CArbon-14 Source Term

CAST

Handling of C-14 in current safety assessments: State of the art

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CAST – Project Overview

The CAST project (CArbon-14 Source Term) aims to develop understanding of the potential release mechanisms of carbon-14 from radioactive waste materials under conditions relevant to waste packaging and disposal to underground geological disposal facilities. The project focuses on the release of carbon-14 as dissolved and gaseous species from irradiated metals (steels, Zircaloys), irradiated graphite and from ion-exchange materials.

The CAST consortium brings together 33 partners with a range of skills and competencies in the management of radioactive wastes containing carbon-14, geological disposal research, safety case development and experimental work on gas generation. The consortium consists of national waste management organisations, research institutes, universities and commercial organisations.

The objectives of the CAST project are to gain new scientific understanding of the rate of release of carbon-14 from the corrosion of irradiated steels and Zircaloys and from the leaching of ion-exchange resins and irradiated graphites under geological disposal conditions, its speciation and how these relate to carbon-14 inventory and aqueous conditions. These results will be evaluated in the context of national safety assessments and disseminated to interested stakeholders. The new understanding should be of relevance to national safety assessment stakeholders and will also provide an opportunity for training for early career researchers.

For more information, please visit the CAST website at:
http://www.projectcast.eu

CAST		
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Executive Summary

Workpackage 6 regroups the Waste Management Organisations (WMOs and other national organisations involved in safety assessments) participating to CAST with the aim to consider the results from WP2 to WP5 at the scale of their repository system and to analyse what is their impact in terms of long-term safety. WP6 can be seen as an integration exercise to ensure that the results from WP2 to 5 are relevant to the safety cases for the end-users, the WMOs, as possible.

The first phase of WP6, prior the analysis of the experimental results delivered by the project, aims at taking stock of the C-14 knowledge from the angle of safety assessments. Deliverable D6.1 gives an overview on this for each national program. On the basis of this compilation, commonalities and differences between disposal concepts regarding the impact of the uncertainties linked to C-14 release from radioactive waste disposal will be identified as well as the way to cope with them. This thorough discussion will be the object of deliverable D6.2.

Deliverable D6.1 is structured as follows: The focus is on geological or sub-surface systems and the waste families containing relevant amount of C-14. Each national contribution is about 10 pages long and shares the same table of content. The repository concept is first described to provide the context in which the safety assessment studies takes place. Next, the inventory of C-14 for each important waste family is provided. The expected evolution

scenario assumed for C-14 containing waste is then outlined with a focus on the

release mode (contribution to the instant release fraction vs. congruent mode), the transport models and the speciation and chemical behaviour of C-14 along the transport pathway to the biosphere.

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RWM contribution to D6.1

Helen Kendall

Date of issue of this report: 7/07/2015

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1 Geological Repository Concept

At the current stage in the United Kingdom (UK) Government programme to manage radioactive wastes, a preferred site for a Geological Disposal Facility (GDF) has not been identified. Therefore, any safety assessments produced as part of an overall safety Case are at a 'generic' stage. We call it a generic safety case because it must cover a range of possible disposal environments and facility designs. Nevertheless, this work builds on more than 25 years of experience studying geological disposal and undertaking safety assessments in the UK. It also draws on the extensive body of knowledge and experience in other countries gained through overseas radioactive waste management programmes.

At the current stage of the Radioactive Waste Management Limited (RWM) programme we are examining a wide range of potentially suitable disposal concepts so that a well-informed assessment of options can be carried out at appropriate decision points in the implementation programme. Drawing from this work we have set out illustrative concepts for three generic geological settings, including the associated variants on rock formations that might overlie the GDF host rock.

We are using these illustrative concepts to:

- further develop our understanding of the functional and technical requirements of the disposal system;
- further develop our understanding of the design requirements;
- support the scoping and assessment of the safety, environmental, social and economic impacts of a GDF;
- support development and prioritisation of our research and development programme;
- underpin our analysis of the potential cost of geological disposal; and
- support assessment of the disposability of waste packages proposed by waste owners.

We have set out the illustrative concepts solely for these purposes. We do not intend that one of these illustrative concepts is necessarily the one that we would use in the relevant geological setting. At this stage, no geological disposal concept has been ruled out. The key aspects of RWM's proposed approach to optioneering are described in [1].

We selected the illustrative geological disposal concepts following consideration of the concepts identified in the studies of disposal concepts for ILW/LLW [2] and for High Level Waste (HLW) and Spent Fuel (SF) [3]. We selected concepts that are well-developed and supported by extensive research and development and have been subject to detailed safety assessment, regulatory scrutiny and international review [4]. RWM's safety assessment studies at this stage assume that a GDF could be a 'co-located' facility. This means that the illustrative disposal concept examples for ILW/LLW and HLW/SF shown in Figure 1 are assumed to be developed at a single site. This is seen as a reasonable and conservative approach for assessment purposes at this stage. The illustrative concepts are listed in Figure 1 and the attached notes present the key reasons why these examples were selected.

The use of generic geological settings does not imply that any specific sites are being considered. The host rock descriptions correspond to three distinct general rock types that are considered potentially suitable to host a disposal facility for higher activity wastes, based on studies carried out in the UK and internationally, and which occur in the UK. They are described as follows:

- Higher strength rocks - these would typically comprise crystalline igneous, metamorphic rocks or geologically older sedimentary rocks, where any fluid movement is predominantly through divisions in the rock, often referred to as discontinuities. Granite is a good example of a rock that would fall in this category.
- Lower strength sedimentary rocks - these would typically comprise geologically younger sedimentary rocks where any fluid movement is predominantly through the rock mass itself. Many types of clay are good examples of this category of rocks.
- Evaporites - these would typically comprise anhydrite (anhydrous calcium sulphate), halite (rock salt) or other evaporites that result from the evaporation of water from water bodies containing dissolved salts.

These illustrative geological disposal concepts are described in the GDF Design Report [5]. A schematic illustration of a generic GDF comprising multiple barriers is shown in Figure 2.

Host rock	Illustrative Geological Disposal Concept Examples[d]	
	ILW/LLW	**HLW/SF**
Higher strength rocks[a]	UK ILW/LLW Concept *(NDA, UK)*	KBS-3V Concept *(SKB, Sweden)*
Lower strength sedimentary rock[b]	Opalinus Clay Concept *(Nagra, Switzerland)*	Opalinus Clay Concept *(Nagra, Switzerland)*
Evaporites[c]	WIPP Bedded Salt Concept *(US-DOE, USA)*	Gorleben Salt Dome Concept *(DBE-Technology, Germany)*

Notes

a. Higher strength rocks – the UK ILW/LLW concept and KBS-3V concept for spent fuel were selected due to availability of information on these concepts for the UK context.

b. Lower strength sedimentary rocks – the Opalinus Clay concept for disposal of long-lived ILW, HLW and spent fuel was selected because a recent OECD Nuclear Energy Agency review regarded the Nagra (Switzerland) assessment of the concept as state of the art with respect to the level of knowledge available. However, it should be noted that there is similarly extensive information available for a concept that has been developed for implementation in Callovo-Oxfordian Clay by Andra (France), and which has also been accorded strong endorsement from international peer review. Although we will use the Opalinus Clay concept as the basis of the illustrative example, we will also draw on information from the Andra programme. In addition, we will draw on information from the Belgian super container concept, based on disposal of HLW and spent fuel in Boom Clay.

c. Evaporites – the concept for the disposal of transuranic wastes (TRU) (long-lived ILW) in a bedded salt host rock at the Waste Isolation Pilot Plant (WIPP) in New Mexico was selected because of the wealth of information available from this United States Environmental Protection Agency (EPA) certified, and operating facility. The concept for disposal of HLW and spent fuel in a salt dome host rock developed by DBE Technology (Germany) was selected due to the level of concept information available.

d. For planning purposes the illustrative concept for depleted, natural and low enriched uranium is assumed to be same as for ILW/LLW and for plutonium and highly enriched uranium is assumed to the same as for HLW/SF.

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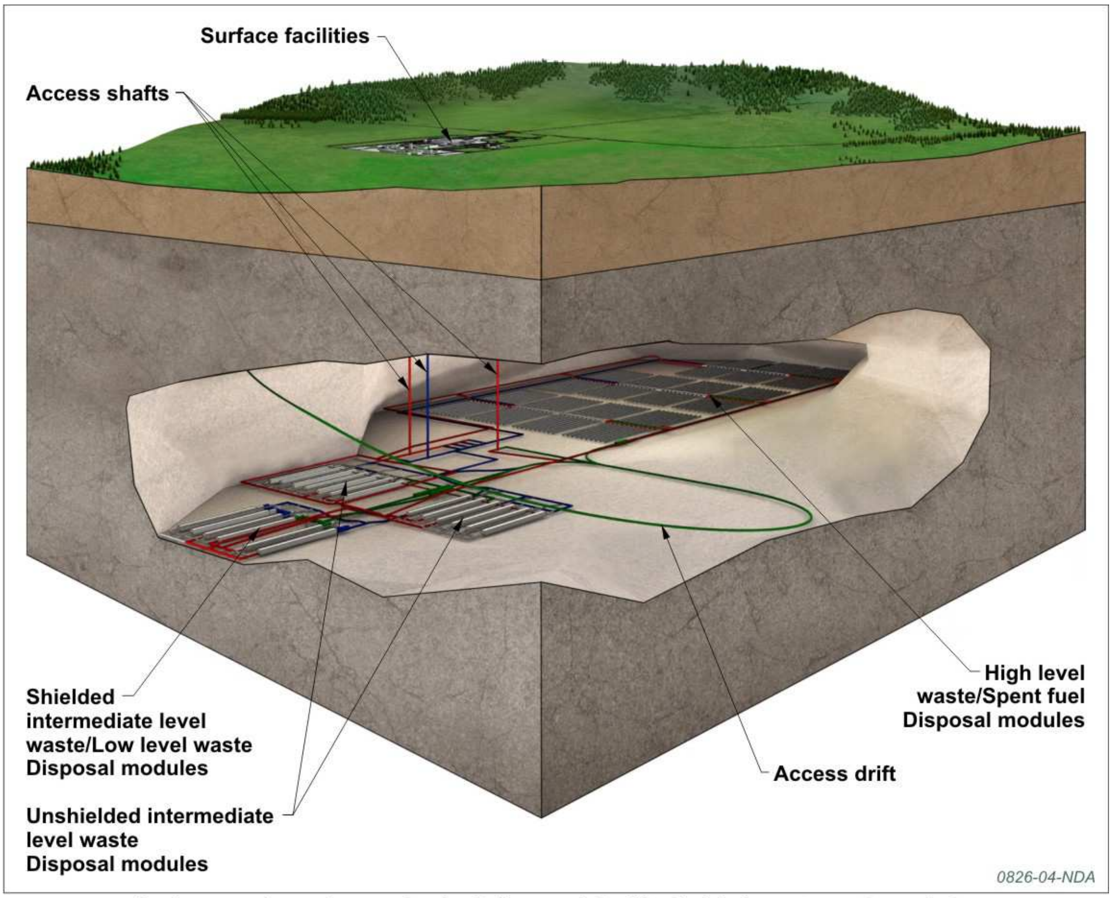
Figure 1 Illustrative geological disposal concept examples for different waste types



An impression of a geological disposal facility in higher strength rock for higher activity radioactive waste

Figure 2 Schematic illustration of the layout of a generic GDF (not host rock specific) for all UK higher activity radioactive wastes[1]

2 C-14 Inventory

The most recent safety assessment undertaken by RWM was part of the 2010 generic Disposal System Safety Case (DSSC) suite of documents [6]. This was based on the 2007 UK Radioactive Waste Inventory [7]. However, at the time of publication of the generic DSSC, an updated gas assessment using the 2007 Inventory data was not available. Therefore the safety assessment presented in the 2010 generic DSSC [8] used a previous gas assessment [9] which was based on an update to the 2004 UK Radioactive Waste Inventory [10].

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1 SF: spent fuel, UILW; unshielded ILW; SILW: shielded ILW

The breakdown of the carbon-14 (C14) inventory by material type was based on the 2004 UK Radioactive Waste Inventory but with enhancements as follows:

- the latest data on the inventory of power station moderator and reflector graphite were taken into account;
- where waste streams contained more than one material type of interest and where the C14 had arisen in each material as a result of neutron activation, the total C14 activity for the stream was apportioned to each material so that material totals could be generated;
- where well-established data on Magnox and uranium metal corrosion were available, the total C14 activity declared for the streams was apportioned between remaining metal and oxide/hydroxide corrosion products in proportion to the fraction of the original metal in each chemical form;
- certain inventory waste streams, were known to contain irradiated uranium and Magnox metal but no C14 activity was declared in their inventory - where possible additional activity estimates were developed for such missing contributions.

