6.24E-03	3.55E+07	6.27E-03	3.55E+07
Np-237*	1.08E-08	2.44E+06	9.93E-09	2.69E+06
U-238	5.61E+00	2.25E+07	5.63E+00	2.23E+07

* Total dissolved concentration, no difference is made between the Np-237 from initial inventory and ingrowth

The result of the benchmark is satisfying, because the calculated concentrations and fluxes are very similar in the time dependency and the maxima.

5. Conclusions and recommendations

The overburden (more precisely: the host rock surrounding rock formations) represents the third compartment of the repository system next to the repository (including the waste form) and the host rock. It has been modelled as a porous medium of different layers. The starting point of the PA model for this compartment are the aquifers at the top and bottom of the clay layer. The end point is the interface with the biosphere model.

A detailed discussion of the geological structure and evolution of the Netherlands yields several potential evolutions of a site in Boom Clay. These potential evolutions can be described by scenarios, which are applicable for performance assessments. For each of these scenarios transport times of dissolved species from the top and bottom of the host rock to the surface water have been determined in OPERA-PU-DLT621.

The calculations for the aquifer system have been performed by the computer code ORCHESTRA, which is applicable to all compartments of the system. The benchmark of the computed results has been done by comparison with results of the computer code POSA.

The transport in the aquifer has been modelled by one-dimensional contaminant advective transport including decay and ingrowth and the option of sorption (retardation). Due to the scale of the aquifer system, diffusion has not been taken into account because it is of minor relevance. Dispersion was only taken into account in the POSA model but by comparison with the ORCHESTRA model its effect was shown to be negligible. The retardation along the transport path has been modelled by a linear sorption isotherm (K_d concept). This model can be used also for calculations without retardation: by specifying a K_d equal to zero (R=1) the retardation can be omitted in the calculations. In the calculations the layered structure of the aquifer system in the overburden has been modelled by one aquifer segment with mean characteristics, especially with mean retardation factors.

The test calculations show good agreement of the results produced with the two computer codes. Thus, as in earlier investigations, ORCHESTRA turned out to be an adequate tool. In a test case the transport has been split into two layers with different retardation factors. The expected behaviour could be demonstrated: for single nuclides (activation and decay products) no difference in the resulting concentrations near the interface with the biosphere occurred. For decay chains with different K_d values of the individual nuclides, the resulting concentrations were influenced by the different retardation in the layers.

The selection of safety and performance indicators may be refined. In the present report, the concentrations and fluxes of radionuclides (either in activity or in amount) were sufficient to explain all the results. Further indicators, such as inventory, may be appropriate in future calculations to highlight special effects in the calculations.

6. References

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Appendix 1 Details of the validation calculations

A-1 Definition of the test case

Table A-1 gives the test data for the PA model 'Aquifer'. As a first step, the separation into layers has not been taken into account. In the following, the suffix “aq” of the variables refers to “aquifer”.

Table A-1 Test data for transport in the aquifer as used in ORCHESTRA and POSA calculations. The entire transport path is modelled by a single layer.

Parameter	*Value*		*Dimension*
	NRG	*GRS*	
density of rock	2.6	2.6	kg/l
cross section of path (total)	5,000	5,000	m^2
diffusion coefficient	0	0	m^2/s
volume flow rate *	$2.22 \cdot 10^{-1}$	$2.22 \cdot 10^{-1}$	m^3/yr
length of path	23,000	23,000	m
porosity of layer	0.3	0.3	-
dispersion length	-	50	m
residence time	$3.1 \cdot 10^{4}$	-	yr
effective flow velocity *	$7.42 \cdot 10^{-1}$	$7.42 \cdot 10^{-1}$	m/yr

* effective flow velocity = distance 23,000 m/time 31,000 yr = 0.742 m/yr
volume flow= effective flow velocity 0.742 m/yr * porosity 0.3 = 0.222 m^3/yr

The test values for the distribution coefficient and associated retardation factors are compiled in Table A-2.

