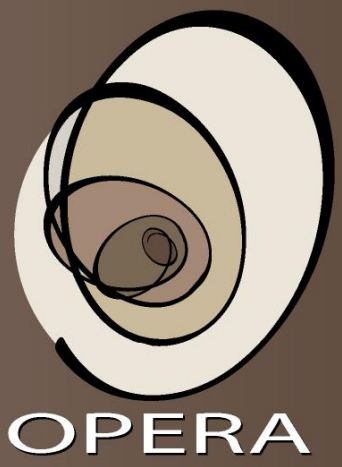

Safety assessment of uranium on very long timescales

OPERA-PU-NRG733/745

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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Summary

In the OPERA research programme, performance assessment calculations were performed for a disposal concept in Boom Clay, covering a period of 10 million years. However, also beyond 10 million years, relevant radiotoxicity concentrations in the biosphere may appear. This would be caused by the very slow release and migration of disposed uranium and its daughter nuclides though the Boom Clay host rock. In this report, additional analyses are performed to address the safety of the OPERA disposal concept in Boom Clay on very long terms, i.e. beyond one million years.

Assessment calculations for periods beyond 1 million years are disputable in radioactive waste management, because these are accompanied by increasing uncertainties on human behaviour, geological processes and other phenomena. In this report it is explored how uncertainties on the long term evolution of the disposal and its surrounding environment can be addressed. In addition, the outcomes of performance assessment calculations are put into perspective, given the fact that the second exposure peak occurs in a very distant future, far beyond human imagination.

The outcomes of additional analyses show that radiotoxicity concentrations in the biosphere on the very long term are dominantly related to ^{238}U and its daughter nuclides. Because uranium is a natural abundant element, the radiological consequence of the presence of any additional uranium and its daughters can be compared with natural background concentrations in the Netherlands and with studies on natural analogues. A number of lessons learned and conclusions are presented.

Samenvatting

Als onderdeel van het onderzoeksprogramma OPERA zijn lange-termijn veiligheids-berekeningen uitgevoerd voor een eindbergingsconcept in Boomse Klei over een periode van 10 miljoen jaar. Aanvullende analyses lieten zien dat ook na 10 miljoen jaar relevante radiotoxiciteitsconcentraties kunnen optreden in de biosfeer, veroorzaakt door de zeer langzame migratie van uranium en zijn dochternucliden door de Boomse Klei. In dit report zijn additionele analyses uitgevoerd om de veiligheid van het OPERA bergingsconcept op zeer lange termijn, i.e. na meer dan een miljoen jaar in kaart te brengen.

Veiligheidsberekeningen over periodes van meer dan een miljoen jaar zijn in de eindbergingswereld omstreden, omdat er naarmate de tijd vordert steeds grotere onzekerheden optreden zoals bijvoorbeeld over het menselijk gedrag of geologische processen. In dit report is gekeken hoe deze onzekerheid over de langetermijn ontwikkeling van de berging en het omliggende milieu geadresseerd kan worden. Daarnaast is nagegaan hoe de uitkomsten van berekeningen over zeer lange perioden in het juiste perspectief kunnen worden geplaatst gezien de radiologische gevolgen van uranium in de eindberging slechts op zeer lange termijn van belang zullen zijn, ver buiten het menselijke voorstellingsvermogen.

De resultaten van additionele analysen laten zien dat radiotoxiciteitsconcentraties in de biosfeer op zeer lange termijn vooral gerelateerd zijn aan ^{238}U en zijn dochternucliden. Omdat uranium een natuurlijk in het milieu voorkomend element is, kunnen de radiologische consequenties van de aanwezigheid van additionele hoeveelheden uranium worden vergeleken met natuurlijke achtergrond concentraties in Nederland, en met resultaten van studies naar natuurlijke analogons. Het rapport bevat tevens een aantal conclusies en “lessons learned”.

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 these proposals, research was proposed for the tasks described in the OPERA Research Plan [2].

In the OPERA research programme, all safety relevant aspects of a given generic reference disposal concept for radioactive waste [1] are considered and assessed in order to evaluate the long-term safety of such a facility [2]. The programme follows in general terms the methodology known as *'Safety Case'* [3, 4, 5]. The central part of the Safety Case is formed by safety assessment calculations that investigate potential radiological risks of a disposal concept. In OPERA, safety assessment are performed over a period of 10 million year, however, a second peak related to disposed depleted uranium can appear in the biosphere far beyond this period.

1.2. Objectives

As is generally agreed in radioactive waste management, uncertainty on the long-term safety assessment of a disposal facility increases in the long-term, making safety assessment calculations after 1 million years disputable. At the OPERA Safety Case Group meeting of 21 of April, 2017, the participants were very aware that communication of the calculated long-term radiological impact of disposed uranium is not straightforward, and there was agreement that there is some need to address these long-living radionuclides in a different way than the radionuclides that contribute to the earlier peak of the effective dose rate in the biosphere.

This report aims to provide additional arguments for understanding and interpreting the potential hazard related to uranium and its daughters on the *very long term* - which, in this report, refers to the period beyond 1 million years after disposal - by use of independent evidence [6], and comparison with 'natural analogues' [7, 8, 9]. The main objective of this endeavour is to provide additional considerations for argumenting safety in the very long-term, consistent with, and in support of the OPERA safety assessment results and underlying OPERA outcomes.

1.3. Realization

This report has been compiled by NRG.

1.4. Explanation contents

Chapter 2 elaborates why it is necessary to look beyond the period of one million years. Chapter 3 analyses how risks can be assessed on geological time scales, and explains why analyses on the very long term can be limited to ^{238}U. It also gives a brief overview on the uranium waste inventory of OPERA, and summarizes natural background concentrations of U in Boom Clay and in the biosphere. In the final section of Chapter 3, observations on natural analogue studies on uranium are presented, and comparisons with the OPERA disposal system are made. Chapter 4 explains how uncertainties in the modelling of uranium migration are addressed in the OPERA safety assessment, and which additional uncertainties need to be considered when looking at very long timescales. In Chapter 5, a synthesis of the analyses in this report is given and conclusions are formulated.

Additional material in support of the main text can be found in a number of appendices.

2 Why addressing timescales beyond 1 million years?

The long-term safety of a geological disposal facility is a key topic in radioactive waste management. An important principle of radioactive waste management with a broad support base is that a similar level of protection should be provided for future generations as for the current generation (e.g. [10, p.7]). It is recognized that safety assessments should be performed sufficiently far into the future to ensure that any peak in the potential radiological impact originating from the disposal facility has been taken into account. On the other hand, very long timescales are accompanied by large uncertainties, making assessment calculations for periods beyond 1 million years disputable.

The safety assessment calculations performed in OPERA cover a period of 10 million years. Within this assessment period, a peak occurs at about 220.000 years, from the contribution of radionuclides that migrating relatively fast through the Boom Clay host rock. Thereafter the calculated risk indicators decrease by many orders of magnitude [23].

During the first 10 million years no contribution of uranium and actinides to the overall risk is visible, due to (1) the limited solubility of uranium, and (2) the very slow migration of these nuclides through the Boom Clay host rock. However, when looking to timeframes beyond 10 million years, uranium and its daughter nuclides cause a second peak of relevant magnitude (Figure 2-1). While doubts on the usefulness of model calculation over such long times scales are justified, a long-term safety assessment limited to 1 or 10 million years could be argued as unsatisfying because it does not tell the 'whole story'.

This raises two major questions:

1. What are the relevant uncertainties on the very long term (i.e. beyond one million years), and how can these be addressed?
2. How can the outcomes of a safety assessment be put into perspective, given the fact that the second peak occurs in a very distant future, on a geological timescale far beyond human imagination?

These questions will be addressed in the next chapters.

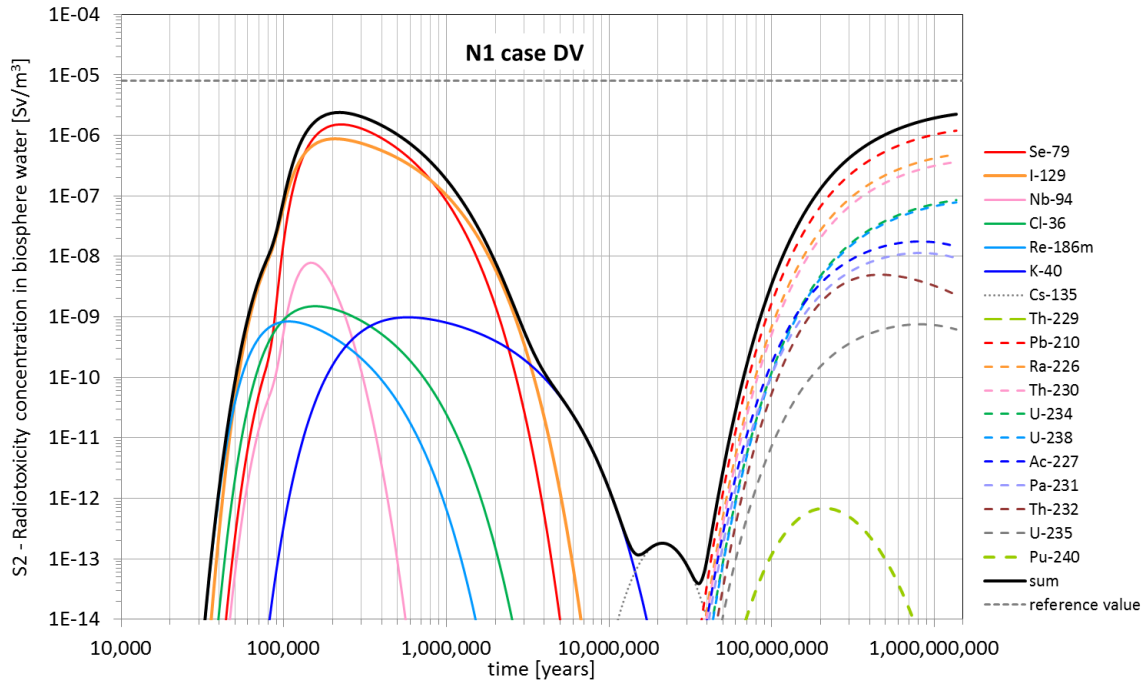

Figure 2-1: Radiotoxicity concentration in biosphere water in the central assessment case (N1-DV), computed over 1.5 billion years. PA-model 9.3-multiwaste.

3 Assessment of uranium on the very long term

It can be generally noted that the uncertainty about the long-term evolution of a disposal system and its natural environment is higher in the biosphere than in the subsurface for the following considerations:

- In the *biosphere*, assessment calculations apply assumptions on presently existing human and animal diets, land use, agricultural practices, as well as water management aspects in order to estimate the future human exposure [11]. However, diets and agricultural practices can already change within decades.
- The subsurface between the biosphere and the host rock, denoted as '*overburden*', can be assumed to be more stable than the biosphere, although water management measures may affect local groundwater streamlines in the upper part of the overburden in a relatively short term. Glacial periods that occur in intervals of few hundred thousands of years affect this layer more profoundly [12]. Within OPERA little quantitative information is provided that allows to integrate the effects of future glacial periods on the overburden in a safety assessment model, and more detailed analyses of the geological evolution in [12] are limited to one million years. It can, however, be noted that the overburden representation in the OPERA performance assessment model is very conservative: the travel time through the system is irrelevant with respect to the considered time scale, and little dilution takes place by dispersion.
- The *host rock* is assumed to be most stable feature of the disposal system, although some attention needs to be given to potential future human activities in the deep underground (e.g. exploration of gas, oil, and shales, geothermal energy, CO_2-disposal). The host rock is affected by glacial periods in a relatively predictable manner that can be covered by performance assessment [12, 13]. Major changes of the host rock can occur on very long times scales, caused by regional and continental processes such as erosion, sedimentation, tectonic plate movements, and related uplifts or depressions. However, the analyses performed in [12] cover 'only' a period of one million years, and allows no conclusion on the time scales beyond 100 million years.

To address the above identified uncertainties on the very long term, it is recommended to use more than one indicator for the long-term safety in a safety case for the geological disposal of radioactive waste, with each indicator addressing different timescales. This is briefly elaborated in the next section.

3.1. Suitable assessment endpoints for the very long term

The outcomes of safety assessments calculations are generally expressed as so-called '*safety and performance indicators*' [14, 15, 16], which provide an indication of the performance of components of the disposal system, such as the engineered barriers and the host rock, as well as the safety achieved by the overall system.

The '*Effective dose rate*' has been applied widely in radiological safety analyses and can be considered as the leading, *primary safety indicator* [17]. To cover the large uncertainties on the very long term, it is recommended to calculate next to this primary safety indicator additional, *complementary* safety indicators as assessment endpoints on the very long term [14, p.92*ff*]. Based on international recommendations [18, 19, 20], in OPERA, two additional safety indicators were suggested for the OPERA safety case [15, 16]:

- *Radiotoxicity concentration in biosphere water*
- *Radiotoxicity flux from geosphere*

The first additional indicator does not depend on assumptions on the biosphere that are uncertain already before the first peak in the biosphere occurs (a few hundred thousands of years). The *Radiotoxicity concentration in biosphere water* excludes the biosphere and all processes within by focussing on the groundwater that just entered the biosphere.

The second indicator, *Radiotoxicity flux from geosphere,* uses fewer assumptions on the overburden with respect to its dilution in the interface to the biosphere, by focussing on the radionuclide flux out of the Boom Clay host rock: this makes it the most favourable indicator on the very long term. However, this latter indicator suffers from the difficulty to provide a reliable reference value that can be used as yardstick to judge whether an assessment outcome can be considered safe [21].