The 2004 C14 inventory for Intermediate Level Waste (ILW) and Low Level Waste (LLW) is detailed in Table 1, below. HLW and Spent Fuel are not currently considered as the focus in the UK is on transport via the gas pathway and our current assumption is that HLW and Spent Fuel will be packaged in high integrity containers. The impact of uncertainty in the C14 inventory was not assessed at this stage in these calculations.

Subsequent to the 2004 UK Radioactive Waste Inventory, updated inventories have also been produced including the 2007 [7], 2010 [11], and 2013 [12]. RWM also produces a differences report which details any significant differences between the last inventory and the new inventory [13], and then an implications report which describes the implications of the new inventory to the generic Disposal System Safety Case [14]. The current inventory (2013) includes assumptions about a possible new build programme and presents a total figure of C14 at 2200 of 1.77×10^4 TBq. This is larger than the previous inventory presented in Table 1 (and that used in our generic safety assessments), however, as mentioned before, this is because the 2013 inventory also includes wastes from new build nuclear power stations.

The major contributors to carbon-14 in stainless steel are Advanced Gas Reactor (AGR) fuel cladding and assembly components and items from reactor decommissioning. Other ferrous metals comprise mainly mild steel, with the major contributors being items from reactor decommissioning and miscellaneous activated components from reactor operations. Ion exchange resins only make a relatively small contribution to our carbon-14 inventory and are therefore not a priority to RWM at this stage. The waste is packaged differently and grouped according to the shielding of these packages – creating two groups of waste – Unshielded ILW and Shielded ILW. In a GDF these wastes would be disposed of in different vaults.

	Total Activity at 2090 (TBq)		
Material	**Unshielded ILW**	**Shielded ILW and LLW**	**Unshielded + Shielded**
Graphite	1.15E+02	6.23E+03	6.35E+03
Stainless Steel	6.91E+02	2.36E+01	7.15E+02
Mild Steel	1.54E+02	1.08E+02	2.61E+02
Zircaloy & Zirconium	5.90E+01	0.00E+00	5.90E+01
Nimonic alloys	4.29E+01	0.00E+00	4.29E+01
Magnox Metal	6.12E+01	0.00E+00	6.12E+01
Uranium Metal	2.80E+01	0.00E+00	2.80E+01
Corroded Magnox	1.85E+01	0.00E+00	1.85E+01
Corroded Uranium	6.90E+00	0.00E+00	6.90E+00
Non-metals, various types	4.98E+00	9.25E-04	4.99E+00
GE Healthcare waste stream 1B05	2.92E+02	0.00E+00	2.92E+02
GE Healthcare waste stream 1A07	2.18E+01	0.00E+00	2.18E+01
Barium Carbonate	3.17E+01	0.00E+00	3.17E+01
Material Composition Not-Assessed	1.32E+01	2.07E-02	1.33E+01
Total	1.54E+03	6.37E+03	7.91E+03

Table 1 Activity of C-14 Associated with Materials in Waste Streams at 2090 [10].

3 Safety concept

Concentrating and containing solid radioactive waste, and isolating it from the biosphere, is the internationally accepted strategy for the safe long-term management of such materials. In geological disposal, long-term containment and isolation of solid radioactive waste is provided by its emplacement in a facility located underground in a stable geological formation – the underground facilities and the geological environment comprise the geological disposal system. A distinctive feature of geological disposal is the depth of emplacement, 200 -1,000 metres below ground. The depth chosen for disposal in a particular facility, as well as specific elements of its design, will depend on a number of factors including, but not limited to, groundwater conditions, rock stability, host rock composition and the nature of the waste.

Safety is provided by a multi-barrier concept, these barriers are the wasteform, the waste container and overpack (where used), buffer/backfill material, mass backfill, seals, and the host rock. The ensemble of wasteform, waste container and overpack is referred to as a waste package. An overpack is an additional container used for disposal of wastes that are already packaged; for example, conditioned HLW is currently stored in stainless steel containers, and an overpack made of another material would be used to support the longevity of the waste package and the long-term safety of a GDF. The buffer/backfill refers to material placed adjacent to and around the waste package in a GDF. Mass backfill refers to material used to fill some or all of the empty space remaining in the excavated areas of a GDF, for example, tunnels and access-ways. The seals are the final engineered barriers to be emplaced when a GDF is sealed.

The engineered barriers and the natural barrier provided by the geological environment work together to provide the necessary level of safety and ensure that undue reliance is not placed on any one barrier.

4 Treatment of C-14 in safety assessment

In the UK, substantial quantities of carbon 14 are generated in nuclear power reactors [15]. In general, C14 is produced by:

- 14N(n,p)14C reactions with nitrogen present as an impurity in fuels, moderators coolants, and structural hardware;

- 17O(n,α)14C reactions in oxide fuels, moderators and coolants; and

- 13C(n,γ)14C reactions in graphite moderators.

Additionally, a proportion of the products used in life science research and medical drug production incorporate C14. Therefore the possible sources of C14 in the UK Radioactive Waste Inventory are irradiated metals, irradiated graphite and organic materials.

C14 could be present in irradiated metals either in elemental form or as metal carbide. On dissolution, the metal carbide could form hydrocarbons (such as methane) or small organic molecules (such as acetylene). The implications for a GDF of C14 release from irradiated metals depend partly on the corrosion rate of the metal (which, in turn, depends on environmental conditions), the surface area of the metal, the distribution of C14 in the metal and the form in which the C14 is released (as well as other processes such as its migration from a GDF). At present C14 is assumed to be released from the corrosion of irradiated metals in the form of methane, and the rate of release is assumed to be proportional to the corrosion rate.

The chemical form of C14 in irradiated graphite is not known although at least some may be present as compounds of hydrogen, nitrogen or oxygen [16]. The mechanism(s) by which C14 is released from irradiated graphite at low temperatures in aqueous solutions is also not well known at present. In the absence of a mechanistic understanding, a simple empirical approach to modelling the release of C14 from irradiated graphite is used. The release of volatile C14 is assumed to be in the form of methane. Its release rate is assumed to be proportional to the activity of C14 in the graphite and the constant of proportionality is estimated from experimental data.

Organic materials containing C14 may be degraded by microbial action to form a mixture of carbon dioxide and methane. The rates of microbial degradation are determined by a number of factors including the recalcitrance of the organic material and the ability of the microbial population to grow under the conditions in a GDF.

In regard to C14 transport in the groundwater pathway we have made the simplified assumption that C14 is present in inorganic form of carbonate.

The following discussion considers two mechanisms of C14 release via bulk gas and via the groundwater pathway.

4.1 *The Gas Assessment*

4.1.1 The reference case scenario

There is uncertainty about the rates at which C14 will be formed because the rate depends, first, on the release of a radionuclide from the waste matrix and, second, the incorporation of the radionuclide into a gas [17]. In developing our understanding of the consequences of gas generated in a GDF, we have made some assumptions that may lead to larger calculated rates of radioactive gas generation than might actually be the case in a GDF. It should be noted that at this stage in our programme RWM provide a high level consideration of gas migration based on several underpinning assumptions.

The reference case scenario is based on ILW/LLW in a higher strength host rock. For this reference case calculation [9], a temperature of 35°C is applied throughout the operational and post-closure phases. Metals are expected to experience a higher temperature (and therefore higher corrosion rates) during the backfilling period before closure, but the gas generated in consequence would not contribute to the post-closure risk. (This scenario therefore is conservative with respect to the consequences of gas post-closure.) Initially no water is assumed to be associated with the waste. This restricts the gas generating reactions during the operational phase, and so maximizes the post-closure inventory of gas generating materials (mainly the Magnox and aluminium), which is conservative for calculating gas generation rates after repository closure. (The Generic Documents[2] [18] assumed the waste packages were initially saturated. This assumption is not conservative for post-closure calculations as it allows additional corrosion of the reactive metals during the operational phase, leaving less remaining post-closure.)

It is assumed that gas can exchange with the atmosphere until the end of the backfilling period, and therefore the environment in the waste packages will remain aerobic until then. In practice, anaerobic niches are expected to form during this period, at least within some types of waste package. The scenario assumes that these niches occupy only a small fraction of the waste package volumes and thus have only a small effect on the overall gas generation. However, this is an assumption that warrants further examination. Carbon dioxide, including C14-labelled carbon dioxide (in a gaseous form), is expected to react with the cementitious backfill and not to escape from the repository post-closure.

The inventory peaks at 2090 AD, when all of the wastes are expected to have been emplaced. Uranium and the steels corrode aerobically releasing C14-labelled methane. C14-labelled methane is formed also from radiolysis of small organic molecules and from degradation of graphite.

The total dose to members of the public (based on a methodology using Dose Per Unit Release (DPUR) values) resulting from off-site gaseous discharges during the operational period is calculated to be 0.23mSv per year (see Table 2, below). The majority of this arises from discharges of C14. This dose is below the effective dose limit for members of the public of 1 mSv per year and may include some conservative assumptions in its derivation. It is also below the maximum dose to individuals which may result from the discharges from any single site, stated in the Environmental Permitting Regulations 2010, of 0.5 mSv per year. However, this assessment presents a dose to members of the public from carbon-14 that exceeds the design target of 0.01 mSv per year.

[2] An earlier version of our safety assessments which pre-dates the generic DSSC.

Radionuclide	Discharge (becquerels per year)	Food consumption DPUR	External irradiation DPUR	Inhalation DPUR	Dose (milli-sieverts per year)
Carbon-14	1×10^{13}	3.3×10^{-14}	6.4×10^{-20}	3.5×10^{-14}	0.180
Total					0.23

Table 2 Total dose to members of the Public from C14 as calculated in the generic Operational Environmental Safety Assessment (OESA) [19].

Figure 3, shows the results of the reference case calculation for Unshielded ILW (UILW). The facility is closed at 2150 AD and starts to resaturate. As a result, aluminium and Magnox start to corrode at a high rate. In the case of Magnox, corrosion also produces C14-labelled methane, and this is the dominant process for forming this active gas. The part of the inventory considered as Magnox plates are all corroded at 2164 AD, closely followed by the Magnox spheres at 2166 AD. As a result there is a dramatic decrease in the generation rate of C14-labelled methane. From this point on stainless steel becomes the dominant source of C14-labelled methane; corrosion of mild steel, radiolysis of small organic molecules and degradation of graphite are minor contributors. The only other noteworthy feature is at 4940 AD, when methane starts to be generated. This corresponds to the point when sulphate runs out (nitrate was all consumed by 4240 AD), and so methane can start to form. When sulphate and nitrate in the wastes are consumed methanogenesis can start (noting that the gas generation model is a simple model and so makes some assumptions in for the calculations). Initially methane is generated at a high rate from the small organic molecules (modelled as ISA) that are present, and C14-labelled methane is generated at a rate that is just less than that from corrosion of stainless steel. When these small organic molecules have been consumed, the new rate-limiting step for the formation of methane is the mid-chain scission of stopped cellulose. Methane continues to form, but at a much slower rate.