Table A-2Test data for the retardation and distribution of some radionuclides

Radionuclide	$K_{d_o}K_{d,aq}$ *[m³/kg]*		R_{aq} *[-]*
	NRG	*GRS*	
C, Cl, I, Se, Zr, Pd, Cm	0	0	1
Np	1.65e-02	1.65e-02	101
Pa	2.71e-02	2.71e-02	165
Pu	6.78e-02	6.78e-02	412
Ra	3.30e-03	3.30e-03	21
Tc	1.65e-02	1.65e-02	101
Th	1.65e-02	1.65e-02	101
U	1.65e-02	1.65e-02	101
Am	6.78e-02	6.78e-02	412

with the following meaning of the parameters:

R_{aq} retardation factor in the overburden
$K_{d_o}K_{d,aq}$ distribution coefficient in the overburden

Figure A-1 shows the assumed nuclide flow rates at the Boom clay/overburden interface that was used as input for this test calculation.

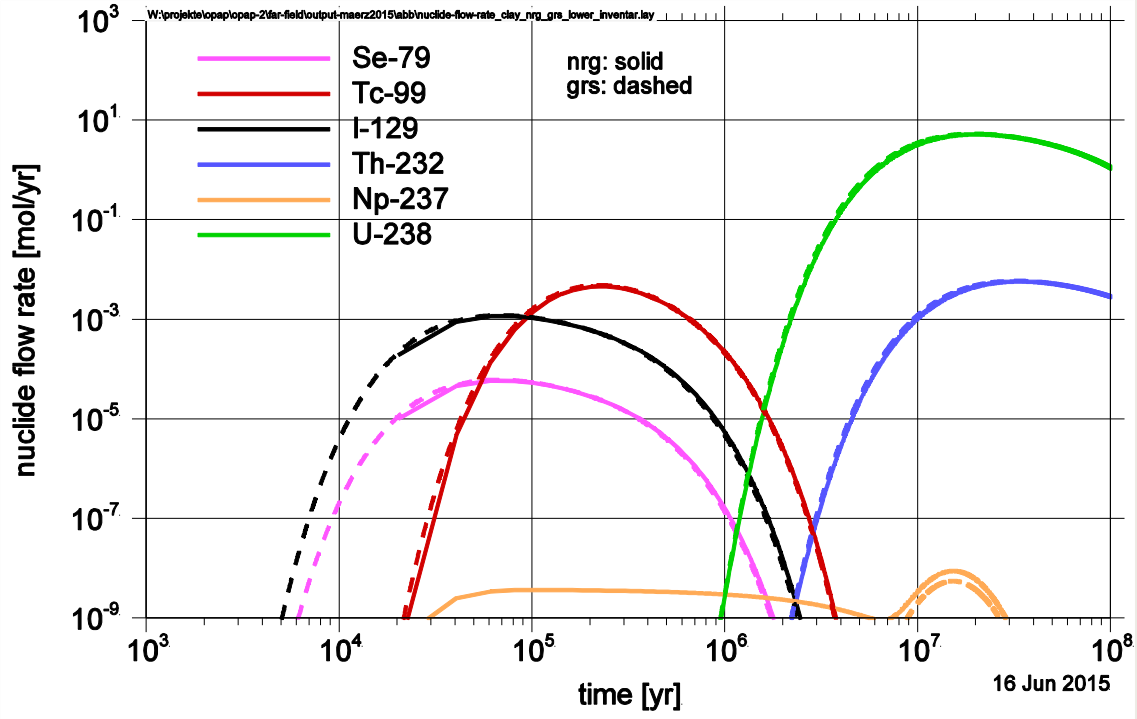

Figure A-1 Nuclide flow rates at the Boom clay/overburden interface as calculated by ORCHESTRA (NRG) and CLAYPOS (GRS)

A-2 Results of POSA calculations

Figure A-2 shows the time dependent molar concentrations calculated with POSA at the inter-section of the Boom clay layer to the overburden for the six radionuclides selected. The position is at 50 m from the beginning of the transport path in the overburden, i.e. the center of the first numerical grid element in the model.