The safety indicator *Radiotoxicity concentration in biosphere water* represents the radiotoxicity of radionuclides in 1 m^3 of biosphere water, in case it is ingested by drinking, and is calculated by

$$\textit{Radiotox. conc. } [Sv/m^3] = \sum_{\textit{all nuclides}} c_n e(50)_n \qquad \text{Equation 1}$$

with c_n the *activity concentration* in [Bq/m^3] of radionuclide *n* in the biosphere water and $e(50)_n$ the *ingestion dose coefficient* [Sv per Bq intake] for adults, based on generally accepted conversion values [22].

The *Radiotoxicity concentration in biosphere water* provides comparable results as the *Effective dose rat*e in the OPERA PA [23], supporting the suitability of the indicator. With this indicator being far more robust on the long term, in the remainder of this document the *Radiotoxicity concentration in biosphere water* will be used as primary assessment endpoint.

In OPERA only the safety indicators *Effective dose rat*e and *Radiotoxicity concentration in biosphere water* were calculated [23]. Nevertheless, the *Radiotoxicity flux from geosphere* can be considered a useful *performance indicator* on the very long term, when applied to compare different scenarios, calculation cases, etc. rather than judging the overall safety of the disposal system by comparison with a reference value.

3.2.'Yardsticks' for the judgement of safety

As shortly noted above, every safety indicator requires a reference value that can serve as 'yardstick' to judge whether a disposal system can be assumed sufficiently 'safe'. In [21], a reference values for the complementary safety indicator *Radiotoxicity concentration in biosphere water* was derived: the recommended reference value is 8 μSv/m^3.

In addition to the above-mentioned radiological indicators, alternative risk indicators may serve as comparison for assessment outcomes with everyday risks or known background concentrations. Figure 3-1 provides an overview of estimators for common risks, natural background concentrations, and exposures. These indicators are subdivided in four categories: incidence rates[1], uranium concentrations, radiotoxicity concentrations, and dose rates. The scales of the four categories are arranged in a way that allows comparing the different categories horizontally: all values at the same height in the figure are comparable when assuming a) an annual water consumption of 1000 l with the indicated

[1] The *incidence rate* is the number of new cases per population at risk in a given time period

uranium concentrations, and b) equilibrium of *U* with its daughter nuclides, as will be discussed in next section.

Figure 3-1 shows e.g. that the reference value of the indicator '*Radiotoxicity concentration in biosphere waste*' is close to the *Annual Average Environmental Quality Standard* (AA-EQS) of uranium [72], and that the natural background radiation exposure in the Netherlands is comparable to the risk of a deadly traffic accident. It can also be seen that the reference value for the radiotoxicity indicator is more conservative than the reference value of the *Effective dose rate*.

A familiar indicator that falls outside the scale of the incidence rate in Figure 3-1 is the population averaged mortality rate: in the Netherlands the rate is about $8.8 \cdot 10^{-3}$ [24].

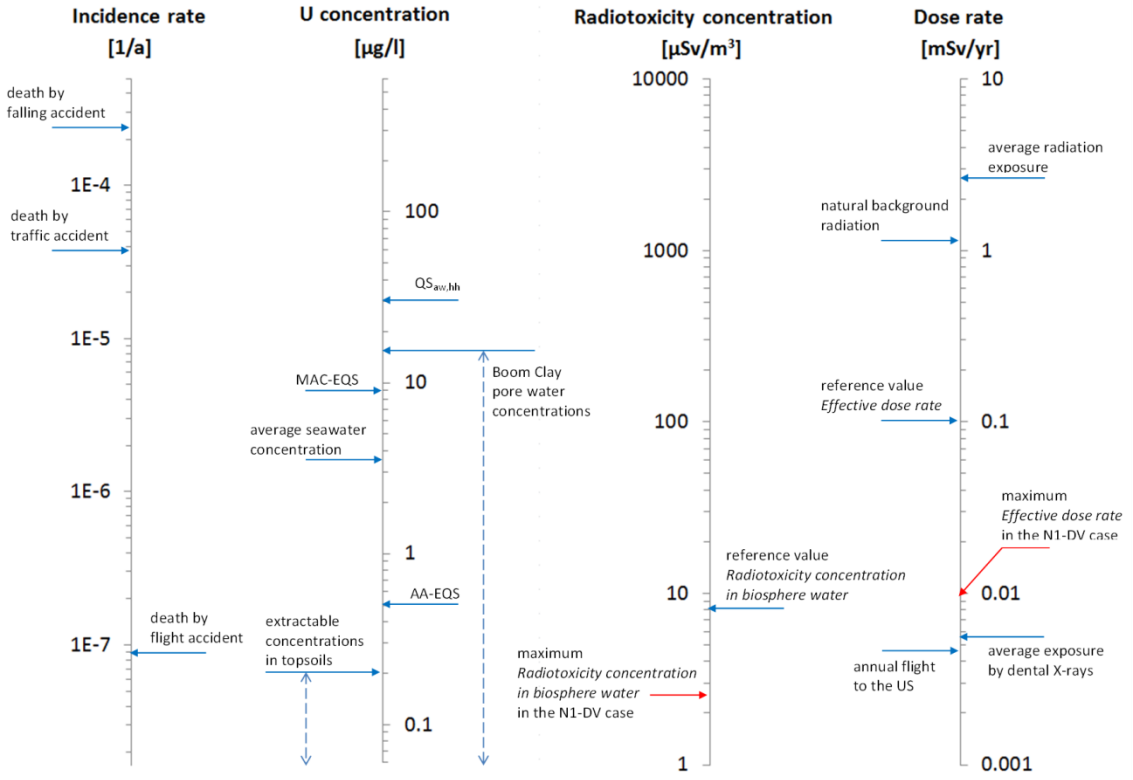

Figure 3-1: Comparison of several risks estimators and reference values in the Netherlands [21, 23, 24, 25, 26, 27, 28, 29, 37, 38, 39, 40, 42, 72,]. *MAC-EQS*: Maximum Acceptable Concentration of Environmental Quality Standard; $QS_{dw,hh}$: Quality standard for surface water; *AA-EQS*: Annual Average of Environmental Quality Standard.

In the next section it is briefly analysed which radionuclides disposed in the facility are relevant for the radiological safety in the long term, and which are not. As will be shown in a subsequent section, this allows simplifying and focusing the safety assessment on the very long term.

3.3.Evaluation of the safety relevant radionuclides on the very long term

For the assessment of the *Normal Evolution Scenario* (NES) of the OPERA disposal concept in Boom Clay on the very long-term, only ^{238}U and its daughter nuclides are of relevance, as will be shown below. The ingrowth of daughter nuclides must be carefully considered, because these can contribute relevantly to the overall risk. Figure 3-2 summarizes the four actinide decay chains, only considering isotopes with half-lives of more than 10 years, as currently implemented in the OPERA PA-model [33].

4N:	$^{248}Cm \rightarrow {}^{244}Pu \searrow$ $^{244}Cm \nearrow$ $^{240}Pu \rightarrow {}^{236}U \rightarrow {}^{232}Th \searrow$ $^{232}U \nearrow$ stable end member
4N+1:	$^{249}Cf \rightarrow {}^{245}Cm \rightarrow {}^{241}Pu \rightarrow {}^{241}Am \rightarrow {}^{237}Np \rightarrow {}^{233}U \rightarrow {}^{229}Th \rightarrow$ stable end member
4N+2:	$^{246}Cm \rightarrow {}^{242}Pu \rightarrow {}^{238}U \searrow$ $\uparrow$ $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra \rightarrow {}^{210}Pb \rightarrow$ stable end member $^{242m}Am \rightarrow {}^{238}Pu \nearrow$
4N+3:	$^{247}Cm \rightarrow {}^{243}Am \searrow$ $\uparrow$ $^{239}Pu \rightarrow {}^{235}U \rightarrow {}^{231}Pa \rightarrow {}^{227}Ac \rightarrow$ stable end member $^{243}Cm \nearrow$

Figure 3-2: Simplified actinide decay chains [33]

In the Normal Evolution Scenario (NES), the leading element determining the radiological risks in very long term is uranium. On the long term, ^{238}U and its daughter nuclides ^{234}U, ^{230}Th, ^{226}Ra, and ^{210}Pb become the most relevant nuclides. After 1 million years these nuclides are in equilibrium with ^{238}U (Figure 3-3, see also Appendix A).

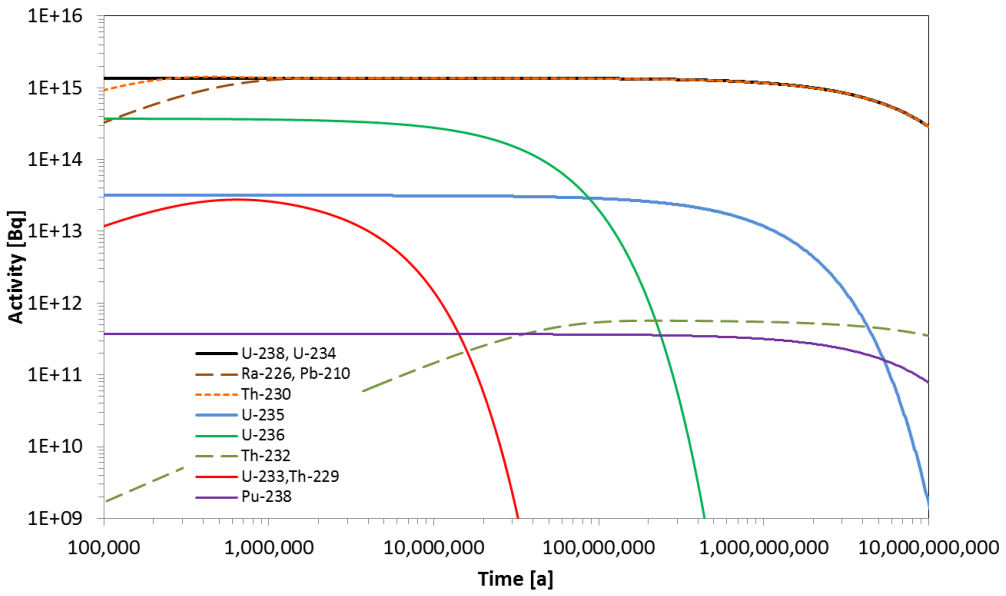

Figure 3-3: OPERA activity inventory of uranium isotopes and other nuclides of the natural nuclide chains relevant after one million years.

Despite that uranium and its daughters are naturally abundant in our environment, it has to be considered toxic, with e.g. uranium mining tailings a relevant point of concern [30; p.291]. To quantify the radiological risk related to disposed uranium beyond 1 million years, it is for the system under consideration sufficient to address the ^{238}U-nuclide chain only: nuclides of the other decay chains contribute at most only a few percent to the total radiological risk. This allows to simplify the long-term safety evaluation of the OPERA disposal concept considerably and avoids the necessity to discuss and understand the complex interrelations of the decay chains indicated in Figure 3-2 and Figure 3-3. Besides, it increases also the computational efficiency of the OPERA performance assessment-model.

All risk analyses can be limited to ^{238}U, when a correction factor is applied for the contribution of the daughter nuclides ^{234}U, ^{230}Th, ^{226}Ra, and ^{210}Pb to the overall risk. This correction factor can be calculated as the ratio of the overall radiotoxicity of all nuclides, and the radiotoxicity of ^{238}U only.

Figure 3-4 shows the evolution of the total radiotoxicity of the uranium disposed in the OPERA facility, the radiotoxicity inventory of ^{238}U only, and the resulting correction factor.

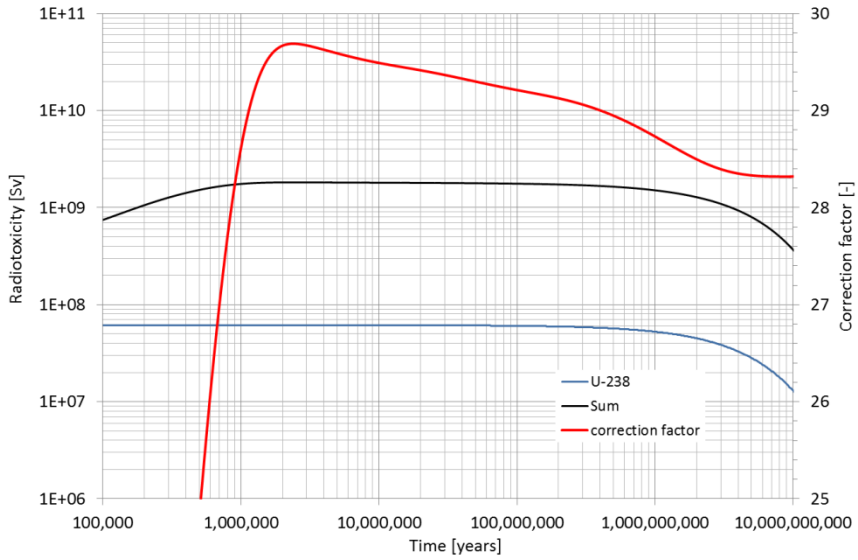

Figure 3-4: Temporal evolution of the radiotoxicity inventory of the OPERA disposal concept, for ^{238}U only and the sum of all natural nuclides chains. The red line indicates the radiotoxicity ratio of ^{238}U and the sum of all nuclide chains ('correction factor').