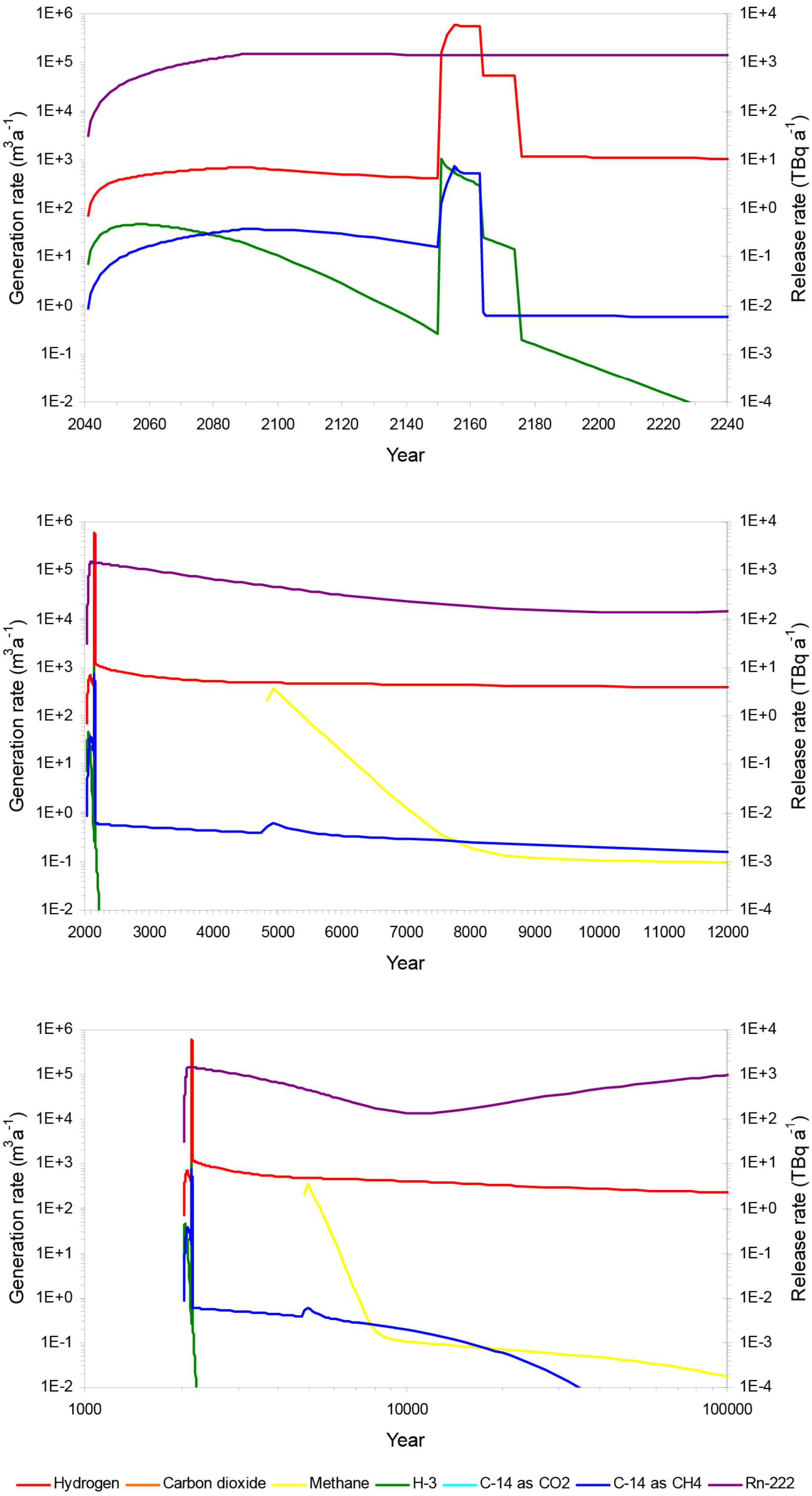

Figure 3 Gas generation from the 2004 UILW inventory for the Reference case – plotted on various time scales. The bulk gases (i.e. H2 and CH4) are plotted vs. the left-hand axis, and the active gases (i.e. 3HH, 14CH4 and 222Rn) are plotted vs. the right-hand axis. CO2 reacts with cement [9].

The behaviour seen for the gas production rate from the Shielded ILW/LLW vaults is analogous to that discussed above for the UILW vaults, except that there is no inventory of readily degradable small molecules (and only a small inventory of cellulose), so the contribution from methane production to the overall gas generation rate is much less significant for this vault type.

The migration of GDF-derived gases and, hence, the radiological impacts of gas generation, would be specific to both a site and the design of a GDF. On the basis of our work to date [1], we conclude the following with regard to gas migration in a range of geological environments:

Higher Strength Rock

In the case of a higher strength host rock, a free gas phase may form and migrate away from a GDF (rates of gas generation are unlikely to be limited by the availability of groundwater). In order to determine where the gas would migrate and when it might be released at the surface, the properties of the host rock and overlying geological formations are important. In particular, geological features (e.g. ‘cap rocks’) may act as barriers to the migration of the gas, while fault zones may or may not act as conduits depending on their ability to maintain a free gas pathway. The volume of water that is available for the gas to dissolve in is also important and would be determined by flow rate and porosity in the overlying rocks. Possible consequences of gas in this geological environment include radiotoxic (i.e. C14 labelled methane) gases to the biosphere. In contrast, an important area of our research work is to investigate the rate at which C14 labelled methane might be produced from the waste, and the potential for this gas to migrate through the geosphere.

Figure 4 shows a gas phase migrating from a GDF located at depth in a higher strength rock. The gas saturation (i.e. the fraction of the pore space in the rock that is occupied by gas) is shaded with a rainbow colour. The different types of rock are demarcated by the black lines. In this simulation, the gas moves upward through the host rock and overlying rocks until it comes to a low permeability formation with a high 'gas entry pressure' (a barrier layer, or 'cap rock'). The barrier layer forces the gas to move towards the right.

Eventually the gas finds a place where a major fault breaks the continuity of the barrier layer, and then it is able to move upwards into the more permeable, near surface rocks. There the gas encounters a large flow of groundwater, into which it dissolves and the free gas phase ceases to exist.

Figure 5 shows the corresponding plume of dissolved gas. The mass fraction of dissolved gas is shown by colour coding, with the red end of the spectrum corresponding to higher concentrations. This example illustrates that the gas pathway will be complex and specific to both a site and the design of the GDF. Recognising this, we recommend that detailed modelling of gas migration should be delayed until sufficient information on possible sites becomes available as part of a site selection process. RWM expect that a GDF situated in a higher strength host rock would be the bounding case for C14 gas migration.

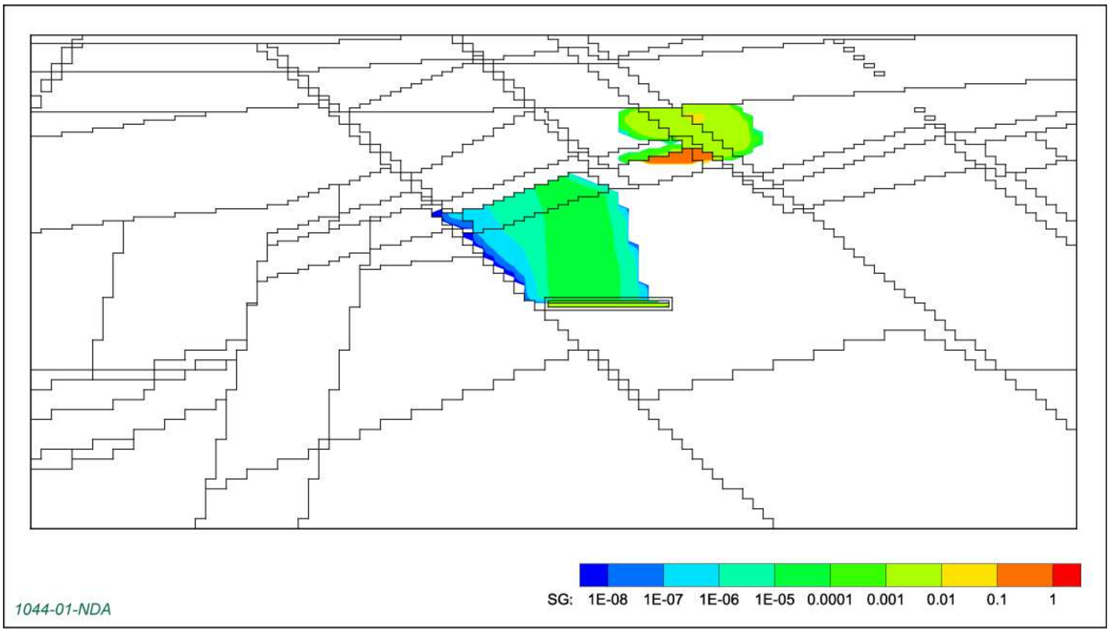

Figure 4 Contour plot of gas saturation at 240 years post-closure for gas migration from a GDF in a higher strength rock [17].

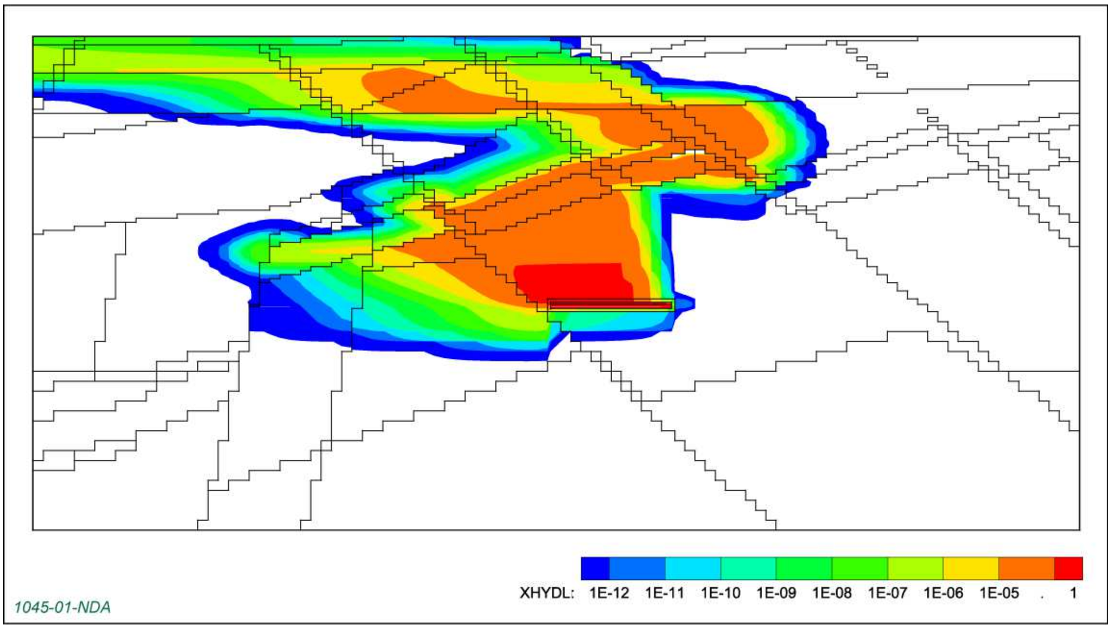

Figure 5 Contour plot of mass fraction of dissolved gas at 240 years post-closure for gas migration from a GDF in a higher strength rock [17].

Lower strength Sedimentary Rock

In the case of a lower strength sedimentary host rock (e.g. clays), the rates of gas generation may be limited by the supply of water from the host rock to a GDF. It would be difficult for any free gas phase formed to migrate from a GDF because the clay minerals typical of such geological environments have small inter-granular pores and the gas entry pressure is high [20]. Depending on the combination of gas generation rate, water inflow and gas migration in solution, the gas may be released through a combination of dilation and microfissuring in the clay. These pathways are then expected to close after the gas pressure has fallen, depending on the properties of the host rock. Possible consequences of gas in this geological environment include over-pressurisation of a GDF, with potential damage to the engineered barriers and the host rock, and the displacement of contaminated water from the disposal areas. However, this could be prevented by suitable design of the EBS and associated seals. Our assessment of such a geological environment would require investigation of the coupled processes of gas generation and multiphase flow in the vicinity of a GDF, and we are undertaking research and development work on this topic within the UK and as part of wider international research in this area [21]. The results of this work will also be relevant to developing our approaches for modelling of such coupled processes in other geological environments.

Work we have undertaken [17] on gas generation in a low-permeability host rock with a large void space surrounding the waste packages, illustrates that this scenario can reduce the maximum release rate of C14 in methane substantially. Our illustrative calculation of gas migration through a mudrock hosted environment somewhat surprisingly, found that for the assumed properties (i.e. the rates of hydrogen generation and water consumption in the vault, the void volume in the vault, and the two-phase flow properties of the host rock) a region of non-zero gas saturation around the vault grew and connected to the top boundary of the host rock before the gas pressure could increase and begin to expel water from the vault. Thereafter the system settled down to a pseudo steady-state, in which the gas leaving the model was approximately equal to the gas generated.