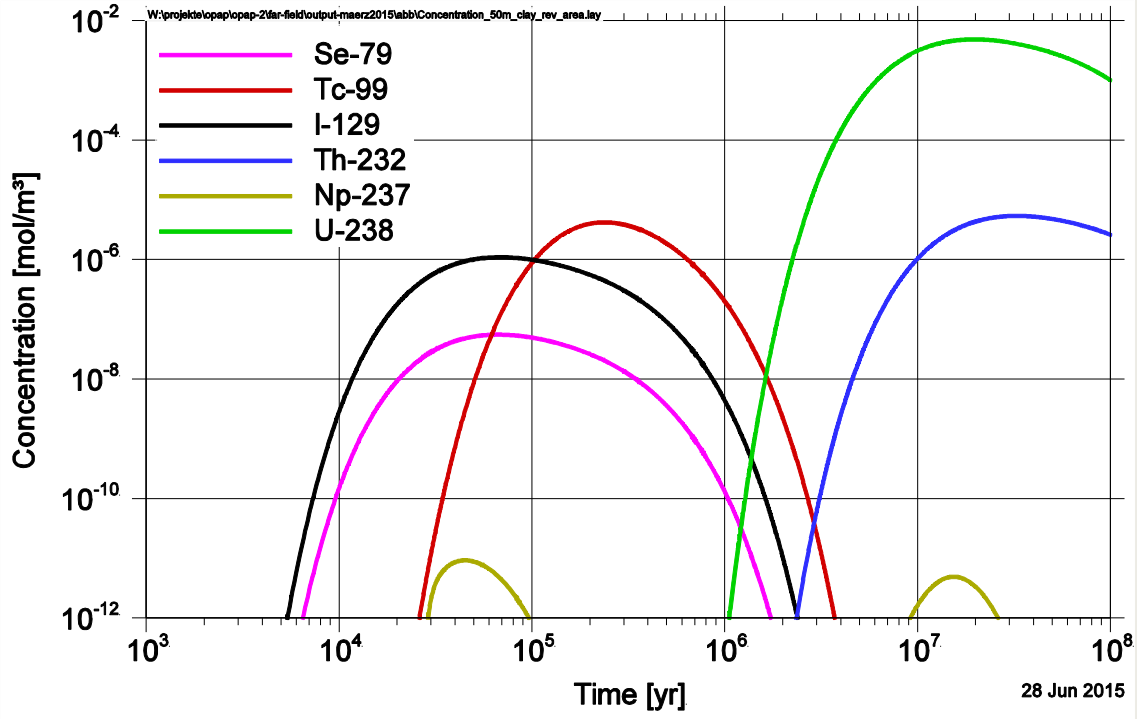

Figure A-2 Time dependent POSA calculated nuclide concentrations in the first numerical grid element of the overburden

The transport of all radionuclides through the overburden is influenced by the flow rate in the aquifer and retardation. Figure A-3 shows the time dependent molar concentrations calculated with POSA at the top of the overburden for the six radionuclides selected.