The correction factor varies after 1 million years only little, a constant values of 29 is assumed sufficient accurate for assessing the radiological impact of disposed uranium on very long time frames. The computation of the *radiotoxicity concentration in biosphere water* (Equation 1) related to the presence of uranium and its daughters can then be simplified to

$$\textit{Radiotox. conc. } [Sv/m^3] = \mathbf{29} \cdot \boldsymbol{c}_{\boldsymbol{238_U}} \cdot \boldsymbol{e}(\mathbf{50})_{\boldsymbol{238_U}} \qquad \text{Equation 2}$$

Focussing the very long term safety assessment of the OPERA disposal concept to a single, natural occurring radionuclide facilitates the understanding of the general system behaviour. It allows also to compare the system behaviour as represented by the OPERA performance assessment model with observations on natural uranium abundance and its behaviour in natural systems (Section 3.5 and 3.6): the radiotoxicity concentrations can

easily be converted to soluble uranium concentrations. This allows a straightforward comparison with measured uranium concentrations in natural systems, as reported in literature: the reference value of 8 μSv/m^3 of the radiotoxicity indicator is equivalent to 14.3 μg/l of uranium without ingrowth, and 0.5 μg/l when accounting for the ingrowth of daughter nuclides (i.e. a factor of 29 lower).

To get a better understanding on the additional uranium concentrations in the environment due to waste disposal, the next two sections elaborate shortly on the uranium inventory of the OPERA disposal concept and provide a brief overview on natural amounts and concentrations in the biosphere and in Boom Clay: this allows to bring the amounts of uranium to be disposed of, and the resulting mobile concentrations into perspective.

3.4.Uranium inventory in the waste disposal facility

In the previous section it was shown that by application of a radiotoxicity correction factor, the assessment on the very long-term can be limited to uranium only. The total anticipated uranium inventory of the OPERA disposal concept is about 110'000 metric tons. 99.6% of the uranium consists of depleted uranium mainly from the uranium enrichment facility Urenco ([31], Table A-1). The tails resulting from this process contain mostly (~99,5%) ^{238}U and smaller fractions of ^{235}U and ^{234}U, and are stored as U_3O_8. The other isotopes of uranium are - as shown in the previous section - of minor relevance[2].

Table 3-1 summarizes the uranium inventory of the OPERA disposal concept in terms of activity, molar and mass amounts. The total amounts of uranium are considerable, taking into account the small nuclear programme of the Netherlands: the equivalent amount of depleted uranium due to all reactor operations in the Netherlands is only 4% of the overall amount to be disposed of ([32], Fig. 5-3).

Table 3-1: Uranium inventory of the OPERA disposal concept (adapted from [33])

Nuclide	Half-life [a]	Activity [Bq]	Amount [mol]	Amount [kg]
^{232}U	$7.0 \cdot 10^{1}$	$9.3 \cdot 10^{12}$	$4.9 \cdot 10^{-2}$	$1.1 \cdot 10^{2}$
^{233}U	$1.6 \cdot 10^{5}$	$2.1 \cdot 10^{9}$	$2.5 \cdot 10^{-2}$	$5.9 \cdot 10^{3}$
^{234}U	$2.5 \cdot 10^{5}$	$1.6 \cdot 10^{15}$	$2.9 \cdot 10^{4}$	$6.8 \cdot 10^{3}$
^{235}U	$7.0 \cdot 10^{8}$	$3.2 \cdot 10^{13}$	$1.7 \cdot 10^{6}$	$4.0 \cdot 10^{5}$
^{236}U	$2.4 \cdot 10^{7}$	$3.7 \cdot 10^{14}$	$6.7 \cdot 10^{5}$	$1.6 \cdot 10^{5}$
^{238}U	$4.5 \cdot 10^{9}$	$1.4 \cdot 10^{15}$	$4.6 \cdot 10^{8}$	$1.1 \cdot 10^{8}$
sum		$3.4 \cdot 10^{15}$	$4.6 \cdot 10^{8}$	$1.1 \cdot 10^{8}$

Because of the very long halve-life of ^{238}U, depleted uranium hardly decays in the long term: less than 0.16% of the emplaced depleted uranium decays within the assessment period of 10 million years. On the other hand, because of the decay of all other disposed radionuclides, the radiotoxicity of the OPERA inventory is dominated after 10'000 years by depleted uranium (Figure 3-5), and only after billions of years, relevant decay of ^{238}U is visible in the graph.

[2] For a complete overview all uranium isotopes and the related waste sections see Appendix A, Table A-1

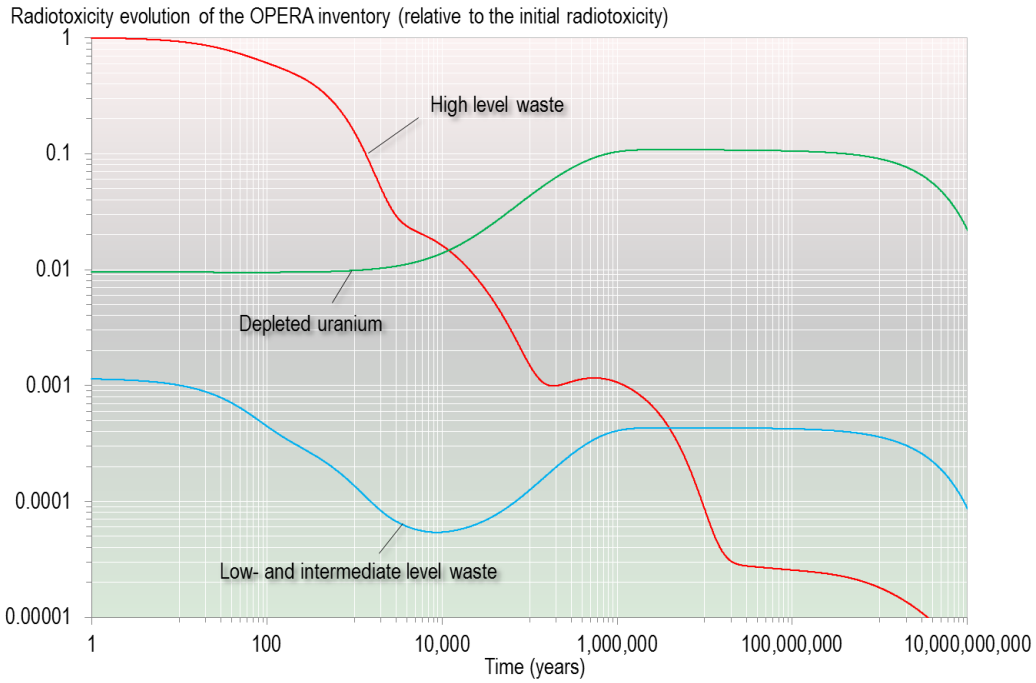

Figure 3-5: Radiotoxicity evolution of high-level waste (HLW), depleted uranium, and low- and intermediate level waste (LILW), relative to the initial radiotoxicity inventory of the OPERA disposal concept.

Due to the low solubility of uranium and its slow migration through the Boom Clay host rock, the disposed depleted uranium is also strongly confined in the disposal facility and its near vicinity: in the OPERA safety assessment calculations of the Normal Evolution Scenario ([23]), only 0.23% of the uranium has migrated out of the disposal facility after the assessment period of 10 million years.

3.5. Natural background concentration of uranium in Boom Clay

In Boom Clay present in the subsurface of the Netherlands, uranium concentrations of about 0.8 - 8.2 mg/kg are measured, with a median value of about 3 mg/kg [34] These values are largely consistent with the uranium concentrations reported for Boom Clay in Belgium, ranging from a few mg to slightly more than 10 mg/kg [35, 36]. To relate these natural abundances of uranium with the amounts present in the OPERA disposal concept: about 180 km^2 of Boom Clay[3] contains a comparable amount of natural uranium as the OPERA inventory (110'000 tons).

Amounts of uranium in the topsoil of the Netherlands depend on the soil type and depth; median values range from 0.6 - 2.4 mg/kg, with an overall range of 0.05 - 10.9 mg/kg [37]. Thus, in general the total amounts of uranium in Boom Clay are roughly three times larger than the mean values found for most soil types in the biosphere in the Netherlands, i.e. there is only a small uranium gradient present between Boom Clay and overlying biosphere soils.

Soluble concentrations of *U* in Boom Clay were not measured in OPERA. For Boom Clay in Belgium, typically concentrations of several tenths of a µg to a few µg per l of pore water are found [62, 38]. Incidentally, high concentrations of 15 µg/l or more are measured as well [39]. Extractable amounts of *U* in the topsoil of the Netherlands as reported in [37] can serve as estimator for soluble concentrations: here, median concentrations range from

[3] assuming a layer thickness of 100 m

0.03 - 0.2 µg/l, with a maximum concentration of 3.5 µg/l. In [40], *U* concentrations up to 3 µg/l in groundwater of the Netherlands are reported. Average concentrations in Dutch river water are about 0.6 µg/l [41]. In [42], average concentrations of 0.02, 3.3 and 0.9 µg/l are reported respectively for rainwater, salt water, and river water of the Netherlands. In conclusion, soluble concentrations of *U* in the biosphere are usually below 1 µg/l, with exception of salt water, and concentrations in Boom Clay are generally higher than in biosphere waters. Figure 3-1 (on p.9) gives an overview on soluble concentrations.

The numbers above provide some global information on total amounts and soluble concentrations of uranium in Boom Clay and the biosphere. However, they are not suitable to estimate uranium mobility for the performance assessment of a geological disposal by using the solid-solution partitioning that can be estimated from the above numbers: it cannot be assumed *a priori* that uranium disposed in the waste facility will be as immobile as currently is observed for uranium present in the Boom Clay since 30 million years. The solubility and geochemical behaviour of uranium will be elaborated further in the next chapter.

In order to get some idea on the level of realism (or conservatism) of the performance assessment model used in OPERA, additional calculations were performed to analyse the effect of natural uranium background concentrations in the host rock on uranium concentrations in the biosphere, and the overall migration of disposed uranium: Figure 3-6 shows that early concentrations of *U* due to natural background concentrations in Boom Clay are more than 100 times smaller in the biosphere than the assumed initial concentration in the Boom Clay. Furthermore, Figure 3-6 shows that the presence of the natural background concentrations of uranium in Boom Clay does not affect the peak concentrations from uranium migrating out of the disposal system into the biosphere.

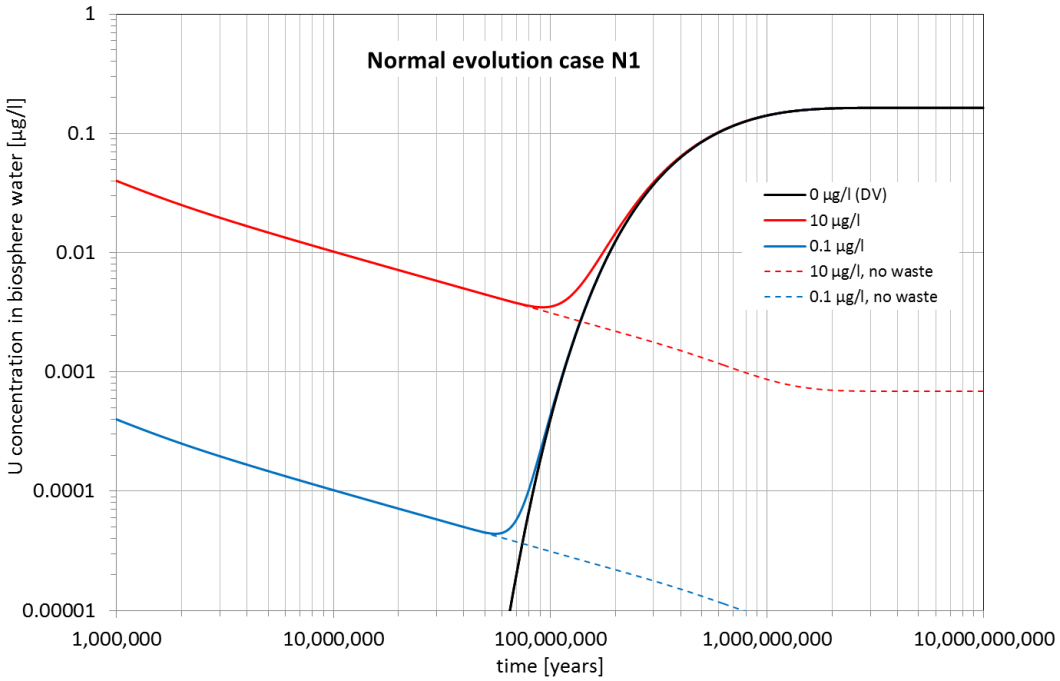

Figure 3-6: ^{238}U concentrations in biosphere water for natural background concentrations of 0, 0.1, and 10 µg/l of uranium in the host rock and with or without OPERA waste disposal.

In the next section, some lessons learned from so-called 'natural analogue studies' on uranium are summarized to gain some insight of uranium mobility in natural subsurface systems.

3.6. Observations from natural analogues studies on uranium

Natural analogue studies have been widely used in the past by organizations such as the IAEA [43], the European Commission's (EC) and Natural Analogue Working Group [44], and NEA's Integration Group for the Safety Case (IGSC [45]). The main reason to consider these studies is to build confidence in the ability of disposal systems to perform over the long term as predicted by safety assessment models. Several detailed and review studies summarize aspects of natural (and anthropogenic) analogues for radioactive waste disposal in deep geological repositories, e.g. [46, 47, 48, 49, 50, 51, 52, 53, 54, 58].

The present report does not intend to summarize and evaluate all these studies. Instead, topics of interest for assessing the contribution of uranium only on very long time scales are highlighted, and conclusions and recommendations about the application of natural analogues in relation to the disposal of significant quantities of uranium are commented.