Evaporite Host Rock

In the case of an evaporite host rock, a GDF environment would likely be even more isolated from infiltrating water than for a lower strength sedimentary host rock. Many of the issues that arise are similar to the case of lower strength sedimentary rocks, with the one difference that an evaporite host rock will creep (i.e., move slowly under the influence of the lithostatic pressure) to a greater extent than a lower strength sedimentary host rock and fill voidage. Work in this area [17] demonstrated that a potential issue is that the generated gas, especially when combined with convergence of the rock, could flush contaminated brine a short distance away from the disposal facility. This process has been assessed by other radioactive waste management organisations, and it is suggested that the releases of radionuclides will be insufficient to compromise a safety case.

4.1.2 Sensitivity analysis

For the gas generation assessment we have undertaken four gas generation variant scenarios [9], a varying temperature case, an operational case an exclusion of carbonation case and a small organic molecules case. The results of these sensitivity analyses are discussed in more detail in [9]. However, the safety assessments [19,22] use the data specifically from the reference case.

The gas release rate data used in the generic OESA, are based on likely upper bound generation rates from the reference case calculation, including any short-lived 'spikes', for hydrogen and methane (including C14 bearing methane) relating to the corrosion of waste emplaced in the GDF after backfilling operations have occurred in the ILW/LLW disposal areas. Note that backfilling has the effect of heating the waste packages as it cures (cement curing is exothermic) and backfilling may make available free water that can be used in e.g. corrosion reactions. Emplacement of a cementitious backfill could therefore contribute to a short-lived enhancement of corrosion rates and hence gas generation rates during the operational period [19].

However, prior to backfilling of the ILW/LLW disposal areas, peak gas generation rates are expected to be at least an order of magnitude lower, no more than about 0.5×10^{12} becquerels per year for C14 bearing methane. Considering the peak annual gas generation rates during the operational period (prior to backfilling and closure), we note the following with relation to C14:

- The peak annual gas generation rate for carbon-14 is about 20 times less than the likely upper bound.

Radioactive Waste Management is undertaking additional sensitivity calculations on gas generation using 2007 Inventory data.

4.2 *The Groundwater Pathway*

4.2.1 The reference case scenario

In the 2010 generic Post-closure Safety Assessment (PCSA) [22] we present some illustrative example calculations for the groundwater pathway. In order to represent radionuclide transport by groundwater at the current generic stage of the UK Site Selection Process, we need an appropriate way of representing a generic groundwater flow field that is fit for use as part of our disposability assessment process. We have developed reference benchmark models for the PCSA in such a way that calculated doses are likely to be conservative - that is, for an actual site, they would be unlikely to be greater than those calculated using the reference models and reference parameter values. The reference model for the groundwater flow field has been developed consistent with this approach. Therefore, we have chosen to base our model representation on a possible flow field for a higher strength host rock setting, as this is the most conservative approach in terms of radionuclide transport by groundwater. Groundwater flows are likely to be greater for such a host rock than for a GDF located in a lower strength sedimentary host rock or an evaporite host rock.

We have developed high level conceptual and mathematical models to be used in the generic PCSA. The conceptual models reflect the available data and understanding of the different processes affecting the disposal system. The models have been developed to represent the different components of the system, including:

- the engineered system or near field, comprising the excavated vaults and their contents, including the waste materials, waste packaging, backfill and structural materials, and shafts and access tunnels;
- the geosphere or far field, comprising the rocks in which a GDF is constructed and those that surround them, extending to the surface; and
- the biosphere, comprising the environment accessible to humans, including the soil and surface rocks, surface water bodies, oceans and the atmosphere.

At this time in the site selection process, as a site for a GDF has not been identified simplistic assumptions have been made about the different components of the system in order to present some illustrative calculations as part of the post-closure safety case.

Our current assumption for the reference case in the generic PCSA groundwater pathway calculations is that the ILW inventory is immediately available in solution once the facility is resaturated. This is a cautious assumption. We recognise the complexity of the carbon groundwater system and are currently developing our treatment of carbon. In the generic PCSA calculations, we have continued to use the carbon dataset from GPA03 [23] in which the aqueous carbon was assumed to be present in the form of carbonate and a single value was adopted for carbon in the groundwater pathway.

The illustrative calculations presented in the generic PCSA (based on a higher strength host rock, using simplified assumptions for C14, as mentioned before) show that C14 is not a key radionuclide. Figure 6, demonstrates that the risk presented from C14 (which is not present as a key radionuclide) is below the annual risk guidance level of 10^{-6} that we are given by our regulators [24].

Figure 6 Reference case mean radiological risk against time for key radionuclides for ILW and LLW [22]

5 Key issues and priorities

Using the current modelling basis, the calculated release of carbon-14 exceeds the risk guidance level and is dominated by: corrosion of reactive metals (in the operational and early post-closure time frame); corrosion of irradiated stainless steel and leaching of irradiated graphite.

Following publication of the generic DSSC, it has been decided to adopt a collaborative approach to tackling issues related to carbon-14 by establishing an Integrated Project Team, in which the partners work together to address all aspects of the issue [25].

The overall aim of the integrated project is:

"To support geological disposal of UK wastes containing carbon-14, by integrating our evolving understanding from current and pre-existing projects, in order to develop a holistic approach to carbon-14 management in the disposal system"

The project focuses on the fate of gaseous C14 species in a fractured higher strength host rock as a bounding case. The work that has been completed in Phase 1 of the project aimed to

- summarise the current understanding, identify knowledge gaps and clarify key uncertainties, so the subsequent work in Phase 2 is appropriately focused;
- capture the understanding in a live list called the "modelling basis";
- identify the approaches, models and tools required to support assessments.

The Phase 1 work has shown, however, that there is considerable scope for reducing the calculated radiological consequence for these wastes: The inventory is based on assumed material precursor concentrations (and information from the waste producers), which may be overestimated. Work in Phase 2 that will improve the inventory. Some carbon-14 might be lost from wastes before disposal. Work in Phase 2 aims to allow these losses to be taken into account. The graphite release model is based on limited data and additional data are now available. Work in Phase 2 will update the modelling basis for release of carbon-14 from graphite, based on data arising from the RWMD programme and from other relevant work. There are no available data on carbon-14 release from irradiated reactive metals. An experimental programme to gather this data has started and will form part of Phase 2. There is only very limited data on carbon-14 release from irradiated stainless steels. An experimental programme to gather this data will be undertaken through the CAST programme.

Other priorities include, C14 gas migration in a hard fractured rock geological environment (which may be important) and the treatment of C14 in the groundwater pathway.

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CArbon-14 Source Term

CAST

Andra contribution to D6.1

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List of Contents

1 Geological Repository Concept

The classification of French radioactive wastes as a function of their management is given in Table 1.

Table 1: Classification of French radioactive waste

<table>
<tr><th></th><th>Very-short-lived (VSL)
Radioactive half-life < 100 days</th><th>Short-lived (SL)
Radioactive half-life ≤ 31 years</th><th>Long-lived (LL)
Radioactive half-life > 31 years</th></tr>
<tr><td>Very-Low-Level Waste
(VLLW)</td><td rowspan="3">VSLW
Managed first through on-site decay and then disposed of as conventional waste.</td><td colspan="2">VLLW
Disposed of at the Cires facility located in the Aube district</td></tr>
<tr><td>Low-Level Waste
(LLW)</td><td rowspan="2">LILW-SL
Disposed of at the CSA facility located in the Aube district, which took over from the CSM facility (Manche district) today monitored in post-closure phase</td><td>LLW-LL
Near surface repository (between 15 and 200 meters).
Under development.</td></tr>
<tr><td>Intermediate-Level Waste
(ILW)</td><td>ILW-LL
Deep disposal, at 500 meters, under development. Commissioning planned for 2025</td></tr>
<tr><td>High-Level waste
(HLW)</td><td></td><td colspan="2">HLW
Deep disposal, at 500 meters, under development. Commissioning planned for 2025</td></tr>
</table>

1.1 *Surface repository (CSA facility: LILW-SL)*

The LILW-SL wastes are disposed of at the CSA (Centre de Stockage de l'Aube) facility. Carbon 14 inventory comes mainly from activated materials, graphite and ionic exchange resins.

1.2 *Near surface repository (LILM-SL)*

In France, graphite waste were initially considered to be disposed of in a specific repository located up to 200 meters underground. However, recent advances in terms of radiological characterization and decontamination treatment processes have opened new management possibilities.

Two management scenarios are being assessed (see Figure 1):

- Sorting prior to disposal. In this scenario, graphite stacks would be disposed of in a near surface repository (ca. -15 m), together with radium bearing waste while other graphite waste, including sleeves, would be disposed of at the forthcoming deep geological disposal "Cigéo" (-500 m) which is planned to open in 2025,

- An alternative option is based on graphite waste treatment, i.e. selective decontamination of graphite by means of thermal/chemical processes. According to decontamination rates, decontaminated graphite would then be gasified or co-disposed of with radium bearing waste in a near surface disposal facility while extracted radionuclides would be trapped, conditioned and disposed of at "Cigéo" together with ILW-LL. The main benefits of this option are to decrease the cost of the disposal (near surface disposal facility is less expensive than deep geological disposal facility) and to improve the safety (chlorine 36 is disposed of in deep geological disposal; carbon 14 is in inorganic form).

These scenarios are being investigated from technical, safety, cost and risk points of view in order to be able to decide on the optimized management option at mid-2015, as requested by the French National Plan for the Management of Radioactive Materials and Waste 2013-2015 [DGEC & ASN, 2014].

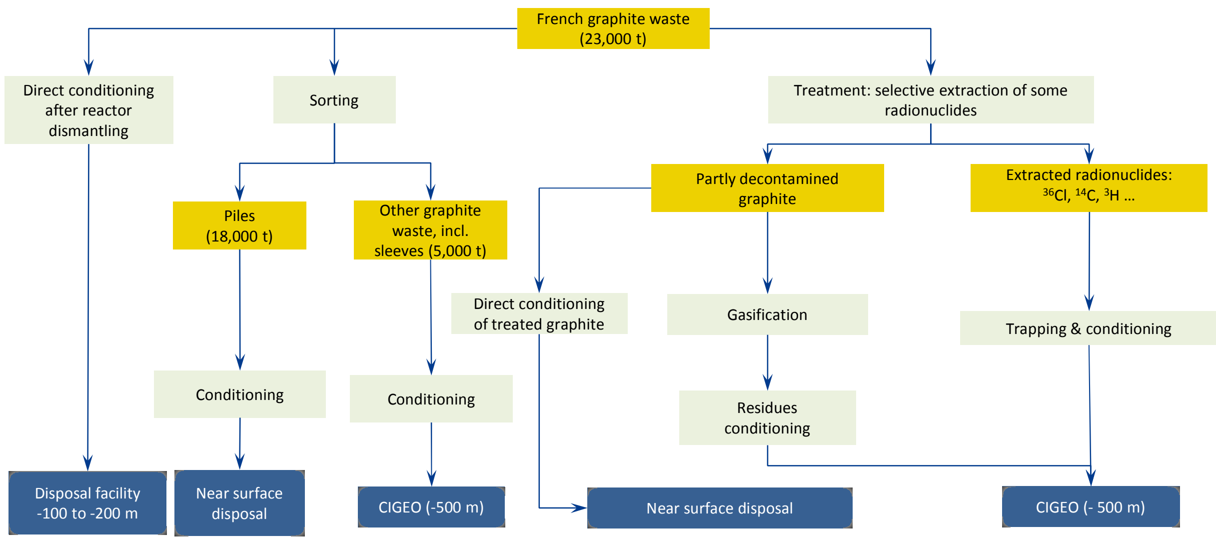

Figure 1: Management scenarios of graphite

1.3 Deep geological repository (Cigéo – ILW-LL / HLW)

Andra has studied the safety and feasibility of disposing of intermediate-level long-lived radioactive waste and vitrified high level radioactive waste in a deep geological repository in the framework of the Dossier 2005 [Andra, 2005a]. According to the 28th June 2006 Act, a reversible waste disposal in a deep geological formation and corresponding studies and investigations shall be conducted with a view to selecting a suitable site and to designing a repository. The application for the authorization will be reviewed in 2017, and subject to that authorization, the repository will be commissioned in 2025. The industrial center for disposal in geological medium (Cigéo) will be located in the Callovo-Oxfordian argillites layer.

This formation has been studied to a large extent by Andra's underground research laboratory in Bure, East of France and a series of boreholes. The Callovo-Oxfordian host rock is a claystone (argillites) consisting in quartz (25 %wt.), carbonates (30 %wt.), clays (40 %wt., mainly interstratified illite/smectite, but also minor amount of chlorite and kaolinite) and other accessory minerals (3 %wt. of pyrite). Its average porosity is 18%. The pH (7.2) is controlled by both carbonates and silicates equilibria. The redox potential is close to the hematite/goethite-magnetite buffer: -180 mV at 25 °C.