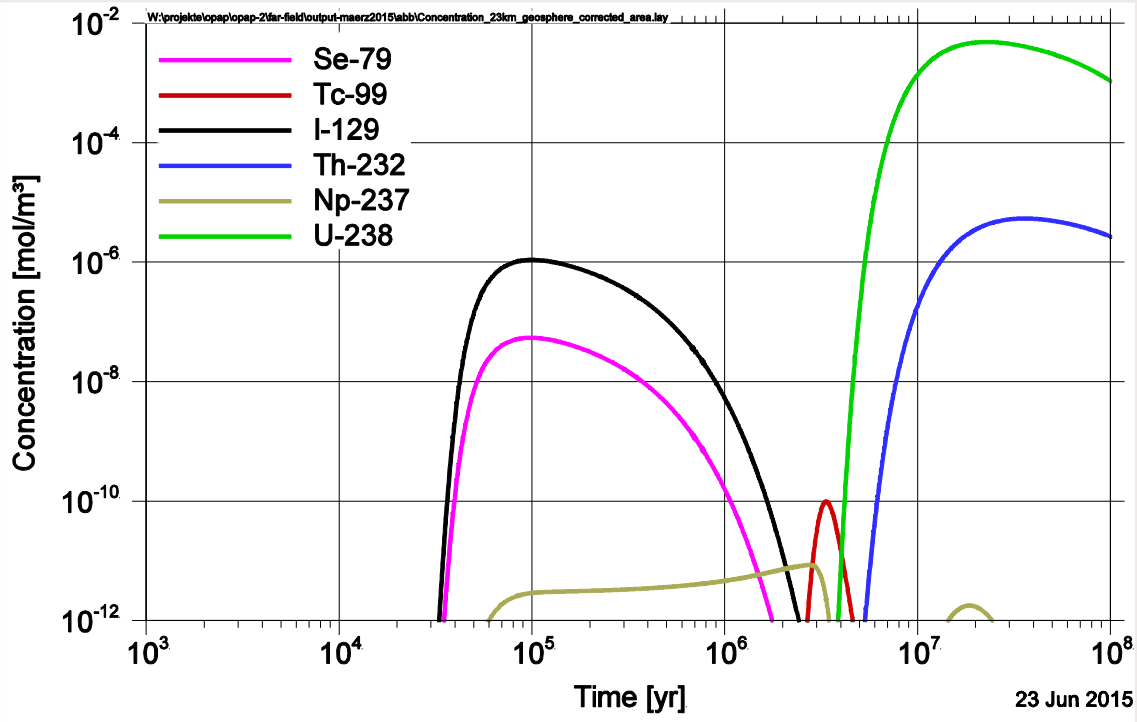

Figure A-3 Time dependent POSA calculated nuclide concentrations at the end of the transport path in the overburden

Compared to the concentrations at the top of the overburden (Figure A-3), for Se-79, I-129, Th-232 and U-238 the maxima of the concentrations slightly decrease within 10^8 years, while for Tc-99 the concentration drops strongly due to radioactive decay. Figure A-4 demonstrates the changes in the concentration curves over time for three selected radionuclides. For Tc-99 the changes are most pronounced.