3.6.1. Cigar Lake uranium deposit

A well-known natural analogue with respect to uranium ore isolation in the deep underground is the Cigar Lake uranium deposit: the Cigar Lake uranium ore body is one of the largest in the world, and is likely the most mentioned and investigated natural analogue of deep geological disposal facilities. Cigar Lake is located at the Athabasca Basin province in northern Saskatchewan, Canada, at a depth of 450 m. The uranium ore is unusually rich in uranium, with an average ore grade of 21% and a maximum of 60%. Although the ore body is located within a rather permeable sandstone, according to ([54], p.171-174) there is no radiological indication of its presence at the surface reported. Figure 3-7 shows a schematic cross section through the Cigar Lake uranium deposit.

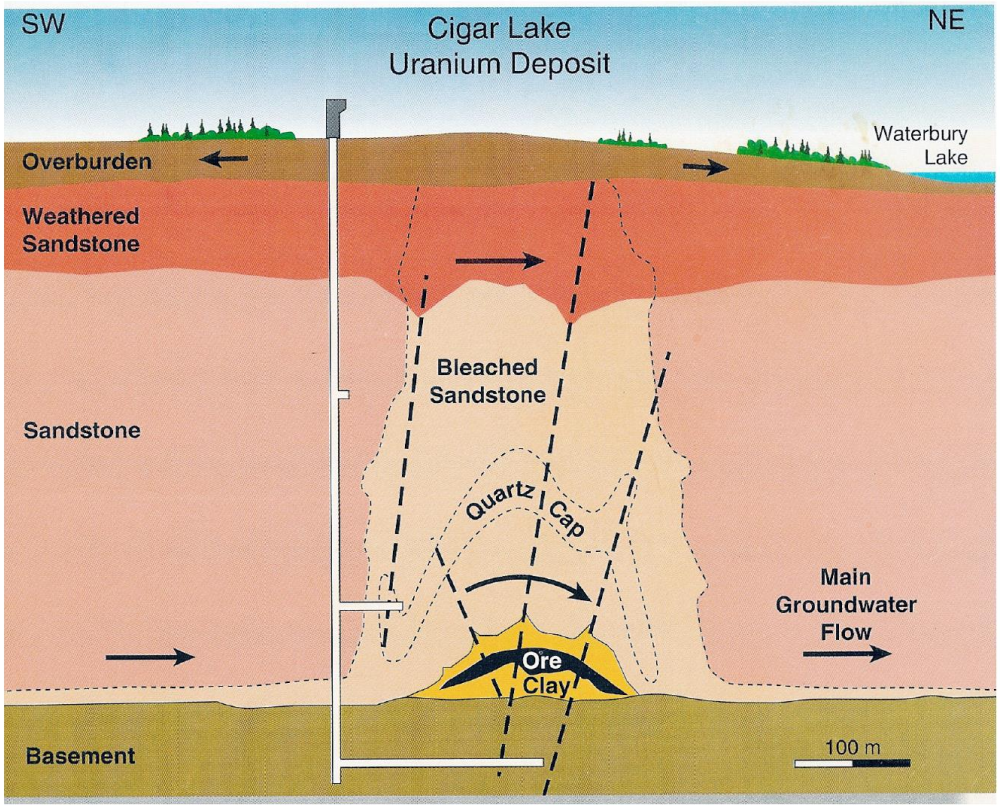

Figure 3-7: Schematic cross section through the Cigar Lake uranium deposit showing major lithological types, extent of the hydrothermal surroundings and groundwater flow pathways (after [55], Figure 3).

Studies on the Cigar Lake site are extensively reported in the literature and, in terms of geological disposal, the key characteristics of this particular site are ([54]; p. 172):

- Qualitative/general evidence of the very long term (about 1,300 million years) chemical stability of a rich ore body, containing high concentrations of *U*;
- Effective containment of radionuclides under present conditions with no significant surface radiological signature of the ore, despite the relatively high permeability of the enclosing host sandstone formation;
- The clay-rich layer surrounding the uranium ore appears to protect the ore from degradation, despite the presence of microbes, dissolved organics and other colloids in the ore pore-waters.

Uncertainties and limitations concerning the utilization of Cigar Lake as a natural analogue are:

- The age of the Cigar Lake ore body (1,300 million years) makes the definition of boundary conditions and its evolution for the greater part of its history difficult. This is an inherent limitation to natural analogues in general. As such, the analogy with a geological disposal facility should not be overestimated;
- Further studies would be constrained by the fact that uranium mining has commenced at Cigar Lake and this will have already considerably perturbed the natural system due to the ongoing excavation by freezing/water-jet boring.

In case of the OPERA geological disposal, several relevant differences between the Cigar Lake and the expected geochemical conditions of Boom Clay in the Netherlands apply:

- Uranium waste is present as U_3O_8 rather than as uraninite and pitchblende in case of the Cigar Lake [55].
- The concentrations of bicarbonates in the Cigar Lake case are much smaller (<100 mg/l [55]) than the values measured in Boom Clay at Mol (±900 mg/l [66]),
- The pH is lower (6 - 8) compared to Boom Clay at Mol (±8.5) or the pH in the disposal sections (>12).

Despite of some notable differences, the general behaviour of the OPERA PA-model representing the OPERA disposal concept is not principally different from the Cigar Lake natural analogue:

- Within the calculation period of the OPERA safety assessment (10 million years), only a minor fraction of the disposed uranium is released into the host rock: about 0.2% for the 'base case' scenario [56]. Extrapolated linearly to a time frame comparable to the age of the Cigar Lake ore, after 1'300 million years still 70% of the disposed uranium will be confined in the disposal facility[4].
- For the fast migration case HR-1 of the OPERA NES [57], the average concentration of uranium in the upper boundary of the Host Rock compartment is less than 2 µg/l within the assessment period, and less than 1 µg/l in the overburden. This is in the order of magnitude of what is measured in groundwater at the Cigar Lake [54, 55].

Thus, on one hand it can be stated that for the OPERA disposal concept the depleted uranium is fairly well confined in the deep underground, even on geological time scales. On the other hand, groundwater concentrations of 1 µg/l of ^{238}U as measured in case of Cigar Lake already represent a relevant, although natural hazard: the concentration is equivalent to 0.6 µSv/m^3 or 7% of the reference value of the radiotoxicity indicator (see Section 3.1), but when accounting for ingrowth of daughter nuclides from the originally

[4] In addition, during that period another 18% will be lost due to natural decay to ^{234}U and other daughter nuclides

present uranium, a radiotoxicity concentration equivalent to 1 µg/l of ^{238}U will exceed the reference value of 0,8 µSv/m^3 (see Section 3.3).

3.6.2. Uranium roll fronts

An example of natural analogue for the transport behaviour of uranium through the overburden is the so-called 'roll-front uranium deposit'. These roll-front deposits, schematically shown in Figure 3-8 ([58], Section 2.1), form in inclined, porous sedimentary subsurfaces, consisting of a reduced zone in front of the ore deposit, and an oxidized zone in its wake.

The boundary between inflowing oxidizing water and the existing reducing environment is called a redox front, where uranium minerals begin to precipitate out of the groundwater and minerals such as uraninite (UO_2, partly U_3O_8) and coffinite can be formed. As result, a local accumulation of uranium can occur, eventually leading to economically mineable uranium concentrations.

Several geologic factors are necessary in order for a uranium ore deposit to form [59]: in the case of roll-front uranium deposits, apart from the presence of uranium itself, a key requirement is the existence of a reducing environment in the groundwater system. Roll-front deposits are also bounded above and below by impermeable rock layers such as shales or mudstones.

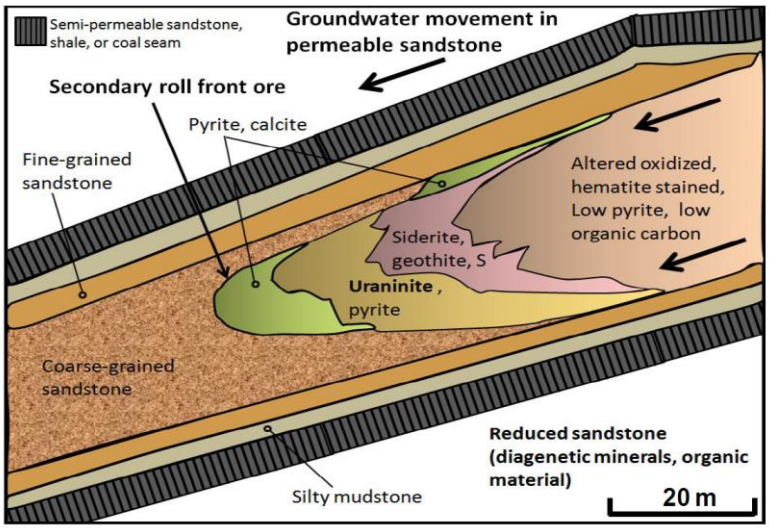

Figure 3-8: Schematic representation of a roll-front uranium deposit [58].

With respect a safety assessment of uranium on the very long term, insufficient information is available in OPERA to quantitatively evaluate the processes described above. In general, localized reduction and oxidation of uranium in the overburden may either decrease or increase radiological risks:

- chemical reduction of uranium generally *decreases* the mobility due to precipitation, but insufficient is known to judge whether reduction can occur in the overburden or Boom Clay in the long term;
- Localized accumulation of uranium may lead to *increased concentrations* in case of changing geochemical conditions, e.g. as result of periodic ice coverage and/or permafrost ('chemical time bomb' [60]).

In conclusion, it is recommended to assess in future safety evaluations the potential formation of uranium accumulations due to the migration of uranium in the overburden.

4 Uncertainties in modelling uranium migration on the very long term

Uncertainties concerning the future safety of a deep geological disposal facility are not only related to its long term evolution, but are also attributed to the conceptual representation of the disposal system and the main migration processes in a safety assessment model in general.

As discussed above, a key uncertainty on the very long term is the regional and continental evolution of the host rock on geological time scales. Concerning that topic, not much can be concluded from the information provided in OPERA. All statements in the remainder of this chapter are therefore based on the principal assumption that in the Normal Evolution Scenario, also in distant future a continuous, subterranean host rock layer of about 100 m thickness will be present ([61]; p.43). However, as indicated in Figure 4-1, an assessment on the time scale of billions of years go beyond regional and continental evolutions of the geosphere: from current understanding on the evolution of the solar system, human life as we know today will be unlikely in about 2 billion years. Increased solar radiation will lead to massive changes of the earth's ecosystem, and after 5 billion years, the Sun is expected to collapse, emitting far more energy than today.

In the next section, the general uncertainties and their handling in the safety assessment are established, with particular focus on a) the effect of these uncertainties on the overall safety on the very long term, and b) potential additional uncertainties related to the assessment on the very long term.

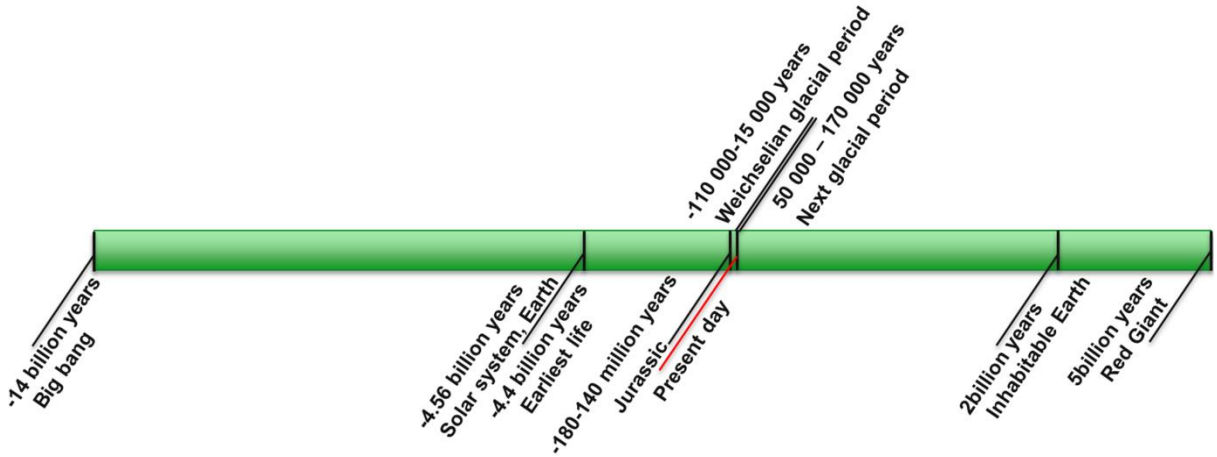

Figure 4-1: Events on geological time scale.

4.1. General geochemical behaviour

Under conditions expected in Boom Clay, uranium is present as (more soluble) *U(VI)* [62]. Although in the given redox range, mixed valence uranium oxides (i.e. U_4O_8 and U_3O_8) might affect the solubility of *U*, it is argued that these species should not be used in geochemical model calculations, since they lead to erroneous results underestimating the real solubility [63]. Therefore, these uranium oxides are conservatively not applied in OPERA for the derivation of the K_d-values in [66] and the solubility of uranium in [65], although some limiting effect on the migration rate is likely.

The solution chemistry of *U* is strongly influenced by bicarbonate concentration in the solution, and relevant amount of bicarbonates are present in Boom Clay in Mol [64, 36]. Soluble uranium can also be strongly complexed with dissolved organic matter (DOC),

which is also present in Boom Clay. The strong binding of *U* to DOC affects the overall amount of *U* present in solution, and can increase it by a factor of 40 [65] or more.