The host rock is a argillaceous layer of at least 150 m of thickness with good confining hydraulic and diffusive properties:

- The hydraulic permeability is of some 10^{-14} m/s and the hydraulic vertical gradient is less than 0.1 on average on the site,
- The host rock has significant anionic exclusion and the order of magnitude of the effective diffusion coefficient is around 10^{-12} m^2/s for anions and 10^{-11} m^2/s for neutral or cation species.

These properties imply that migration of soluble gas in the host rock is mainly managed by diffusion.

The reference design for the surface installation and the disposal areas are illustrated in Figure 2. Separate disposal areas are envisaged for IL-LL waste and HL waste to ensure phenomenological independence between them.

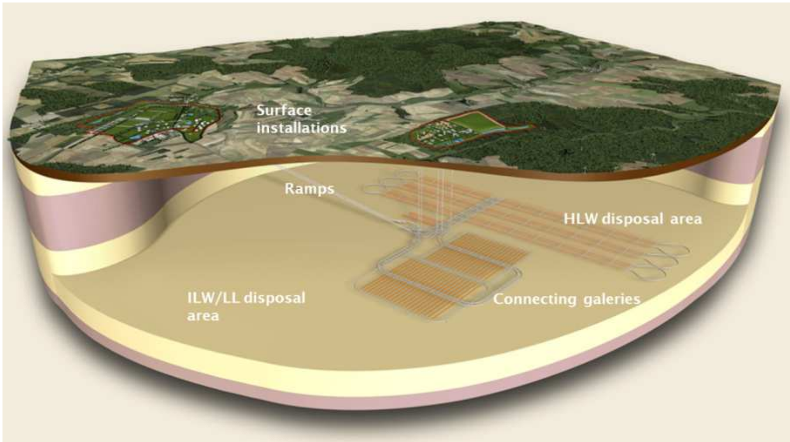

Figure 2: Schematic illustration of design for Cigéo

Upon their arrival at the facility, high-level waste packages will be conditioned one by one in disposal carbon steel containers and laid down one behind the other in tunnel-like disposal cells equipped with a carbon steel sleeve (Figure 3). The disposal carbon steel container is designed to prevent water from leaching the vitrified waste as long as the temperature is over 70 °C (less than 1,000 years) for the most exothermic ones.

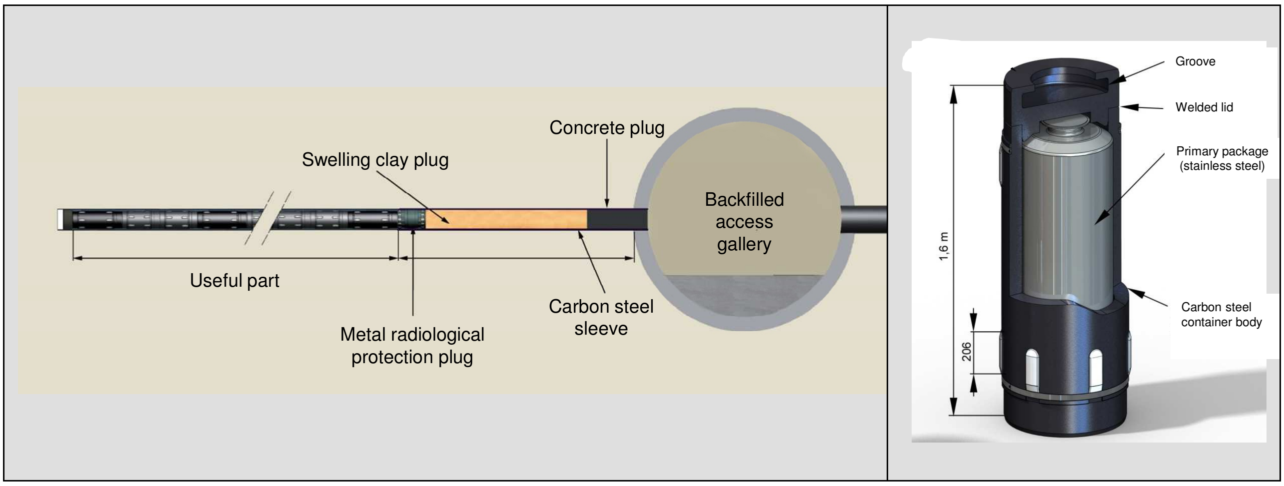

Figure 3: Schematic illustration of the design for HL waste disposal cell (left) and HL waste package (right)

Before being transferred to underground installations, intermediate-level long-lived (IL/LL) waste packages will be placed in concrete containers (disposal packages). In the repository, they will be juxtaposed and stacked in dedicated disposal cells (Figure 4).

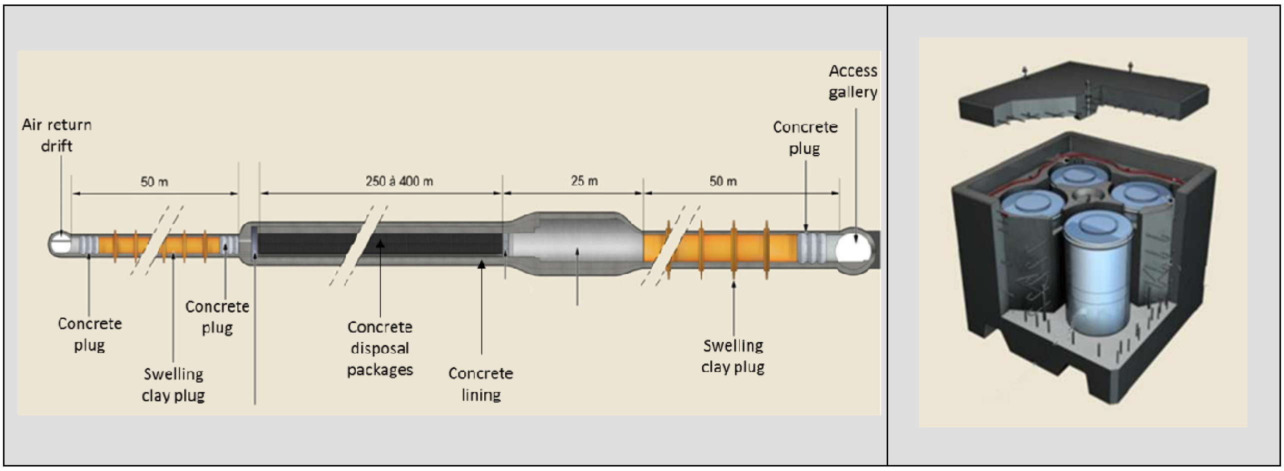

Figure 4: Schematic illustration for IL-LL waste disposal cell (left) and IL-LL waste disposal package (right)

Andra's work in the WP6 (PA) will focus on Cigéo. Accordingly, the paragraphs below will deal only with deep geological disposal.

2 Carbon 14 inventory

2.1 *Origin of the data*

The radiological inventories, together with the data related with the primary waste packages, are described by the Producers in the form of "knowledge files" dedicated to each waste family. In order to allow efficient updates and easy access for all authorized staff, Andra has developed a corporate tool which gathers the files content in a homogeneous and formalized template. However, the present level of knowledge is different from one family to another, depending, in particular, on the waste production history and background.

2.2 *Carbon 14 inventory per waste type*

A preliminary estimation of the inventory of carbon 14 is given in Table 3. The level of knowledge concerning the inventory varies from low to good. The level of knowledge is not specific to ^{14}C but to the whole inventory. A good level of knowledge may only be achieved for already produced waste packages.

Table 2: Carbon 14 inventory

Waste type	Repository	Number of packages	^{14}C activity (GBq/waste package)	Total ^{14}C inventory (TBq)	Level of knowledge
ITER	Cigéo (ILW)	~4,700	~9	~40	intermediate
Hulls and end caps (CSD-C…)	Cigéo (ILW)	~55,000	1 - 30	~1,000	Intermediate – good
Vitrified waste (CSD-V)	Cigéo (HLW)	~50,000	~3	~150	Intermediate – good
Graphite	Cigéo or LLW-LL repository			~1,000	Low – intermediate
Bitumen waste	Cigéo or LLW-LL repository	~40,000 (Cigéo) and ~30,000 (LLW-LL repository)	< 0,3 (Cigéo) < 0,03 (LLW-LL)	~5 (Cigéo) ~1 (LLW-LL)	Low
IER	Centre de l'Aube (LILW-SL)			~2	Low - intermediate

There are also other activated materials (vessel internals, control rod assemblies…) which contain carbon 14 and are mainly made of stainless steel. Their carbon 14 inventory is being consolidated.

2.2.1 ITER

ITER (International Thermonuclear Experimental Reactor) is an international nuclear fusion research project. The carbon 14 inventory relies on calculation. There are no available data.

2.2.2 Hulls and end caps

The carbon 14 inventory is assessed from the tritium inventory, which is a tracer for volatile radionuclides. Tritium inventory is correlated with cesium 137 inventory, which is measured by gamma spectrometry, as a function of burnup.

CEA, AREVA and EDF sought to determine more realistically the average grade of activated elements, including N, C and O present in the cladding tubes of Zircaloy-4 and M5^{TM} alloy [MARIMBEAU, 2004]. For Zircaloy-4, the analysis focused on over 21 years of manufacturing, more than 100,000 fuel assemblies. The compilation and analysis of results led, in France, to adopt and recommend the actual values shown in Table 3 for the calculations of activation products inventories. It is thus noted that the nitrogen content is 34 ppm in the Zircaloy-4 and 27 ppm in the M5^{TM}.

Table 3: Actual values of the contents (mass ppm) in precursors elements of 14C in the claddings in Zircaloy-4 and M5^{TM}, after [MARIMBEAU 2004]

	Actual values derived from analysis of castings or tubes (mass ppm)	
	Zircaloy-4	M5^{TM}
N	34±10	27±4
C	140±30	45±8
O	1250±170	1350±70

The determination of the amount of carbon 14 produced in the spent fuel claddings has been carried out either by calculation, using codes like ORIGEN or CESAR, or by experimental measurement. The carbon 14 specific activity is close to 50 kBq/g.

2.2.3 Vitrified waste

Carbon 14 inventory was derived from spent fuel inventory (10 % of the initial inventory) and claddings inventory (~0.1 % of the initial inventory).

2.2.4 Graphite

There are two main ways of forming carbon 14 by neutron activation in graphite: activation of the most abundant isotope of nitrogen (^{14}N) or activation of natural isotope 13 of carbon (^{13}C). The origin of carbon 14 in the graphite from the reactions ^{14}N (n,p) ^{14}C and ^{13}C (n,c) ^{14}C highly depends on the nitrogen content in the graphite under operating conditions. The method used to assess the carbon 14 inventory is based on computation and radiochemical measurements [PONCET & PETIT, 2013]. The specific activity ranges from 10^4 to 10^5 Bq/g.

2.2.5 Bitumen waste

Carbon 14 activity is always below the detection limit. Therefore carbon 14 inventory is assessed using half of the detection limit value.

2.2.6 Ionic exchange resins

The inventory was calculated using the scaling factor method based on ^{60}Co measurement.

2.3 *Building carbon 14 inventory for PA and SA*

Post closure safety assessment of Cigéo brings the necessity to take account for all the waste radioactivity content likely to have been disposed in the repository at the moment of its closure. The Nuclear Safety Authority also requests that the possible radiological impacts would not be under-estimated and that uncertainties are accounted for.

Andra has developed a methodology for building a reference radiological inventory to be considered in the post-closure safety case assessment. First step is assigning a “knowledge level” to each waste family, including the waste families for which no inventory is available at date. At last, in order to account for uncertainties, the reference inventory is estimated with application of margin factors set with consideration to the “knowledge levels”. This methodology has proved to be a good framework for exchanges between Andra and the Producers resulting in improving the knowledge of the waste.

3 Safety concept for post-closure safety

The protection of people and the environment constitutes the fundamental objectives assigned to a nuclear waste repository in deep geological formation. To conform to the assigned safety objectives and the National French Regulations [ASN, 2008], the repository is designed in order to guarantee and demonstrate the safety, during operational phase and after closure. With respect to the regulations and safety guide, the repository when closed will have to conform to safety objectives without intervention (passive safety). The concept must be designed in order to keep the impact as low as reasonable according to the state of scientific and technological knowledge and taking into account the economic and societal aspects. The concepts are designed with respect to the “defence in depth” principles.