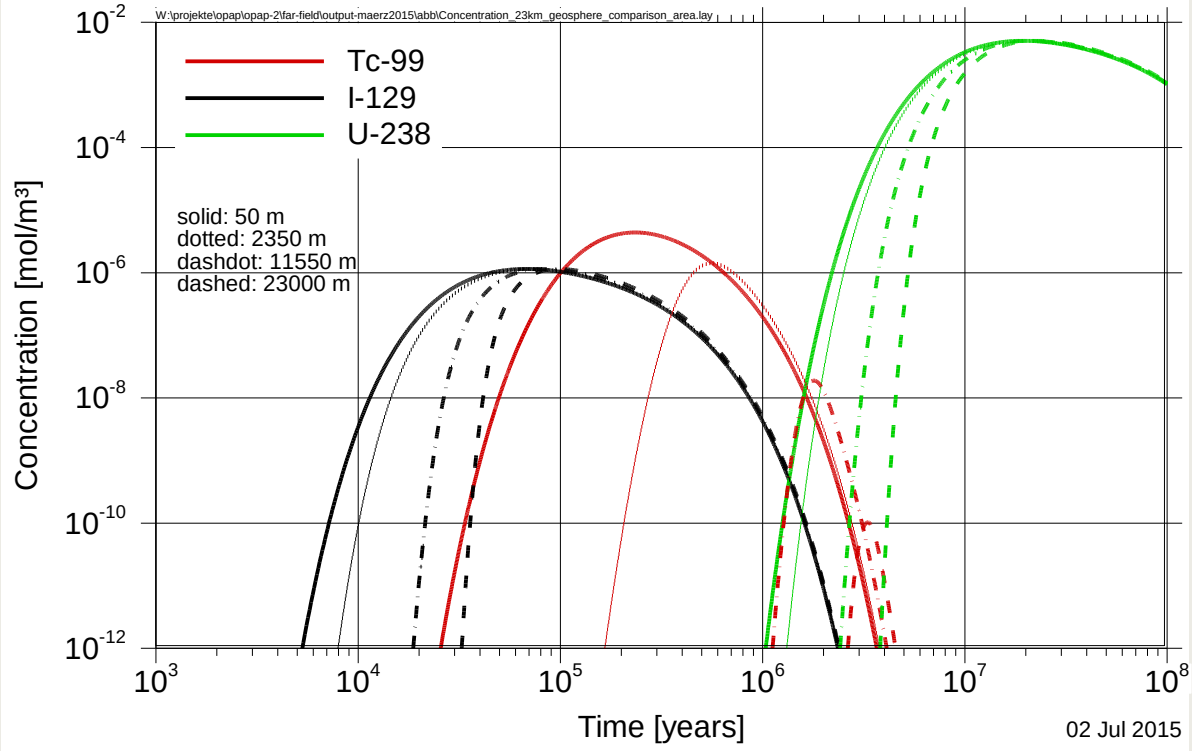

Figure A-4 Temporal evolution of POSA calculated nuclide concentrations along the transport path in the overburden for selected radionuclides

In Figure A-5 the nuclide flow rates from clay to aquifer are compared to the flow rates at the top of the overburden. As already seen in Figure A-4, the curves clearly show the effect of travel time in the overburden for non-sorbing radionuclides (Se-79, I-129) as well as the retardation effect (R_C = 100) for sorbing radionuclides. For Tc-99 additionally the effect of radioactive decay is remarkable.

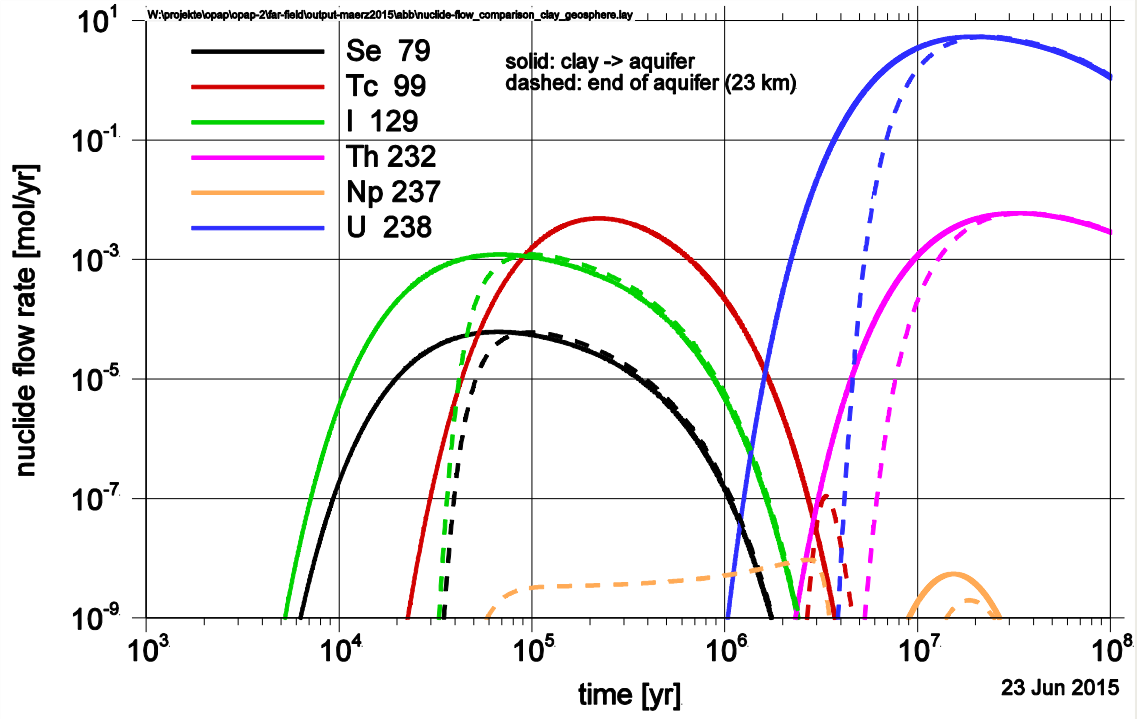

Figure A-5 POSA: Comparison of nuclide flow rates at the Boom clay/overburden interface and at the end of the transport path