Calculations performed in [66] for a variety of conditions expected in the Netherlands show that the DOC-bound fractions dominate the soluble amounts over a large range of conditions, mainly dependent on the presence of bicarbonates in solution, and the overall concentration of dissolved uranium (see also Section 4.2). In case of high concentrations of dissolved *U*, $UO_2(CO_3)_3^{4-}$ becomes the dominating uranium-species in solution. This is consistent with the general trend in the experimental findings in [36].

The further geochemical analysis is not straightforward. The geochemical model described in [66] assumes equilibrium with amorphous UO_2, equivalent to [64]. This results in comparable small concentrations of non-carbonated uranium-species (median value: $9{\cdot}10^{-11}$ mol/l), with soluble concentrations varying as result of the assumed variation of the redox potential of about 4 *pe* units. However, due to the high affinity of uranyl for carbonate, large fractions of carbonate will be complexed with uranium, resulting in solubilities up to the order of magnitude of the assumed bicarbonate concentration, and higher than the DOC-bound fractions. E.g. assuming a fixed bicarbonate concentration of 900 mg/l and no solubility limitation by mixed valence uranium oxides results in a median overall solubility of $8{\cdot}10^{-4}$ mol/l (10- and 90-percentiles: $9{\cdot}10^{-5}$ and $2{\cdot}10^{-3}$ mol/l, respectively).

The soluble uranium concentrations in the OPERA safety assessment are much higher than measured in the Cigar Lake natural analogue, this is mainly related to the different pH, bicarbonate- and DOC-concentrations. The relevance of the geochemical modelling results are difficult to confirm due to lack of robust experimental data, therefore a best estimate of the overall solubility of U in Boom Clay pore water of $1{\cdot}10^{-4}$ mol/l[5] was suggested to be used in the OPERA safety assessment in [65]. However, large uncertainties with respect to this value are noted as well.

4.2. Solubility limits

From the previous section it is evident that there are relevant uncertainties with respect to the derivation of a solubility limit of uranium under the conditions prevailing in Boom Clay host rock. However, the leading limit for uranium is the solubility in the waste compartment, which is determined to be lower than in the host rock [65]. The actual value of this parameter used for analysing the OPERA NES is $1{\cdot}10^{-5}$ mol/l. A second subcase addressing a lower solubility is defined in [33], Table 3-10, with a uranium solubility of $1{\cdot}10^{-6}$ mol/l. It is evident that the solubility of *U* strongly influences the maximum radiotoxicity concentration in all compartments of the disposal system (Figure 4-2). However, the general uncertainties discussed in the previous section and in [65] apply here as well, and cannot be resolved by geochemical modelling only.

With respect to long term evolution of the uncertainties about the solubility limit of uranium it is reasonable to assume that these will decrease in time rather than increase:

- due to the 'aging' of cementitious materials in the waste section of the disposal facility, the pH is expected to decrease, and as consequence, the solubility of uranium will decrease relevantly (see Figure 4-2 in [65]);
- according to the 'Ostwald phase rule' [67], more soluble mineral phases will appear first, and more stable phases are formed later on. This points to the expectation that experimental determination may overestimate solubilities, and even if the formation of more stable uranium minerals cannot be demonstrated

[5] or 24 mg/l

experimentally, this does not exclude that these will determine the solubility on the long term.

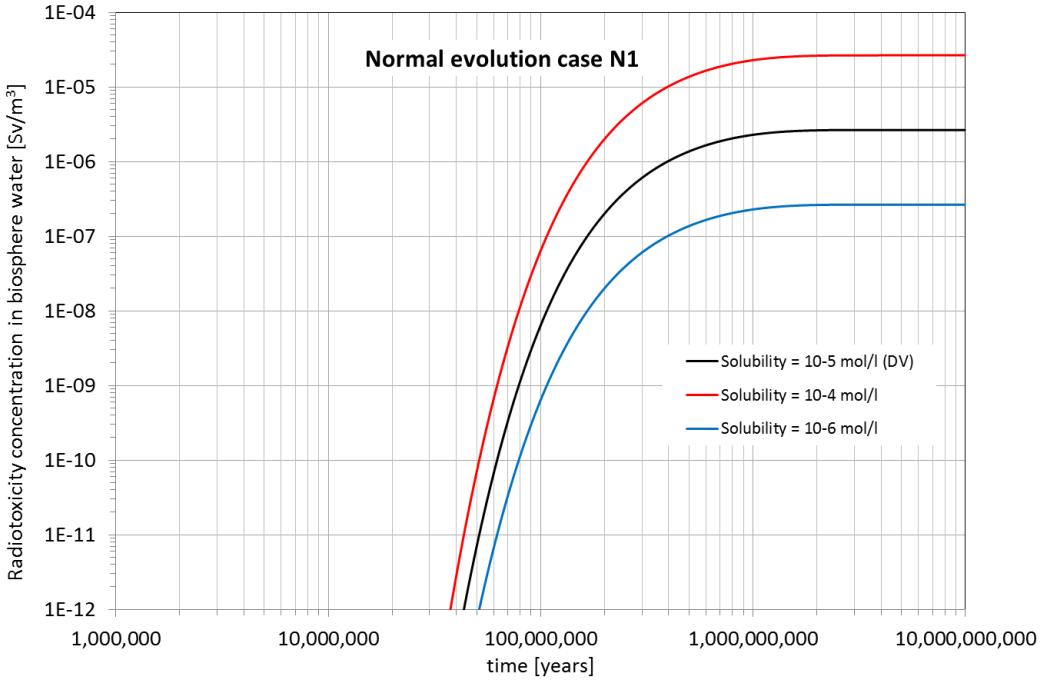

Figure 4-2: Radiotoxicity concentration in biosphere water from ^{238}U and its daughter nuclides for a solubility limit for uranium according to the N1 case DV (black), a 10 times higher (red) and a 10 times lower (blue) solubility limit. Note that the high solubility limit is beyond the parameter values defined for the NES case in [33] (see also [65]).

4.3. Sorption behaviour

The representation of uranium-sorption in Boom Clay by a constant K_d-value was evaluated in [66, 68] for a range of properties of Boom Clay in the Netherlands (see Table F-11 in Appendix F) and implemented in the OPERA performance assessment model [33]. In order to address uncertainties with respect to the concentration of dissolved organic carbon (DOC) in Boom Clay at a specific future location [66], three cases are distinguished:

- a ***base case*** with a DOC concentration of 100 mg/l,
- a ***low DOC* case** with a DOC concentration of 20 mg/l, and
- a ***high DOC case*** with a DOC concentration of 200 mg/l.

Table F-12 in the Appendix summarizes the lower, upper and central K_d-values and retardation factors for the cases considered in the RANMIG project ([66], Tables B1-B3).

The range of Boom Clay properties (Table F-11 in Appendix F) covers a large variation of ionic strengths and different DOC concentrations expected on different locations, and these can be assumed to cover long term variation of Boom Clay properties in an evolving or changing environment as well.

Some remaining uncertainties are acknowledged, however, from general system understanding it can be argued that the K_d-value of ^{238}U influences the time period when uranium enters the biosphere, but has a only a minor effect on the maximum value of the *radiotoxicity concentration in biosphere water* in the NES. This is due to the limited solubility of uranium in the waste (see previous section), which keeps the dissolved

uranium concentration in the disposal compartment constant and at a relatively low value. Radioactive decay has little effect since the concentrations of ^{238}U halves only every 4.5 billion years.

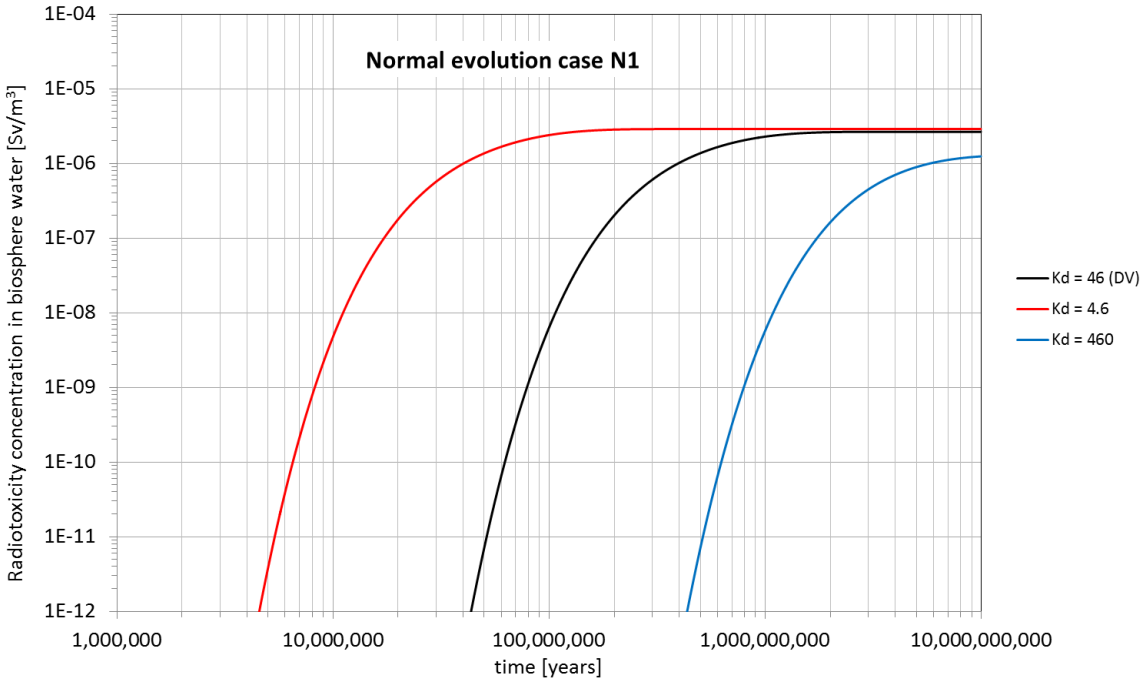

Figure 4-3: Radiotoxicity concentration in biosphere water from ^{238}U and its daughter nuclides for a K_d-value in Boom Clay according to the N1 case DV (black), a 10 times higher (red) and a 10 times lower K_d-value (blue).

In conclusion, it can be noted that some uncertainties exist in relation to the sorption behaviour, but there is no reason to assume that these will increase on very long term. Besides, these uncertainties affect the maximum radiotoxicity value only marginally.

4.4.Diffusion

In OPERA, the role of diffusion in the complex and heterogeneous Boom Clay has extensively been analysed [69]. In addition, a reference database with diffusion parameters has been elaborated required to calculate the migration of different radionuclides through the Boom Clay. The data presented in Appendix F, Table F-13, summarize the migration parameters values deduced for the Boom Clay for conditions as expected for the OPERA reference disposal concept ([69], Table 6-9).

The diffusion properties listed in Table F-13 are valid for salinities and *in-situ* effective stress ranging from the Belgian conditions (low salinity and in situ effective stress 2.4 MPa) and the highest expected salt level and in situ effective stress for a Dutch situation (sea water ±0.6 M, in situ effective stress 6.9 MPa). The used methods, data and assumptions are documented in [69].

The diffusion parameter in Table F-13 covers a large range of ionic strengths and different DOC concentration, which are assumed to cover the long term variation of Boom Clay properties in an evolving and changing environment. The most important unknown on the very long term is the thickness of the overburden that affects the porosity of the Boom

Clay. Here, with increasing overburden, e.g. due to glaciation, the pore space may decrease, potentially affecting migration in two manners:

- a decrease of the pore space volume leads to a temporary increase of radionuclide migration due to Boom Clay pore water that is squeezed out into the overburden [13][6];
- a smaller pore space results in slower diffusion, with the diffusion accessible porosity might change even stronger than the water filled porosity due to partially breakdown of the channel network through which the nuclides migrate.

Together with the K_d-values resulting from OPERA Task 6.1.2 (see previous section), these data determine the overall migration rates in the OPERA PA calculations. Thus, comparable to sorption, the range of values for diffusion properties given in Table F-13 mainly affects the time step of the peak radiotoxicity concentration, but not the peak value itself (see also [57]).

[6] and - in case of periodic events as glaciation - leads to a decrease of radionuclide migration in the period thereafter

5 Synthesis and conclusions

In Chapter 2, two major questions were raised:

1. What are the relevant uncertainties on the very long term (i.e. beyond one million years), and how can these be addressed?
2. How can the outcomes of a safety assessment be put into perspective, given the fact that the second peak occurs in a very distant future, on a geological timescale far beyond human imagination?

With respect to the first question, it was argued that uncertainty in the long-term evolution decreases with depth, i.e. processes in the biosphere are uncertain on a shorter timescale than processes in the deeper underground. As conclusion, it was recommend to apply additional safety indicators that rely less on assumptions on the most uncertain compartment on the long term, the biosphere.

The safety indicator *Radiotoxicity concentration in biosphere water* needs no assumption on the biosphere evolution and was also applied in [23], with a reference value defined in [21]. As part of the present work, additional calculations are performed for this indicator, covering a time period of 1.5 billion years (Figure 2-1). Although the maximum value was not reached during that period, it can be assumed that the peak value will remain well below the reference value for this calculation case.

A safety indicator *Radiotoxicity flux from geosphere* or *Radiotoxicity flux from host rock* would be even more suitable to assess the safety on geological time scales, since this indicator is less dependent on uncertainties related to the overburden evolution. However, no robust reference value could be provided [21], a problem not unique to the OPERA Safety Case [19].