Safety of the repository thus relies upon a series of components and design provisions with the objectives of confining and limiting the migration of the radionuclides and chemical toxics toward the biosphere. To conform to safety objectives, it implies that characteristics and performance of the components must be such that safety objectives are maintained against reasonably imaginable disturbances (internal or external) or in case of a failure of one of the components.

For Cigéo, in the Dossier 2005 Argile [Andra, 2005b], the fundamental function of protection was split into safety functions (See Chapter 3 of the safety evaluation tome):

(i) Isolating waste from surface phenomena and human intrusion which forms one of the principles of a repository in a deep geological formation. It aims at keeping waste out of reach of the populations in order to prevent them from being exposed to radioactivity. Repository is considered at depth of about 500 m in the Callovo-Oxfordian argillites.

(ii) Resisting the circulation of water. Limiting advection contributes to slow and better control of the kinetics of deterioration processes and radionuclide transfer, which can become comparable to diffusion. This function depends on the properties of the host formation (the Callovo-Oxfordian argillites). A set of design provision, including seals and overall architecture, allows those properties to be preserved and reconstituted where they have been disturbed. In that respect, design aims at limiting disturbances of the argillites.

(iii)Limiting the release of radionuclides and immobilising them within the repository. This function aims at preventing radionuclides from being placed into solution, and when inevitable, fostering their precipitation. This function is generally effective in the near field, as close as possible to the waste packages. This function is maintained during the life of the repository and takes various forms depending on the timescales and type of waste. For intermediate level waste, conditions are created in order to limit radionuclides releases, including an alkaline to neutral pH that globally favours limiting the solubility and a reducing potential. The function depends on a cement based environment. In those components, organic compounds are limited as well as other materials that can favour radionuclide migration. For vitrified waste, the first sub-function is to prevent the arrival of water in contact with the glass during the period characterised by a relatively high temperature, e.g. as long as the temperature doesn't allow to reliably account for the behaviour of vitrified glass, taking into account limit of knowledge. This function is performed by an overpack that isolates the glass from the water. After, release of radionuclides is controlled by the dissolution rate of the glass, together with design provisions contributing at maintaining diffusive regime.

(iv) Delaying and reducing the migration of radioactive nuclides. Delay means increasing the time taken to transfer them to the biosphere, which enables their impact to be significantly reduced thanks to radioactive decay. Reducing means in a dual sense, in both time and physical extent. For a given mean migration time and a constant quantity, the greater the distance covered by a radioactive nuclide flow, the less noxious it becomes and the longer it takes to arrive to the biosphere. Several favourable characteristics of the Callovo-Oxfordian argillites contribute to this function (diffusive transfer mode, sorption phenomena, precipitation, and a large thickness of sound argillites between the repository and the surrounding formations, of at least 50 meters on both sides).

For the future license application of Cigéo, a post-closure safety assessment will be conducted, upon the new regulations and safety guides since 2005 [ASN, 2008], and new scientific and technical knowledge acquired since 2005. Andra has initiated the updating of qualitative and quantitative safety analyses which will allow the definition of scenarios (normal evolution scenarios and altered evolution scenarios) and their associated assumptions for quantification of indicators. Other indicators than dose are assessed to show more clearly the repository's intrinsic performances without requiring any assumptions on the surface environment and the biosphere. The indicators allow particularly assessing the performance of individual component with respect to their safety functions (for example, molar fluxes of radionuclides reaching the roof of the Callovo-Oxfordian formation). They enable to characterize the role of the components. They allow comparing different situations in order to see which one is the most favorable with respect to the limitation of the radionuclide transfers, but they cannot be compared to thresholds.

4 Treatment of carbon 14 in safety assessment

4.1 *Treatment of carbon 14 in the 2005 feasibility assessment report*

In the feasibility assessment report [Andra, 2005b] of deep geological disposal in clay layer, the safety assessment considered the two forms of carbon 14, gas and solute, to assess the migration and potential impact of carbon 14. They were treated in two separate approaches.

The first approach considered migration of carbon 14 in solute inorganic form taking into account all the inventory of carbon 14. It was assumed that disposal was fully saturated from the beginning of post-closure. In those saturated conditions, migration by advection/diffusion/dispersion of carbon 14-solute was taking into account together with sorption coefficient and solubility limit in each porous media (claystone, concrete, bentonite). Both transfer pathways through repository structures and host rock have been quantified in the same calculation case. These quantifications have been considered for both normal evolution scenario and altered evolution scenarios (i.e. “seal failure” scenario and “borehole” scenario). For each scenario, sensitivity analyses were performed, taking into account lower value of sorption coefficients, for all the inventory of carbon 14.

Second approach assumed that all inventory of carbon 14 was likely to migrate instantaneously under gaseous form from the waste packages through the galleries and shafts, (without any exchange with clay layer under dissolved form), up to the top level of the Callovo-Oxfordian layer (roof). This very conservative assumption was chosen due to the lack of knowledge, at the time, about hydraulic transient within the repository structures, and thus about carbon 14 in two-phase flow approach, and about speciation and source term. As knowledge has significantly increased during the last decade, this type of very simplified assumption for carbon 14 migration will not be used again for further safety scenarios.

4.2 Next safety assessment of carbon 14 for the license application of Cigéo

Next assessment of Cigéo will consider the increase of knowledge on inventory and speciation of carbon 14 and its source term, better and more precise understanding of 2-phase-flow at all scales, and the use of a large data set derived from intensive scientific program over the last 10 years. Specific assumptions for performance and safety assessments will be derived from this knowledge and the residual uncertainties at this stage of the program. It is however envisaged to consider the treatment of both gaseous and solute pathway with a 2-phase-flow approach. Previous to the safety assessment, simulations are conducted considering many sensitivity studies (models and input data) deduced from uncertainties on speciation, organic/inorganic part, retention for inorganic part, 2-phase flow input data such as corrosion rate, hydro-dispersive parameters from host rock… Choices relative to the evaluation of the carbon 14 transfer in the normal evolution scenario will be made in order to maximize each pathway. As such, for example for the gaseous phase, conditions that maximize re-saturation time and migration of carbon 14-gas to the shafts will be considered.

As mentioned in the previous section, Andra has initiated the updating of qualitative and quantitative safety analyses which will allow the definition of scenario and associated hypotheses and conceptualisation, including gaseous and solute forms of carbon 14.

4.2.1 Phenomenological results to be used in the definition of safety scenarios for the migration of carbon 14

In order to be able to define safety scenarios regarding carbon 14 migration different phenomenological numerical simulations were undertaken at global repository scale. The main phenomenological processes taken into account are as follows:

- Generalized Darcy two phase flow equation for water and gas convection
- Fick's law for water and gas diffusion

- Henry's law for state changes (from gas to solute or vice-versa)

Due to the transfer properties of the Callovo-Oxfordian claystone (cf. § 1.3), migration of soluble gas in the host rock is mainly managed by diffusion. High diffusive travel time (i.e. several hundred thousands of years to reach the host rock upper or lower boundary from the repository) also implies a very low level of soluble gas at top and bottom of the clay layer. It means that significant amount of dissolved carbon 14 never reaches the surrounding aquifers through the host rock as it has disappeared by radiological decay earlier. Rapid carbon 14 gaseous flow to the upper aquifer may only occur through the repository drift system.

The main gas produced in the repository is hydrogen generated either by water or polymer radiolysis or by corrosion; it is this gas that governs the extension of desaturation process in the whole repository and the migration of carbon 14 mimics the extension of the H_2 gas phase. Just to fix the order of magnitudes, the total number of moles of H_2 produced in the repository is of several billions whereas the total number of moles of carbon 14 produced in the repository is of several hundred at most (based on the 2009 inventory).

As an example the following paragraphs explicit some results produced for the 2009 architecture version of Cigéo, thus the description depicts only a general behaviour and the values presented below have to be considered only as orders of magnitude.

For the exercise done in 2009, carbon 14 source term (only inorganic form) is based on congruent release with glass dissolution (HLW) or zircaloy corrosion (ILW-LL). The first release of carbon 14 takes place in ILL-LL waste zone (Figure 5, MAVL zone). It is also this waste zone that produces the majority of the carbon 14, the HL waste zone (HA zone in Figure 5) accounting only for some percent in the total generation. The arrival of carbon 14 at shafts level (located between IL-LL waste zone and HL waste zone) and eventually in the upper Oxfordian aquifer begins after several hundred years with a maximum at around 20,000 years, and represent only a very small part of the total amount of carbon 14 produced in the repository (less than 0.1 %, see Figure 6). Most of the carbon 14 is disappearing by radiological decay in the Callovo-Oxfordian water while moving under dissolved form toward the surrounding aquifers (only an even smaller part reaching these aquifers after migration through the host rock).

These results are representative of Andra's understanding of carbon 14 from the waste storage zones toward the geosphere, but have to be updated using the final architecture that will be retained for the DAC Dossier (construction licence application), the latest inventory (carbon 14 inventory has drastically increased since 2009) and the latest values for phenomenological parameters, mainly host rock permeability, host rock diffusion coefficient and corrosion rates (used to assess the H_2 flux production).

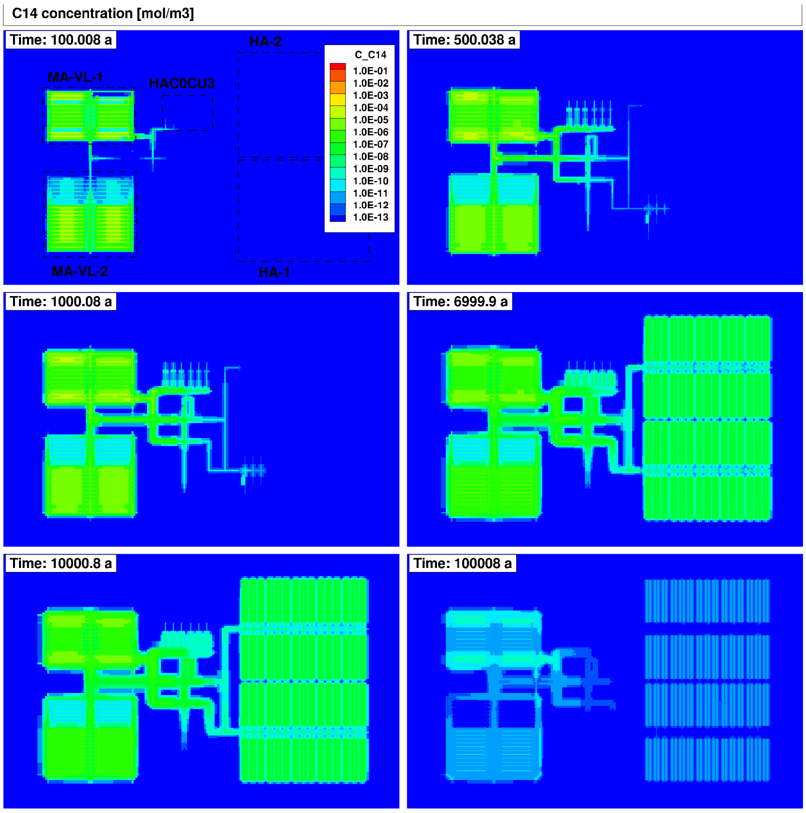

Figure 5: Example of a possible evolution with time of concentration of carbon 14 at repository level (2009 repository architecture)

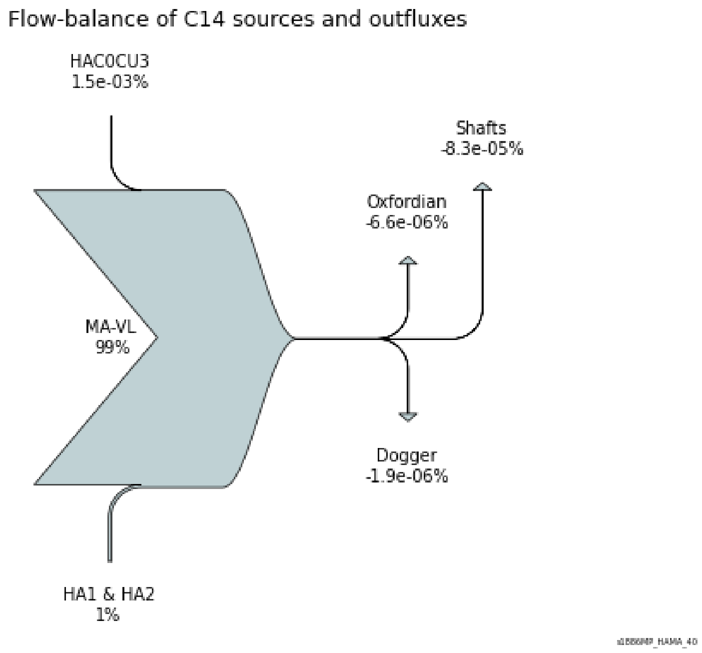

Figure 6: Chart of a possible carbon 14 flows (2009 repository architecture)

5 Key issues and priorities

In the 2005 feasibility report no critical issues regarding carbon 14 migration and impact were determined. Nevertheless, in the forthcoming safety report, carbon 14 migration and impact will be assessed using as much as possible of the phenomenological knowledge gained during the last decade and the main uncertainties, and therefore outstanding issues regarding to carbon 14, are the following:

- Carbon 14 inventory and speciation (graphite, stainless steel, zirconium alloy, ionic exchange resins…),
- Organic species migration both in concrete and in claystone,
- Improvement of understanding on two-phase flow carbon 14 migration for all disposals.