A-3 Results of ORCHESTRA calculations

For the presentation of the ORCHESTRA results similar figures as for POSA are shown. As the curves are almost identical to the POSA curves, a detailed discussion of the results is omitted here.

In Figure A-6 the time dependent concentrations at the interface between Boom clay and the overburden are shown for the ORCHESTRA calculation. The position is at 230 m from the beginning of the transport path in the overburden, i.e. the centre of the first numerical grid element in the model. The calculated small increase of the concentration from clay to aquifer is as in the case of POSA (see Figure A-2) due to different cross sections of the flow path, which yield different volumes for dilution.

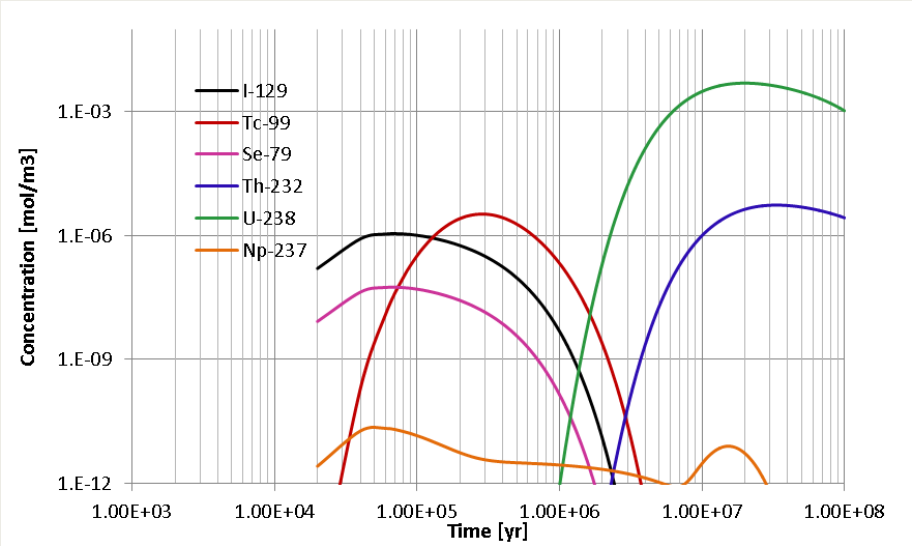

Figure A-6 Time dependent ORCHESTRA calculated nuclide concentrations in the first numerical grid element of the overburden

The time dependent concentrations of the selected radionuclides calculated with ORCHESTRA for the end of the transport path are shown in Figure A-7. The performance indicator *Radiotoxity concentration in the overburden* compartment is obtained by calculating an average activity in the compartment and then converting these activity-based concentrations to radiotoxicity based concentrations.