With respect to the relevant uncertainties about the regional or continental evolution on geological time scales, not much can be concluded on basis of the information provided in OPERA, since these do not cover that long times scales. In general, the applicability of the PA-model used in OPERA depends on the assumption that also in future a continuous, subterranean host rock layer of about 100 m thickness is present. Changes in groundwater composition that may slowly alter the host rock's pore water composition are assumed to be largely covered in the NES by the range of conditions used to derive the migration parameters. Periodic "squeezing" of the Boom clay layer by glacial periods was shown to contribute only little to the overall migration [12]. The largest effect on the radiotoxicity concentration in the host rock as well as in the biosphere has the solubility limit of uranium, and general uncertainties in the determination of the solubility have been noted. However, looking to the evolution of the uncertainty of the solubility limit, it is expected that solubility will only decrease on the very long term.

When looking at the timescales beyond a billion year, uncertainties on long term are not limited to regional and continental processes: on this times scale, our solar system's evolution results in dramatic changes of the Earth's ecosystem even before the uranium concentrations in the biosphere have reached their maximum concentration. These changes question the presence of human life as we know it today, making the assessment results presented here very questionable.

With respect to the second question, it was shown that on geological time scales it is sufficient to understand the solubility and migration behaviour of ^{238}U, rather than assessing the inventory of all radionuclides. This allows comparing assessment outcomes with measured natural background concentrations in Boom Clay and the biosphere. Furthermore, analysis of natural analogues with respect to uranium confinement and migration in the overburden provided a number of "lessons learned'.

While large question marks can be placed on results calculated for such long calculation time frames, is it shown that - within the uncertainties discussed above - in principle a similar level of protection can be provided for future generations as for the current generation. It can also be stated that the additional safety assessment calculation performed has extended sufficiently far into the future to ensure that even the very late appearance of uranium in the biosphere is covered. It is, however, necessary to develop a more detailed 'story line' on the very long term evolution of the host rock compartment, i.e. far beyond the period covered in [12]. The 'story line' must address the principal regional and continental evolution processes on the very long term, and should also mark unresolvable uncertainties. Such an analysis must be performed in a manner that would either support the current safety assessment approach, where the host rock is largely stable in its properties and extend, or would allow the integration additional processes models related to the long term evolution of the host rock in the existing OPERA performance assessment model.

Summarizing, despite the uncertainties discussed above, a number of conclusions can be made:

- When assessing safety in the very long term (beyond 1 million year), in the Normal Evolution Scenario (NES) only ^{238}U and its daughter nuclides are of relevance.
- It is, however, insufficient to consider environmental concentrations of U only, because the overall radiotoxicity is dominated by its daughter nuclides, resulting in an almost thirty times increase of the overall radiotoxicity.
- The uranium inventory of the OPERA disposal concept is dominated by depleted uranium with an overall amount about 25 times higher than what could be expected from reactor operations in the Netherlands.
- The amount of *U* in the OPERA disposal is considerable compared to the natural background concentrations: a footprint of about 180 km^2 of a 100 m thick layer of Boom Clay contains an amount of U equal to the amount intended for disposal.
- For the assessment of safety on the very long term, the use of additional safety indicators is recommended. The *radiotoxicity flux from geosphere* is in principle a suitable indicator on the very long term, because it is not based on (uncertain) assumptions on the overburden and biosphere. However, it lacks a robust reference value and can therefore only serve as performance indicator that allows to analyse and compare the system behaviour for different scenarios, calculation cases etc.
- The calculated radiotoxicity concentrations in the biosphere remain below the reference values. Although unresolved uncertainties on the evolution of the host rock on such long time scales (>1 billion years) exist, it could be shown that within the uncertainties addressed in OPERA and the OPERA PA-model, a comparable level of protection of future generation can be provided. The calculation period used in this report was sufficient to cover also the very late appearance of uranium and its daughter nuclides in the biosphere.

Appendix A: Uranium in the waste

Depleted uranium is generated by URENCO resulting from uranium enrichment activities. The tails resulting from this process, mostly containing ^{238}U and smaller fractions of ^{235}U and ^{234}U, are converted to solid uranium oxide (U_3O_8) in France, and stored at COVRA. Table A-1 summarizes the inventory of the various uranium isotopes contained in depleted uranium and the other anticipated waste fractions distinguished in OPERA ([31]; Table 5-2; Table A-7).

Table A-1: Inventory of disposal sections ([31])

Nuclide	Half-life [a]	Inventory of disposal sections [Bq]				
		Vitrified HLW	Spent Fuel	Non-heat-generating HLW	DepU	LILW
^{232}U	6.98E+01	1.41E+12	3.63E+12	1.80E+10	4.24E+12	2.37E+07
^{233}U	1.59E+05	1.53E+09	2.75E+08	1.13E+07	-	2.86E+08
^{234}U	2.46E+05	2.28E+11	5.99E+12	1.72E+11	1.57E+15	7.21E+10
^{235}U	7.04E+08	1.38E+09	7.80E+10	3.61E+09	3.14E+13	1.93E+11
^{236}U	2.37E+07	2.01E+10	9.29E+11	3.03E+10	3.72E+14	1.34E+09
^{238}U	4.47E+09	2.64E+10	1.13E+11	1.13E+10	1.36E+15	5.36E+12

It should be noted that the decay of ^{241}Am, mainly present vitrified HLW, contributes in the long-term relevantly to the inventory of ^{233}U: the ^{233}U inventory of $2 \cdot 10^9$ Bq in 2130 (Table 3-1) will increase to a maximum of $3 \cdot 10^{13}$ Bq after 650'000 years (Figure 3-3). The inventory of ^{234}U will increase strongly, too, due to the ingrowth from ^{238}U. Nevertheless, even with accounting for this, ^{238}U is still the dominant nuclide contributing to risks in the very long term. Other relevant nuclides in the long-term are the daughters of ^{238}U and ^{234}U,, i.e. ^{230}Th, ^{226}Ra, and ^{210}Pb, which are virtually in equilibrium after 1 million years (Figure 3-3).

Following the methodology for analysing the actinide decay chains as outlined in ([33]; Section 2.2), the long-term activities as well as the radiotoxicity values of depleted uranium and daughter nuclides can be calculated. The results are depicted in Figure A-1 (activities) and Figure A-2 (radiotoxicities). These figures show that after about 1 million years the isotopes of depleted uranium and their daughter nuclides are in equilibrium. The daughter nuclides contributing mostly to the total long-term radiotoxicity are, respectively, Pb-210, Ra-226, Th-230, Ac-227, and Pa-231, with the latter two contributing only a few percent to the overall radiotoxicity.

From the decay calculations, it also can be estimated that the total radiotoxicity resulting from disposed depleted uranium in the long term, i.e. after 1 million years, will have increased by a factor 11.4 compared to the initial state. This is due to the fact that the dose coefficients of the daughter nuclides are higher than those of the uranium isotopes themselves. Relative to ^{238}U only, the total radiotoxicity in the long term increases even by a factor of 29 (see Figure 3-4).

The long-term values of the activities and radiotoxicities of depleted uranium and daughter nuclides are given in Table A-2. Values of $e(50)_{ing}$ are obtained from (22; Table 4.1).

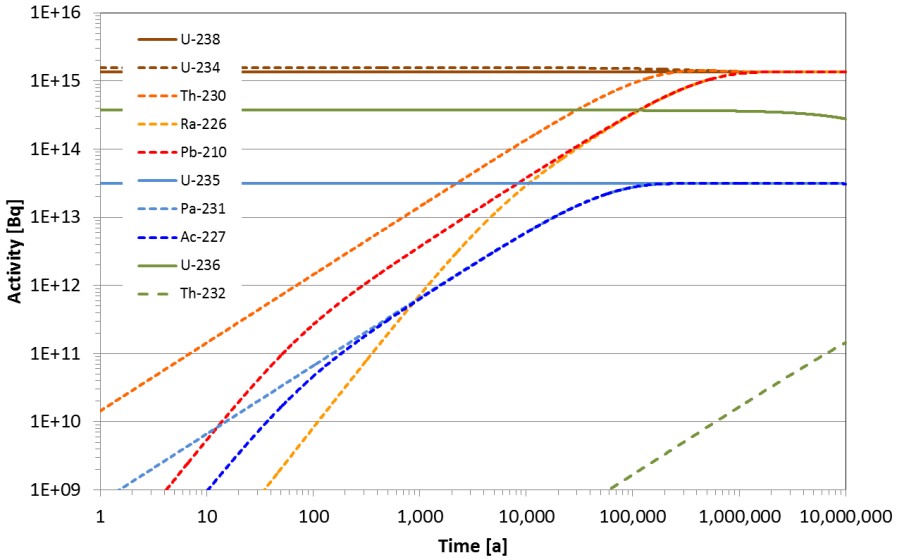

Figure A-1: Activities of depleted uranium and relevant daughter nuclides

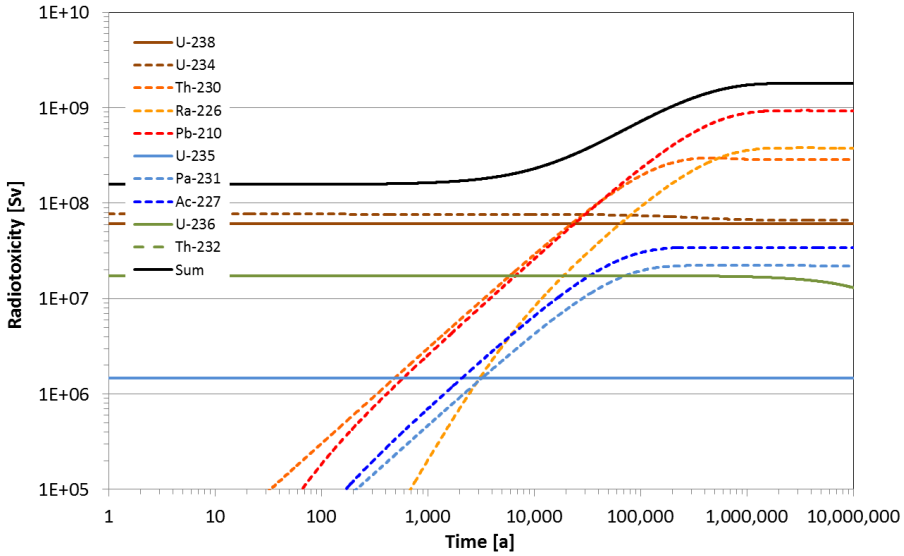

Figure A-2: Radiotoxicities of depleted uranium and relevant daughter nuclides

Table A-2: Nuclide data, activities and radiotoxicities of depleted uranium and daughter nuclides (10 million a)

Isotope	Half life [a]	$e(50)_{ing}$ [Sv/Bq]	Activity [Bq]	Radiotoxicity [Sv]	Inventory [mole]	Inventory [g]	Inventory [kg]
U-238	4.468E+09	4.5E-08	1.36E+15	6.11E+07	4.583E+08	1.091E+11	1.091E+08
U-234	2.457E+05	4.9E-08	1.36E+15	6.65E+07	2.521E+04	5.898E+06	5.898E+03
Th-230	7.540E+04	2.1E-07	1.36E+15	2.85E+08	7.735E+03	1.779E+06	1.779E+03
Ra-226	1.600E+03	2.8E-07	1.36E+15	3.80E+08	1.641E+02	3.710E+04	3.710E+01
Pb-210	2.216E+01	6.9E-07	1.36E+15	9.36E+08	2.273E+00	4.774E+02	4.774E-01
U-235	7.038E+08	4.7E-08	3.10E+13	1.46E+06	1.652E+06	3.881E+08	3.881E+05
Pa-231	3.276E+04	7.1E-07	3.10E+13	2.20E+07	7.688E+01	1.776E+04	1.776E+01
Ac-227	2.177E+01	1.1E-06	3.10E+13	3.41E+07	5.110E-02	1.160E+01	1.160E-02
U-236	2.370E+07	4.7E-08	2.77E+14	1.30E+07	4.968E+05	1.172E+08	1.172E+05
Th-232	1.405E+10	2.3E-07	1.46E+11	3.35E+04	1.546E+05	3.587E+07	3.587E+04

Appendix B: Uranium in the host rock

The lateral and depth-related heterogeneity of the Boom Clay in the Netherlands has been studied in OPERA Task 5.2.1 [34]. For this purpose, a total number of 152 Boom Clay samples were selected from 17 cores taken at various locations in the Netherlands. Subsequently, geochemical and grain size analyses were performed and statistically analysed. The samples show considerable variability, mainly in their clay, quartz, carbonate and pyrite contents. The main part of the heavy and trace elements can be associated with the clay mineralogy. Strontium and uranium are linked to calcite and organic carbon respectively.

Geographically, three different groups of Boom Clay are recognized in [70] which differ by their constitution. The Boom Clay in the southern part of the Netherlands consists of coarser, silty upper and lower layers. The central layer is finer grained and more clay-rich with occasional silty layers. This is consistent with the cyclic alternation of clay- and silt-rich layers found in the Belgian Boom Clay. In the Southeast of the Netherlands, the Boom Clay has higher carbonate content than in the Southwest. The Boom Clay in the north of the Netherlands is significantly different from the Southeast and Southwest. The Boom Clay in the north is fine grained and clay- and carbonate-rich over the total depth interval. Both the pyrite and organic carbon content are important parameters due to their reactivity and potential impact on the safety function 'delay and attenuation of releases'. The pyrite and organic carbon contents vary among the samples but they do not show geographic or depth-related variations. For the present project only the results obtained for uranium are considered.

Table B-3 shows selected statistics of the distribution of the uranium content (mg/kg) in Boom Clay ([34], Table 11). Note that the dataset contains one outlier with extreme uranium content, for which a large concentration of the mineral apatite was suggested ([34], p.18).