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CArbon-14 Source Term

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ONDRAF/NIRAS contribution to D6.1

M. Capouet & D. Boulanger

Date of issue of this report: 08/09/2015

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1 Geological Repository Concept

ONDRAF/NIRAS, the Belgian Agency for Radioactive Waste and Enriched Fissile Materials, is considering the safety and feasibility of disposing of low level and intermediate-level long-lived radioactive waste (category B waste), and vitrified high level radioactive waste and spent fuel (category C waste) in a geological repository excavated in poorly indurated clays. The reference host rock, on which ONDRAF/NIRAS focuses its RD&D is the Boom Clay formation. Boom Clay consists of an alternation of silty clay and clayey silt, with a high content of pyrite and glauconite in the silty layers. Bands differ by (limited) variations in grain size, organic matter and carbonate content. The carbonates regulate the pH (8.3) while the pyrite controls the redox potential (-270 mV). The reference design for the disposal of category B and C wastes in Belgium is illustrated in Figure 1. Separate disposal areas are envisaged for category B and C wastes, which would be excavated and operated sequentially. Ypresian Clays are also investigated as an alternative host rock.

Primary waste packages will be emplaced into pre cast concrete disposal waste packages: the “supercontainer” for category C waste and the “monolith” for category B, examples of which are illustrated in Figure 2. In the supercontainer, containment is achieved by placing the primary waste container in a carbon steel overpack surrounded by a Portland Cement concrete buffer. An outer stainless steel envelope may also be placed around the buffer. The supercontainer is designed to provide complete containment of radioactivity at least during the thermal phase. This corresponds to the period over which the performance of safety-critical disposal system components, primarily the retardation capability of the Boom Clay, could be more difficult to substantiate due to the complex, coupled processes associated with the thermal transient. The thermal phase is expected to last for at least hundreds of years after emplacement of vitrified HLW, and up to a few thousand years for spent fuel.

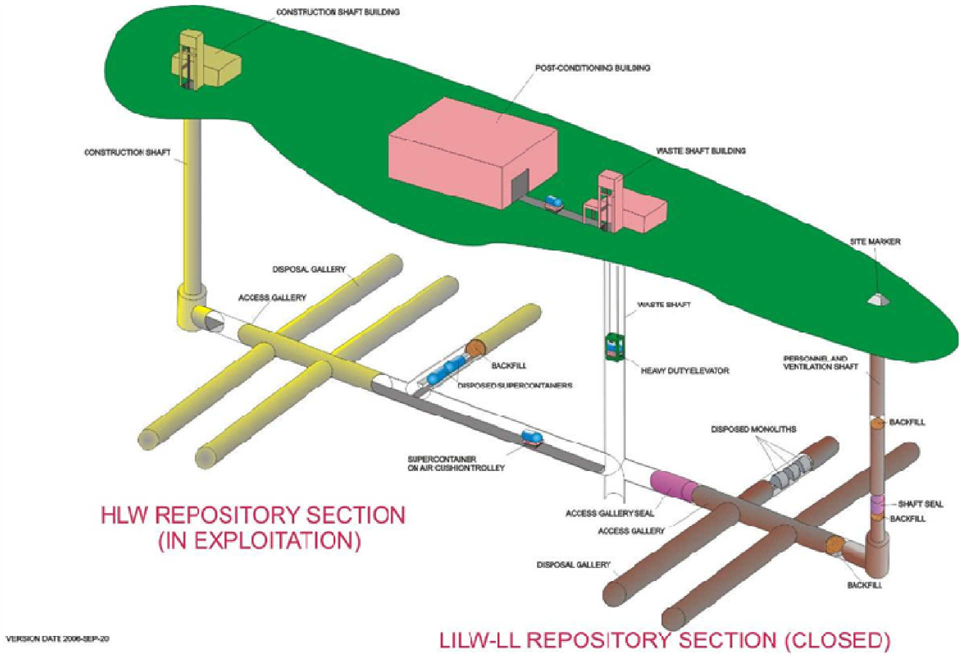

Figure 1 Reference design for the disposal of category B and C waste in Belgium

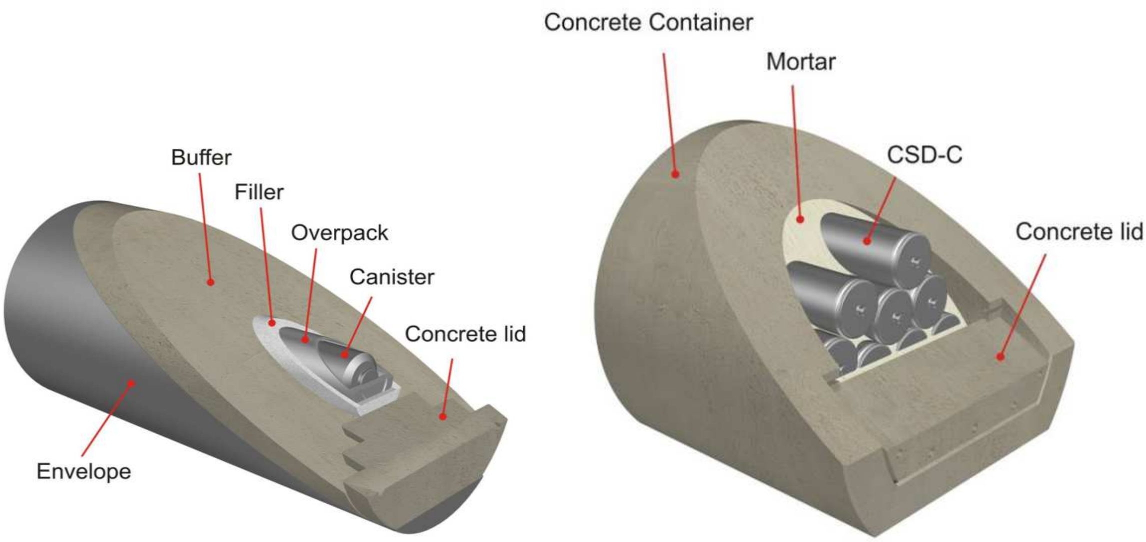

Figure 2. Example of a supercontainer and a monolith design for vitrified HLW canisters and compacted waste ('CSD-C'), respectively.

Category B waste does not generate significant amounts of heat, but does not meet the radiological criteria to be compatible with surface disposal. Generally, these wastes have been encapsulated in cement, or immobilized in a bituminized matrix. The primary waste containers are emplaced in large pre-cast concrete containers and the void space is filled with mortar. A concrete lid is emplaced to close the monolith. The monolith is designed to facilitate contact handling during emplacement, but has no specific post-closure safety function assigned to it.

2 C-14 Inventory

A preliminary estimation of the C-14 inventory of the main waste families of the Belgian programme relevant for long term safety is given in the following table. The CSD-C family shows the most important inventory of C-14 with a total activity of 1.9E14 Bq in the case a scenario of full reprocessing of spent fuel is considered (average burn-up is about 47 GWd/tHM, PWR). The inventory is estimated by ONDRAF/NIRAS assuming the maximum of the tolerance permitted by the waste producer for the content of nitrogen (~80 ppm for Zry4). To date no other sources of C-14 are considered like those eventually associated to surface contaminations. Furthermore, the production yield remains theoretical and is not supported by measurements. Therefore the conservative nature of this estimation must still be confirmed. C-14 inventory of the core internals resulting from the dismantling of the Belgian PWR's amounts a total of 2E14 Bq, resulting in a best estimate of 5E8 Bq/kg of stainless steel. On the other hand, an activity of 1E7 Bq C-14/kg for the resin waste originating from the primary circuits of PWRs is reported. The chemical speciation of carbon sorbed on these resins is an unknown as well as the impact of the conditioning processes (drying) thereon. The inventory for the core internals and the resins are both estimations provided by the waste producers and must still be confirmed. The inventory of the high level waste is as follows: In the scenario of a full reprocessing of spent fuel, the C-14 inventory amounts 1.5E10 Bq/canister of CSD-V. It corresponds to an average burn-up of about 47 GWd/tHM. In the alternative scenario that the reprocessing moratorium is maintained, the C-14 average of UOX is 5.6E10 Bq/assembly (75 ppm of nitrogen is assumed). The claddings are mainly made of M-5/ZIRLO (1/4) and standard zirconium alloy (3/4). C-14 is also present in Be-waste and bituminized waste but in a less important amount.

Table 1: C-14 Belgian inventory as for 2010

Waste type	C-14 activity (Bq/unit)	Total activity (Bq)	Uncertainty
CSD-C	8E7 Bq/kg metal	1.9E14	Conservative, to be confirmed
Core internals	5E8 Bq/kg metal	2.0E14	Best estimate for the most activated parts, uncertainty within one order of magnitude
Resins (PWR primary circuit)	<5E7 Bq/kg resins		Best estimate, uncertainty more than one order of magnitude not excluded.
CSD-V	1.5E10 Bq/canister	4.7E13	Conservative, to be confirmed
Spent fuel total	5.6E10 Bq/assembly 12 feet	6E14	Conservative, to be confirmed
Fuel	4.1E10 Bq/assembly 12 feet		
Struct. parts	5.1E09 Bq/assembly 12 feet		
Claddings	1.0E10 Bq/assembly 12 feet		

3 Safety concept

The safety concept for the category C waste is illustrated in Figure 3 and described as follows [ONDRAF/NIRAS, 2013]. The repository is constructed at depth in Boom Clay. The geological formation is stable and no human or natural events will alter the isolation (*Safety function I*) provided to the disposal system. The containment of radionuclides and other contaminants within the overpack lasts till at least the end of the thermal phase. The Boom Clay pore water will diffuse slowly into the EBS. Eventually, it will start to corrode the envelope (if present), then the overpack and finally the primary waste packages. The containment safety function (*C*) is assigned to the overpack only, although it is recognized that the envelope (if present) and the primary waste packages can extend the containment phase. After perforation of the overpack, the waste will come into contact with the pore water, begin to dissolve and release the contaminants which will diffuse through the near field into the Boom Clay host formation. Some waste matrices can dissolve very slowly and be characterized sufficiently and in a robust way to be assigned a safety function (*R1*). The Boom Clay around the repository will be disturbed by the excavation, construction, operation and post-closure evolution of the repository. It is considered that the spatial extent of these perturbations remains limited to a few meters and is bounded. Transport is diffusion-dominated and migration is further delayed by retention processes (*Safety function R2 and R3, respectively*). After the slow diffusive transport through the Boom Clay formation, during which a large fraction of the radionuclides will have decayed, only a minor fraction will reach the biosphere.

The safety concept for the category B waste is similar to the one developed for C. However, no containment is foreseen. Also, in most cases, no safety function is assigned to the waste matrix.