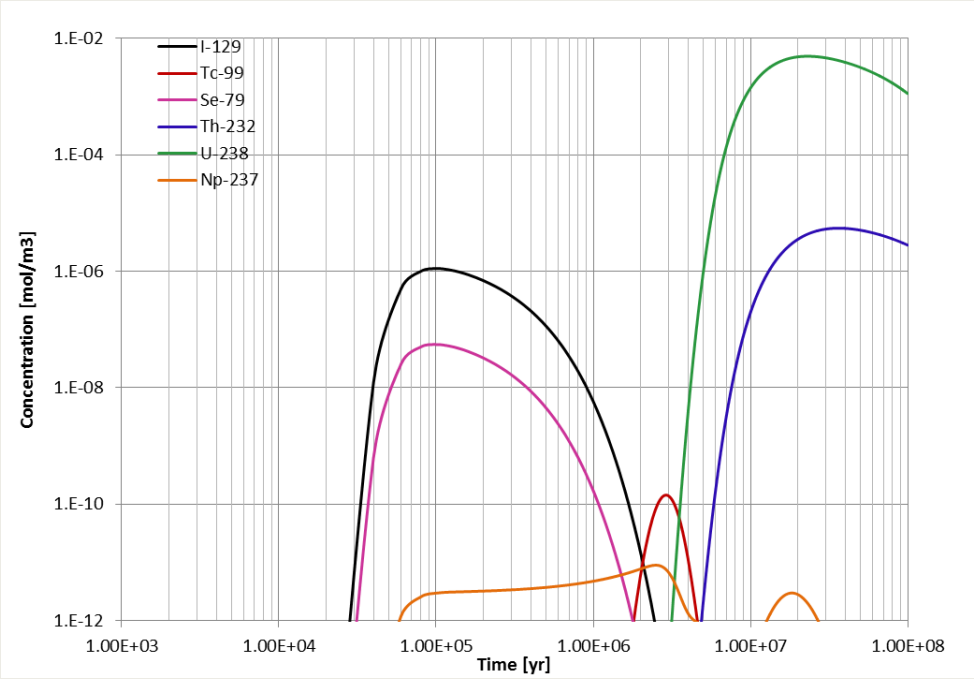

Figure A-7 Time dependent ORCHESTRA calculated nuclide concentrations at the end of the transport path in the overburden

When combined on the same graph, the concentrations at selected positions along the transport path give a good representation of the retardation function of the overburden (see Figure A-8).

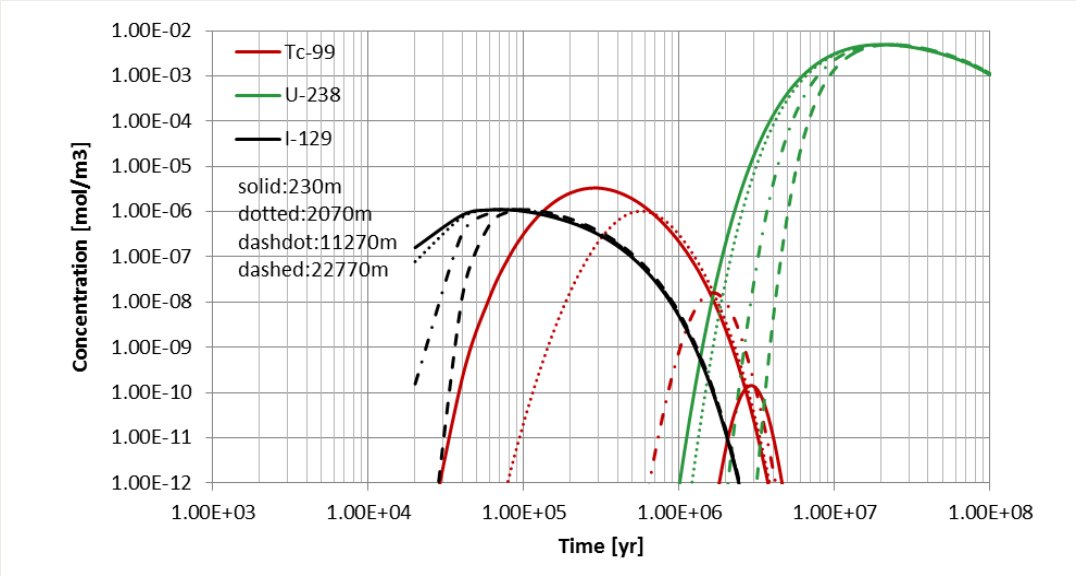

Figure A-8 Temporal evolution of ORCHESTRA calculated nuclide concentrations along the transport path in the overburden for selected radionuclides

The temporal evolution of the radionuclide fluxes across the interface aquifer/biosphere (see Figure A-9) form the base for the rest of indicators related to geosphere compartment.