Table B-3: Distribution of uranium content (ppm) in Boom Clay

Statistical parameter	Value [mg/kg]	Value excl. outlier [mg/kg]
Minimum	0.8	0.8
Maximum	28.3	8.2
Median	3.30	3.30
Average	3.54	3.37

Appendix C: Uranium in the biosphere

Table C-4: Uranium content in the topsoil of the Netherlands - *U - total amount (mg/kg)* ([37], p. 255)

Layer	Soil type	min	P5	P25	median	P75	P95	max	n
upper	marsh	0.139	0.305	0.703	1.79	2.88	8.91	10.9	33
	sand	0.271	0.35	0.497	0.638	0.804	1.37	2.07	178
	marine clay	0.317	0.991	1.64	1.96	2.27	2.71	3.25	115
	fluvial clay	0.451	0.978	1.62	2.32	2.83	3.65	4.04	28
	Loess	2.26	2.27	2.31	2.35	2.39	2.42	2.43	4
lower	marsh	0.0515	0.09	0.368	0.636	1.99	7.39	9.69	33
	sand	0.216	0.297	0.379	0.485	0.656	0.984	2.85	178
	marine clay	0.267	0.519	1.29	1.77	2.22	2.73	4.5	115
	fluvial clay	0.404	0.458	1.6	2.12	3.16	6.43	9.68	28
	Loess	2.18	2.19	2.26	2.31	2.34	2.36	2.36	4
Av. upper		0.295	0.656	1.115	1.677	2.196	4.160	5.065	
Av. lower		0.235	0.341	0.909	1.253	2.007	4.384	6.680	
Average		**0.265**	**0.499**	**1.012**	**1.465**	**2.101**	**4.272**	**5.873**	

Table C-5: Uranium content in the topsoil of the Netherlands - *U - reactive amounts (mg/kg)* ([37], p. 255)

Layer	Soil type	min	P5	P25	median	P75	P95	max	n
upper	marsh	0.011	0.0155	0.0726	0.249	0.706	2.71	2.95	32
	sand	0.007	0.0125	0.0298	0.082	0.135	0.22	0.55	172
	marine clay	0.00102	0.116	0.207	0.276	0.342	0.546	1.02	81
	fluvial clay	0.019	0.122	0.169	0.234	0.475	0.823	1.2	28
	Loess	0.184	0.186	0.194	0.203	0.22	0.244	0.25	4
lower	marsh	0.00101	0.002	0.0119	0.057	0.249	1.21	2.54	31
	sand	0.0035	0.006	0.01	0.0162	0.0325	0.103	0.716	162
	marine clay	0.00101	0.0313	0.15	0.232	0.299	0.577	1.13	77
	fluvial clay	0.0095	0.0466	0.155	0.254	0.593	2.17	3.1	27
	Loess	0.112	0.116	0.129	0.136	0.143	0.154	0.157	4
Av. upper		0.0095	0.0665	0.1196	0.2103	0.4145	1.0748	1.4300	
Av. lower		0.0038	0.0215	0.0817	0.1398	0.2934	1.0150	1.8715	
Average		**0.0066**	**0.0440**	**0.1007**	**0.1750**	**0.3539**	**1.0449**	**1.6508**	

Table C-6: Uranium content in the topsoil of the Netherlands - *U - extractable concentrations (µg/l)* ([37], p. 256)

Layer	Soil type	min	P5	P25	median	P75	P95	max	n
upper	marsh	0.01	0.0155	0.0675	0.185	0.47	1.39	1.66	32
	sand	0.01	0.01	0.03	0.05	0.09	0.15	0.35	167
	marine clay	0.01	0.01	0.05	0.09	0.175	0.357	0.73	79
	fluvial clay	0.01	0.01	0.0275	0.055	0.115	0.38	0.66	28
	Loess	0.01	0.01	0.01	0.015	0.0275	0.0455	0.05	4
lower	marsh	0.01	0.01	0.02	0.09	0.31	0.388	2.83	24
	sand	0.01	0.01	0.01	0.03	0.05	0.173	0.39	135
	marine clay	0.01	0.02	0.08	0.14	0.255	1.02	3.48	71
	fluvial clay	0.01	0.0175	0.08	0.13	0.218	0.633	1.12	16
	Loess	-	-	-	-	-	-	-	0
Av. top		0.0100	0.0114	0.0438	0.0950	0.2125	0.5693	0.8500	
Av. lower		0.0100	0.0144	0.0475	0.0975	0.2083	0.5535	1.9550	
Average		**0.0100**	**0.0129**	**0.0456**	**0.0963**	**0.2104**	**0.5614**	**1.4025**	

Appendix D: Drinkwater quality standards

> *"Depleted uranium is not a significant health hazard unless it is taken into the body. External exposure to radiation from depleted uranium is generally not a major concern because the alpha particles emitted by its isotopes travel only a few centimeters in air or can be stopped by a sheet of paper. Also, the uranium-235 that remains in depleted uranium emits only a small amount of low-energy gamma radiation. However, if allowed to enter the body, depleted uranium, like natural uranium, has the potential for both chemical and radiological toxicity, with the two important target organs being the kidneys and the lungs. The most likely pathways by which uranium could enter the body are ingestion and inhalation. The relative contribution of each pathway to the total uptake into the body depends on the physical and chemical nature of the uranium, as well as the level and duration of exposure."* [71]

Contents of selected chemical elements in drinking water have been reported in *Water quality standards for uranium* [72]. For establishing water quality standards for uranium in drinking water, only natural uranium in the environment is considered, which consists of the three isotopes U-234 (0.0055 wt% abundance), U-235 (0.72 wt%), and U-238 (99.27 wt%).

Under the Water Framework Directive two types of quality standards are distinguished: the *Annual Average Environmental Quality Standard* (AA-EQS) and the *Maximum Acceptable Concentration EQS* (MAC-EQS). The AA-EQS is the concentration which should protect the ecosystem against adverse effects resulting from long-term exposure. The MAC-EQS protects aquatic ecosystems from effects due to short-term exposure or concentration peaks. Both standards are expressed as dissolved uranium, including background levels. Monitoring data indicate that the proposed value is currently exceeded in some of the Dutch surface waters.

Next to the AA-EQS and MAC-EQS, the European Water Framework Directive (WFD) also considers a standard for surface water used for drinking water abstraction. In addition to these WFD standards, additional risk limits apply that can be used for the purpose of national water quality policy, e.g. discharge permits or specific policy measures. These are the *Negligible Concentration* (NC), and the *Serious Risk Concentration* for ecosystems (SRC_{eco}) ([72]; p.9). The quality standard for surface water that is used for drinking water abstraction is referred to as $QS_{dw,hh}$. This is the concentration in surface water that meets the requirements for use of surface water for drinking water production. The $QS_{dw,hh}$ specifically refers to locations that are used for drinking water abstraction.

Table C-7 summarizes the values proposed for AA-EQS and MAC-EQS, as well as the WFD standards (72; p.10).

Table C-7 Proposed water quality standards for uranium ([72]; p.10)

Standard	Name	Value
AA-EQS	Annual Average Environmental Quality Standard	0.5 µg/L
MAC-EQS	Maximum Acceptable Concentration EQS	8.9 µg/L
NC	Negligible Concentration	0.33 µg/L
SRC_{eco}	Serious Risk Concentration for ecosystems	56 µg/L
$QS_{dw, hh}$	Quality standard for surface water	30 µg/L

Appendix E: Natural analogues

One of the more difficult aspects involved in the disposal of radioactive waste in deep geological disposal facilities are the very long time frames required by the safety assessment. Especially in the case of uranium and its daughter nuclides the time scales of concern may extend over millions of years after disposal into a dedicated facility.

Such time frames are well beyond anything that can be considered in experimental settings like demonstration projects in laboratories and large-scale projects in underground laboratories. Mathematical models can provide long-term estimates on the long-term safety, but they face considerable uncertainties over very long time frames. Due to the uncertainty of predictions made far into the future the reliability of quantitative predictions, relying on experimental data, decreases with increasing timescale.

Therefore, the demonstration of the long-term safety of a deep geological repository may rely less on quantitative predictions and more on qualitative arguments as timescales increase. The application of such an approach is referred to as *multiple lines of reasoning*.

The use of multiple lines of reasoning may add value to the safety case by providing arguments that together build confidence in certain data, assumptions and results underpinning the safety of a facility for the disposal of radioactive waste. The consideration of comparing the processes occurring in a repository following the emplacement of radioactive waste with natural analogues can complement numerical modelling with quantitative data from systems that formed over geologically long time periods, and that have remained stable for millions to billions of years. Furthermore, arguments complementary to scientific ones may be more meaningful to specific audiences ([73], p. 40).

Considerations about the use of natural analogues

General considerations about the use of natural analogues as alternative argument for building confidence in deep geological disposal are the following ([58], Chapter 5:

- The existence of large underground bodies of enriched uranium ore provides a good argument for the potential of disposal of uranium-containing radioactive waste in stable host rocks. This stability allows the altered zone around the ore body to shield the uranium from the effects of water flow through the surrounding rock – and for periods far beyond those required for the hazard from radioactive waste to decay to insignificant radiological properties;
- Natural uranium-rich ores in the sub-surface lack the presence of materials which are disposed in a deep geological repository such as large quantities of steel and concrete, or organic material in the case of LILW. These materials may adversely affect the (chemical) stability of the surrounding rock and enhance the transport of radionuclides released from the emplaced waste.

Although some analogue studies have provided invaluable information regarding repository selection and design, a number of recommendations have recently been inventoried in a study under the auspices of the Canadian Nuclear Safety Commission, CNSC [58]:

- Undertake a systematic review of key information, identify a set of needs (a gap analysis), and decide which of these gaps may best be fulfilled by analogue information.
- Public support for the deep geological repository may benefit from *national* analogues, i.e. host country environments, because it is geographically and culturally familiar, which may provide some reassurance to the public.
- Develop a national analogue program that is structured and can provide input to the development of a safety case, with an emphasis on specific national geological disposal concepts.

- Engage safety assessment specialists early on in the process of safety case development when selecting new analogue studies or evaluating existing analogue information. This will ensure that the required information is obtained for the safety case.
- Integrate natural analogue information with other studies (e.g., hydrogeology, rock mechanics etc.) including laboratory experiments. This will provide more quantitative data that can be used in PA models.
- To build confidence in a geological disposal concept multiple lines of evidence are necessary. Therefore, it is recommended that an attempt is made to evaluate the true potential of analogues for public communication and dialogue through a structured opinion survey.
- In addition to natural (or national) analogues, anthropogenic analogues are judged important for building public confidence in geological disposal concepts.
- Analogue studies for the near-field chemical environment, which is inevitably location-specific, would add confidence to the long-term chemical behaviour of the anticipated host rock due to the presence of alien materials. Example studies could include:
    - high pH plume sites and natural cements,
    - microbial interaction associated with high-grade uranium deposits,
    - natural analogues for sorption and diffusion of radionuclides (e.g., long distance transport),
    - natural analogues of secondary traps for radionuclides,
    - colloidal transport in natural systems, and
    - natural analogues for site-specific matrix diffusion quantification.

Uranium roll fronts

A schematic of the changing *Eh* and *pH* conditions during the transport and deposition of uranium is depicted in Figure E-3 [74].It is noted that the *pH*- and *Eh*-value below are expected to be not representative for the Netherlands.

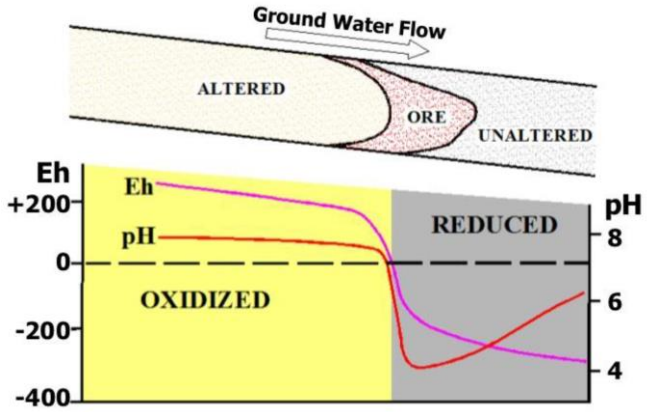

Figure E-3: Postulated *Eh-pH* conditions during transportation and deposition of uranium

Other natural analogue studies

In addition to Cigar Lake, many other studies have been performed in the past about the utilization of anthropogenic and natural analogues in relation the geological disposal of radioactive waste. The following tables provide an overview of relevant analogues for a disposal concept in Boom Clay on the very long term [58].