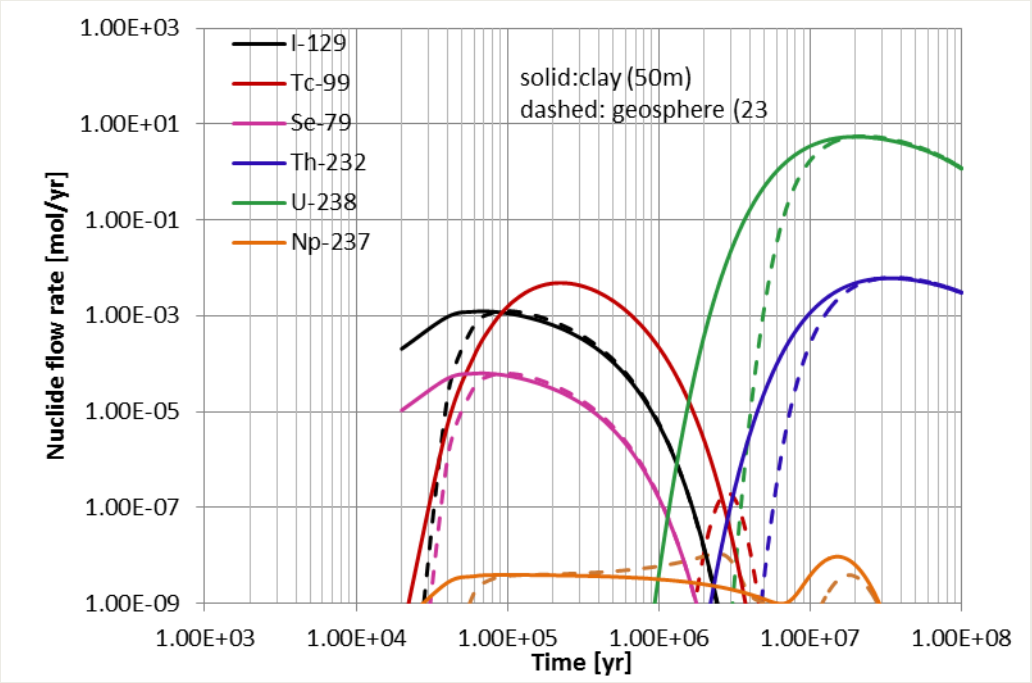

Figure A-9 ORCHESTRA: Comparison of nuclide flow rates at the clay/aquifer interface and at the end of the transport path

When converted to radiotoxicity, these data represent the indicator *Radiotoxicity flux from the compartment overburden* and the integration in time of the radiotoxicity flux represents the indicator *Time-integrated radiotoxicity flux from the compartment overburden*.

The *Retardation due to migration through overburden* can be calculated by dividing the time integrated radiotoxicity flux released from the overburden by the time-integrated (up to time t) radiotoxicity flux released from the waste compartment.

Appendix 2 Advection-dispersion equation

Transport of dissolved species is described mathematically by the advection-dispersion equation. The equation in 1-dimensional form, including decay, ingrowth and linear absorption is:

$$\frac{\partial C}{\partial t} = \frac{GD_w + Lv_{water}}{R}\frac{\partial^2 C}{\partial x^2} - \frac{v_{water}}{R}\frac{\partial C}{\partial x} - \lambda C + \lambda_p \frac{R_p}{R} C_p$$

where:

C	aqueous phase concentration of the solute (mol/m^3),
η	porosity,
v_{water}	effective flow velocity (in the X-direction) of the water in the aquifer (m/s)
L	geometry coefficient accounting for the pore structure driven dispersion due to the water flow through the pores - dispersion length (m)
G	geometry factor accounting for the pore structure (i.e. tortuosity, constrictivity)
D_w	diffusion coefficient of the solute in free water (m^2/s),
R	retention or retardation factor,
λ	radioactive decay constant (s^{-1}),
λ_p	radioactive decay constant of the parent nuclide (s^{-1}),
R_p	retention or retardation factor of the parent nuclide
C_p	aqueous phase concentration of the parent (mol/m^3),

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