Table E-8: Inventory of analogue studies for the engineered barrier system

Location/Site	Analogue
Near-field barrier materials	
Dunarobba Forest, Todi, Italy	Long-term isolation properties of clay
The Philippines	Long-term isolation properties of clay
Cyprus	Long-term isolation properties of clay
Mudrocks altered by igneous intrusions	Thermal stability of clay barriers

Table E-9: Inventory of analogue studies for the natural barrier system

Location/Site	Analogue
Long-term Isolation concepts	
Several locations	Matrix diffusion: long-term isolation properties of the host rock

Table E-10: Inventory of analogue studies for the radionuclide migration in natural systems

Location/Site	Analogue
Retardation in natural systems	
Poços de Caldas: Morro do Ferro, Brazil	Radionuclide migration
Poços de Caldas: Osamu Utsumi mine, Brazil	Radionuclide migration
Poços de Caldas: Osamu Utsumi mine, Brazil	Redox fronts
The El Berrocal Project	Analogue for uranium mobilisation and migration from a radioactive waste repository
Needle's Eye, Scotland	Uranium mobilisation and migration
Broubster, Scotland	Uranium mobilisation and migration
South Terras Mine	Uranium mobilisation and migration
Alligator River, Australia	Uranium mobilisation and migration
Loch Lomond, Scotland	Halogen migration
Colloid migration in natural systems	
Poços de Caldas, Morro do Ferro, Brazil	Colloid transport
Whole system performance	
Oklo	A natural analogue for the long-term behaviour of a GDF
Cigar Lake, Canada	A natural analogue for an entire GDF

Appendix F: Uranium migration in Boom Clay

Table F-11: Expected properties of Boom Clay in the Netherlands

property	min - max
Bulk wet density [kg/m^3]	1.900 – 2.150
Porosity [%]	29 – 43
CEC Boom Clay [meq/100g Boom Clay]	2.0 – 42
SOC [wt. %]	0.35 – 2.0
Proton exchange capacity SHA [meq/g]	1 – 2
DOC [mg/L]	20 – 200
Proton exchange capacity DHA [meq/g]	2 – 6
HFO [g/kg]	0.4 – 3.3
Inorganic carbon [wt. %]	0.0 – 2.5
Total amount Ca [wt. %]	0.2 – 7.3
Total amount Fe [wt. %]	2.2 – 5.4
Total amount S [wt. %]	0.35 – 2.6
Soluble concentration Cl [mg/L]	4 – 20'000
Soluble concentration Na [mg/L]	4 – 11'000
pH [-]	7.7 – 9.2
pe + pH [-]	3.8 – 5.8

Table F-12: Ranges of calculated K_d- and R-values in Boom Clay of the Netherlands for the '*base case*' (100 mg/l DOC). Lower, central, and upper values correspond to 5-, 50- and 95-percentiles of the calculated values, respectively.

Element	$K_{d\text{-}diss}$			$K_{d\text{-}DOC}$			R_{dis}			R_{DOC}		
	lower	central	upper	lower	central	upper	lower	central	upper	lower	central	upper
'base case' (100 mg/l DOC)												
U	7	>10'000	>10'000	16	46	95	33	>50'000	>50'000	77	221	489
Th	>10'000	>10'000	>10'000	16	46	95	>50'000	>50'000	>50'000	77	221	489
Ra	18	1554	>10'000	34	95	275	87	7320	>50'000	161	458	1364
Pb	>10'000	>10'000	>10'000	25	69	237	>50'000	>50'000	>50'000	120	338	1145
Ac	>10'000	>10'000	>10'000	16	46	95	>50'000	>50'000	>50'000	77	221	489
Pa	7	>10'000	>10'000	16	46	95	33	>50'000	>50'000	77	221	489
low DOC case' (20 mg/l DOC')												
U	7	>10'000	>10'000	81	231	473	33	>50'000	>50'000	382	1103	2442
Th	>10'000	>10'000	>10'000	81	231	473	>50'000	>50'000	>50'000	382	1103	2442
Ra	18	1550	>10'000	173	475	1373	87	7315	>50'000	799	2287	6816
Pb	>10'000	>10'000	>10'000	125	347	1184	>50'000	>50'000	>50'000	595	1685	5722
Ac	>10'000	>10'000	>10'000	81	231	473	>50'000	>50'000	>50'000	382	1103	2442
Pa	7	>10'000	>10'000	81	231	473	33	>50'000	>50'000	382	1103	2442
high DOC case' (200 mg/l DOC')												
U	7	>10'000	>10'000	8	23	47	33	>50'000	>50'000	39	111	245
Th	>10'000	>10'000	>10'000	8	23	47	>50'000	>50'000	>50'000	39	111	245
Ra	18	1557	>10'000	17	47	137	87	7330	>50'000	81	230	682
Pb	>10'000	>10'000	>10'000	12	35	118	>50'000	>50'000	>50'000	60	169	573
Ac	>10'000	>10'000	>10'000	8	23	47	>50'000	>50'000	>50'000	39	111	245
Pa	7	>10'000	>10'000	8	23	47	33	>50'000	>50'000	39	111	245

Table F-13: Estimated minimum, maximum and average values for the pore diffusion coefficient D_{pore}, diffusion accessible porosity η and tortuosity for Boom clay for conditions as expected for the future Dutch disposal facility.

	Diffusion accessible porosity η [-]		Pore diffusion coefficient D_{pore} [m^2s^{-1}]	
Element	min	max	min	Max
U	0.07	0.17	$5.7\ 10^{-12}$	$5.7\ 10^{-11}$

Appendix G: Yardsticks for safety assessments

Yardsticks for qualitatively assessing the safety of a facility for the final disposal of radioactive waste may be derived from a number of sources, including legislation or regulation, which typically provide guidelines or limits on dose or risk, as illustrated in Figure G-4 [14; Fig. 15, p.79]. Typical sources include:

- Safety recommendations from international organisations that may relate to radiological safety (e.g. ICRP) or broader health and environmental safety (e.g. drinking water standards);
- The principle that the repository should not significantly perturb the radiological or chemical conditions naturally present in the environment. Corresponding yardsticks can be derived from natural radionuclide concentrations and fluxes;
- Societal values or expectations;
- The results of performance assessments (e.g. a critical minimum container lifetime);
- System understanding, considering the physical processes by which the safety functions of the disposal system are provided.

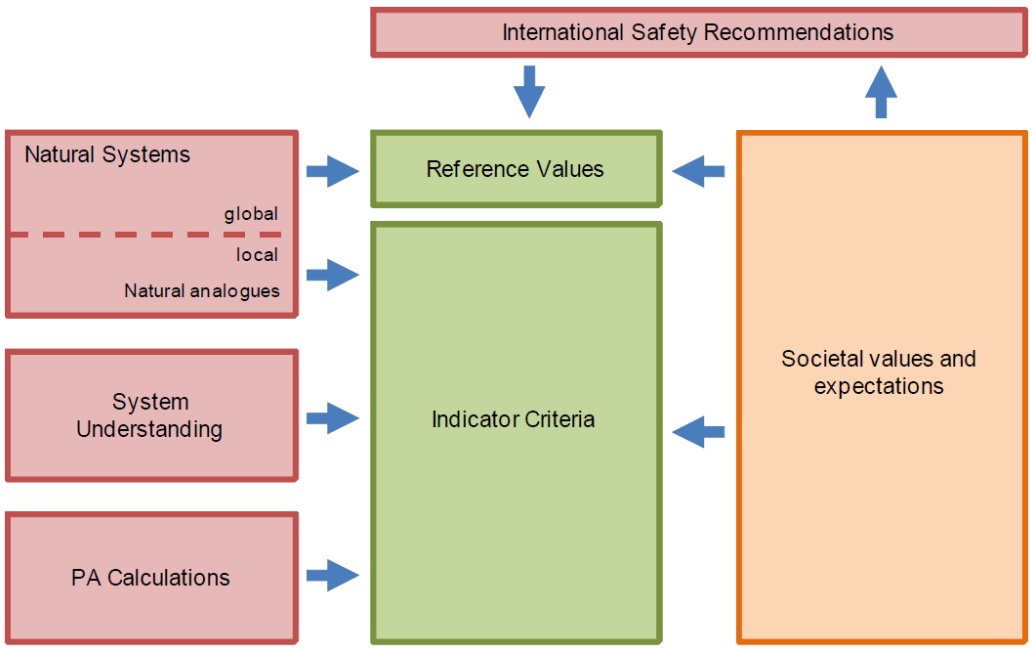

Figure G-4: Sources of references values and indicator criteria

In [17, p.10*ff*] it is noted that in order to constitute a meaningful measure of safety, a safety indicator must be compared to a yardstick that conveys information with respect to the impacts on humans and the environment. It also emphasizes that reference values, against which the yardsticks are to be compared, should be defined in a way that is generally considered to be acceptable.

In principle, reference values for safety indicators can be based on three lines of reasoning:

- dose constraints, that can be related to actual calculated risks;
- natural processes or features, e.g. radiotoxicity fluxes or concentration in groundwater;
- reference values used for other purposes.

While the first type of argument is based on legally or regulatory defined radiological constraints, the second type is related to natural features of a site, e.g. concentrations of naturally occurring uranium in groundwater. In MeSA [14, p.79*f*], it is stated that a repository system can be considered safe if possible radionuclide releases remain low in comparison with the natural radionuclide content of the environment. It is however noted that this is a "*somewhat problematic principle*" because the concentrations and fluxes in natural systems vary widely, and no guarantee exists that the natural environment is safe. This is of interest especially where high local natural concentrations of uranium or other radionuclides are present, while no epidemiological evidence exists suggesting that people have any increased risk of cancers. On the other hand, [14, p.80] noted that in areas with extremely low radionuclide concentrations unduly low reference values may be derived.

Reference values can also be based on limits used for other purposes. E.g. in [19, p.30], a reference value for a flux-related indicator was derived from an existing national regulation on the application of phosphate fertilizer. In such cases one needs to carefully investigate what the rationale behind the 'adapted' reference values or guidelines is: reference values can represent practical attempts to implement a certain policy, e.g. based on feasibility aspects or the desire to steer a certain process, rather than that the values are directly linked to assessed radiological risk limits.

Although there is a small number of universally applicable reference values that may be used in all safety cases, such as internationally agreed drinking water standards, it was acknowledged in *MeSA* that the derivation of appropriate reference values can be difficult. A number of recommendations with respect to the use of safety indicators were provided [14, p.92*ff*], under which:

- Reference values for comparison with safety indicators should have a generally accepted safety significance and, ideally, local context. Good examples of reference values provided where: maximum permissible concentrations defined in drinking water standards such as those provided by the World Health Organisation; measured concentrations in local rivers and ground waters; and measured fluxes in the accessible environment (e.g. due to groundwater discharge or surface erosion)
- It was noted that when using locally derived reference values, care should be given to evaluate spatial and temporal variations, and to express this appropriately.
- Reference values derived from local conditions should be treated with care to incorporate spatial and temporal variations, and to express this appropriately.

'Acceptability' of risks

Additional to the scientific-technical discussion on reference values, it is also important to acquire what a society in general assumes to be an 'acceptable' risk. The concept of defining an 'acceptable' risk evolved from the fact that absolute safety can never be achieved in any everyday activity or industrial practice, including radioactive waste disposal [14, p.83*f*]. In the current understanding, exposure of people and other biota to even very low radiological or chemotoxic substances involves some risk. In the UK, for example, the Health and Safety Executive (HSE) defines' acceptable risks as [[75], p.31]:

> *"a level of risk which, provided there is a benefit to be gained, and proper precautions are taken, does not worry us or cause us to alter our ordinary behaviour in any way".*

The general concept of 'benefit' (or 'justification', see [[76]]) is rather difficult to apply in case of the disposal of radioactive waste due to the long timescales until peak exposures are expected in performance assessment (ten thousand to hundred thousands of years). In [75, p.30*f*], a set of definitions with respect to risk perception was elaborated, covering qualification ranging from '*just about tolerable*' to '*acceptable*' risks:

- 1 in 1000 as the '*just about tolerable risk*' for any substantial category of workers for any large part of a working life.
- 1 in 10,000 as the '*maximum tolerable risk*' for members of the public from any single non-nuclear plant.
- 1 in 100,000 as the '*maximum tolerable risk*' for members of the public from any new nuclear power station.
- 1 in 1,000,000 as the level of '*acceptable risk*' at which no further improvements in safety need to be made.

What a society considers 'acceptable' depends on the complex national context and may differ between stakeholders and members of the public. Risks (and their benefits) are quite differently distributed across societies, and the societal factors and processes that determine whether a risk is acceptable will change with time and may affect the public perception of risk, eventually causing discrepancies in subjectivity judgment and statistically based measures of risks. In [77, p.208], a list of standpoints has been elaborated that could be used as a basis for determining whether a risk is considered 'acceptable' (or, perhaps, 'tolerable'). A risk is 'acceptable' when:

- it falls below an arbitrary defined probability;
- it falls below some level that is already tolerated;
- it falls below an arbitrary defined attributable fraction of total disease burden in the community;
- the cost of reducing the risk would exceed the costs saved;
- the cost of reducing the risk would exceed the costs saved when the '*costs of suffering*' are also factored in;
- the opportunity costs would be better spent on other, more pressing, public health problems;
- public health professionals say it is acceptable;
- the general public say it is acceptable (or more likely, do not say it is not);
- politicians say it is acceptable.

In the PAMINA project, conducted within the 6th Framework Programme of the European Commission, the term acceptable risk was described as the level of loss a society considers acceptable given existing social, economic, political, cultural, technical and environmental conditions [18; p. 26]. In environmental and especially in nuclear sciences there is the general agreement, that a risk of $1{\cdot}10^{-6}$ per year of suffering a serious health effect is an appropriate level as a regulatory constraint or target (e.g. [78]; p.72).

Reference values for risk-related safety indicators cannot be derived technically, when it is not known what level of risk a society considers acceptable. In the current phase of the Dutch disposal programme, no societal discussion could be identified that allows to link what is considered as 'societally accepted' with respect to risks for future generations.

Although discussions in ENGAGED with stakeholders pointed out that the use of reference values is very relevant and stakeholders liked to be engaged [79], no suggestions were given that can be used for the derivation of reference values in that report. However, a 'golden standard' [77, p.208] used in many risks-related field is a risk of $1{\cdot}10^{-6}$ per year (incidence or mortality), e.g. as regulatory constraint or target for a citizen living nearby a nuclear power plant under normal operation [75]. In many other international guidance documents and national regulations, an individual risk of 10^{-6} per year of suffering a serious health effect is often applied as a 'target level' for an acceptable risk [14, p.83*f*]. A value of 10^{-6} per year is discussed and applied also in the Netherlands with respect to risk attributed to nuclear power generation [80, 81].

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