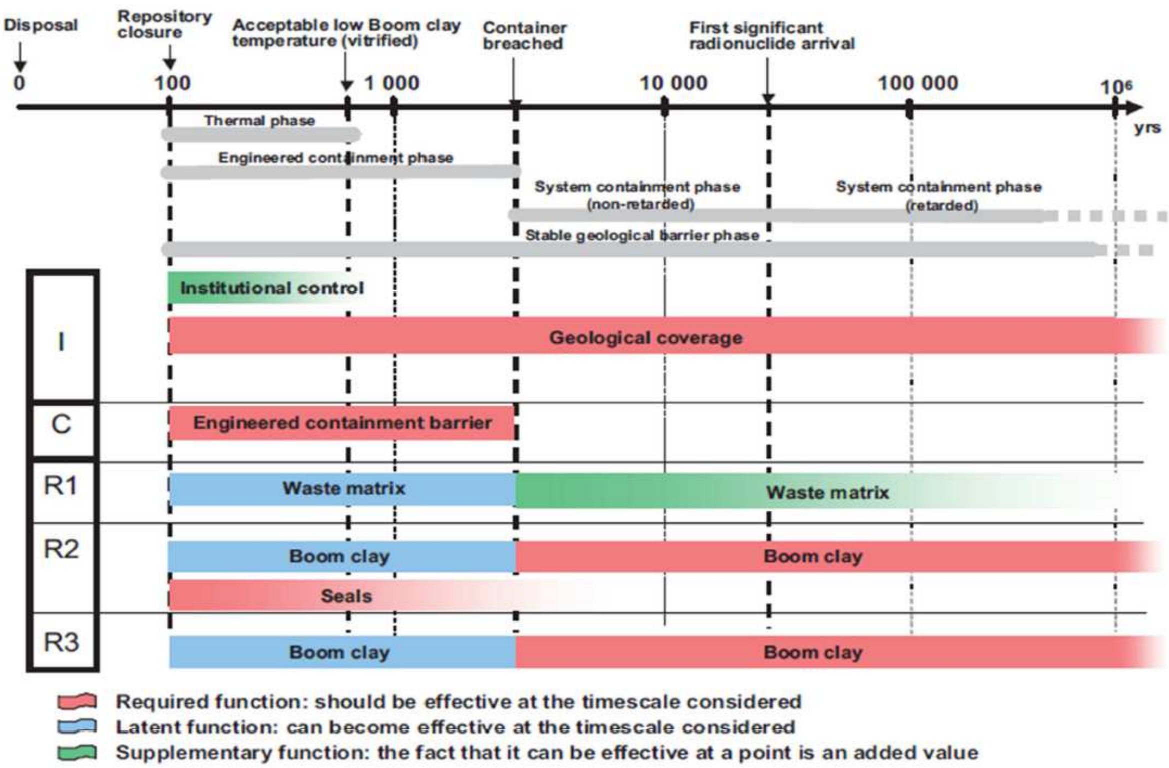

Figure 2 Relationship between safety functions and time for the disposal of spent fuel and high level waste. I = Isolation, C = Containment, R= retardation (time periods are illustrative) [IAEA, 2011].

4 Treatment of C-14 in safety assessment

According to the ONDRAF/NIRAS methodology, a scenario is a set of high-level descriptions of possible evolutions of the disposal system, in a simplified, abstract form. These high-level descriptions share a common time-deployment of the safety functions. A scenario can be represented by different realisations of the system evolution with a specific set of parameter data and model hypothesis. In the reference scenario, safety functions are operating as expected. In the altered-evolution scenarios, one or more safety functions are considered as partially or fully impaired. In the reference case, the system is assumed to be implemented according to the specified design. The assumptions behind this case tend to be conservative. Alternative cases represent different assumptions (e.g. less conservative parametrisation, design options).

4.1 *Spent fuel*

The reference scenario for spent fuel is representative of the impact of the high-level waste. Following the reference scenario, the containment lifetime is assumed to last tens of thousands of years because of the very low passive uniform corrosion rate imposed by the alkaline conditions of the cemented buffer. It is conservatively assumed that all overpacks breach at the same time. The C-14 inventory is provided for the three main parts of the spent fuel assembly: The fuel matrix (UO2 pellets), the structural parts and the claddings. In safety assessment, C-14 is assumed to be homogeneously distributed in each part. The IRF associated to spent fuel includes 20% of the C-14 inventory of the claddings and 15% of the C-14 inventory of the spent fuel matrix. The high ratio adopted for spent fuel comes from 1/ The pessimistic value of 10% proposed by Johnson et al. (2005) and 2/ A supplementary IRF term (5%) for C-14 due to alpha-self irradiation enhanced diffusion. The IRF for the cladding corresponds to the ratio of the oxidized layer over the section of the cladding for a burn-up of about 60 Gwd/tHM. Since the averaged burn-up of the spent fuel assemblies of the Belgian program ranges between 30 and 55 GWd/tHM, applying this IRF on the whole C-14 inventory contained in the claddings is a conservative hypothesis. Furthermore the longitudinal variations of the oxidized thickness due to the burn-up are also accounted for with this high ratio. However future work should confirm if this ratio is conservative with respect to the observed heterogeneous distribution of C-14 between the zircone and the core of the cladding due to surface contaminations.

In the last safety assessment, carbon is assumed to be in an oxidized form in the waste matrices and transported consequently in the clay environment under the form of bicarbonate. The EBS contains large quantities of cementitious materials, whose properties evolve with time. The dissolved carbon in cement pore water is considered to be controlled by the solubility of calcite related to the dissolved calcium concentration which in turn depends on the solubility of Ca compounds such as Portlandite and C-S-H phases. On-going RD&D tends to show that clogging of the concrete buffer will occur [ONDRAF/NIRAS, 2013]. However, adequate support to these evidences is currently insufficient, and this clogging is not taken into account in safety assessment resulting in the conservative assumption of unretarded release for all radionuclides. Experiments on carbonate transport in Boom Clay reveal that it is poorly retarded (no observable isotopic exchange at the scale of the laboratory experiment). In consequence, Carbon-14 is assumed to be not retarded in the safety assessment calculations (either in clay or in cement).

In the case of category C waste, temperature elevation and irradiation of the different concrete layers lead to high temperature and radiations for several decades. More research is under way to assess the thermo-hydromechanical (THM) evolution of the buffer concrete, to confirm in particular the absence of through-going cracks. A high diffusion coefficient (one to two orders of magnitude higher) is assumed in the SA calculations to account for these uncertainties.

Intake to C14 is due to ingestion of water from a well located in the aquifer above the geological system and from contaminated food from the agriculture. It is assumed that the C14, introduced in the root zone soil from contaminated irrigation water (or upwelling of contaminated groundwater) is released in the air by gaseous emission as carbon dioxide, which is then incorporated by the plant via photosynthesis, resulting in increased C14 in the crops.

The very long containment time of the safety concept associated to the diffuse transport through the host rock yields a reduction of the C-14 radiotoxicity as high as 10^{12}, resulting in an entire decay in the primary package [Govearts et al., 2012].

Sensitivity analysis of the transport of C-14 were made by considering hypothetical retardation factors: a weak sorption on clay (R = 2) and a more considerable to strong sorption (R = 100, 1000 and 10 000) on concrete [Raeymaekers et al., 2008]. The corresponding C-14 fluxes into the upper aquifer are shown in Fig.3. It can be seen that even a small increase in the retardation factor of clay has a noticeable impact on the flux. For sorption on concrete much higher retardation factors are required to influence the flux of C-14 into the aquifer. This observation reflects the importance of the clay barrier for the performance of the repository.

4.2 CSD-C

The assessment of compacted waste was performed with the same model parametrisation at the exception that 1/ No containment time had been considered, 2/ a congruent corrosion release of the claddings was assumed, resulting in CSD-C matrix lifetime of the order of 25000 years, and 3/ No IRF was considered. Although this nuclide has the highest dose after Cl-36, Nb-94, Se-79, Tc-99, its contribution to the total dose remains very small (< 0.001) [ONDRAF/NIRAS, 2001; Weetjens and Govaerts, 2012]. Assuming a more conservative calculations (eg: Assuming an IRF) would not change the results in terms of radiotoxicity.

4.3 Gas conversion

The impact of a possible conversion of C-14 in gas in the near field environment has also been investigated. Assuming that the whole C-14 inventory contained in a supercontainer is instantaneously released in the form of methane would contribute for a negligible part to the hydrogen production from the metallic corrosion of the supercontainer and the primary package expected in the long-term. [Yu and Weetjens, 2012] showed that the total gas production from a high-level waste should be well below the maximum diffusive capacity of the host rock. For some B-waste (e.g. CSD-C), the production of gas might well be beyond this threshold. RD&D is on-going to reduce the uncertainty related to the corrosion rates of the diverse metallic parts. If necessary, design solutions will be implemented to organize the storage and transport of gas through the disposal system in a non-disruptive manner. If this alternative speciation is plausible, C-14 might be a safety issue in the case of an accelerated transport through the disposal system to the surface. This altered scenario where all safety functions are bypassed have been analyzed in SAFIR 2 [ONDRAF/NIRAS, 2001] in the context of a poor-sealing scenario. Sensitivity analysis has shown that the impact was dependent of the importance of the advective flux and the length of the transport path.

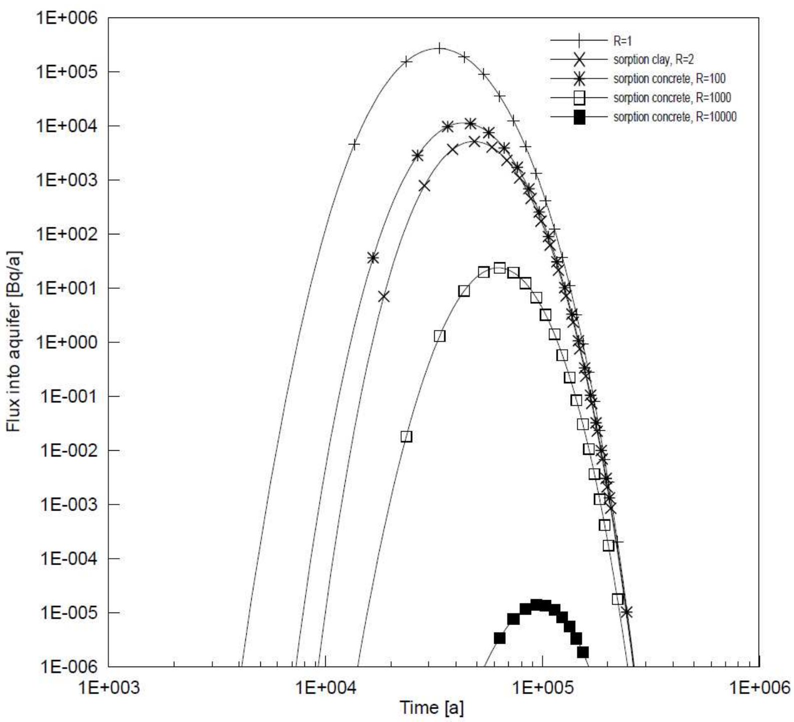

Figure 3 C-14 flux into the aquifer for different retardation factors in clay and concrete

5 Key issues and priorities

The speciation of C-14 in the Belgian inventory is largely uncertain. Regarding its inventory, resin waste show uncertainties of several order of magnitude. For the other waste types, realistic values can be approached provided additional production pathways by activation or surface contamination are identified or discarded. Should C-14 be released in a carbonate form, the impact on radiotoxicity should not be critical. Should C-14 be in a gas form, then the picture could be different in the case of premature release scenarios of B-waste such as CSD-C. Within CAST, sensitivity analysis will be performed to capture more precisely in which conditions an accelerated transport of C-14 bearing flow could be critical.

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RATEN ICN contribution to D6.1

Daniela Diaconu and Crina Bucur

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1 Romanian Concept for Geological Repository

The current Romanian Strategy on Medium and Long Term Management of Spent Nuclear Fuel and Radioactive Waste, including the Disposal and Decommissioning of Nuclear and Radiological Facilities elaborated in 2004 by the national waste management organization called Nuclear Agency and for Radioactive Waste (Agentia Nucleara si pentru Deseuri Radioactive – ANDR) is considering the geological disposal of Spent Fuel and long-lived radioactive waste, without retrievability. A Deep Geological Repository (DGR) shall be available by 2055. The DGR facility will dispose both spent fuel and long lived wastes from the Cernavoda Nuclear Power Plant (NPP). The non-retrievable facility will be located at 500 - 1000m depth. In horizontal emplacement rooms where spent fuel (SF) and the Long lived-Intermediate Level Waste (LL-ILW are placed in locations separated by a buffer area of non excavated material.

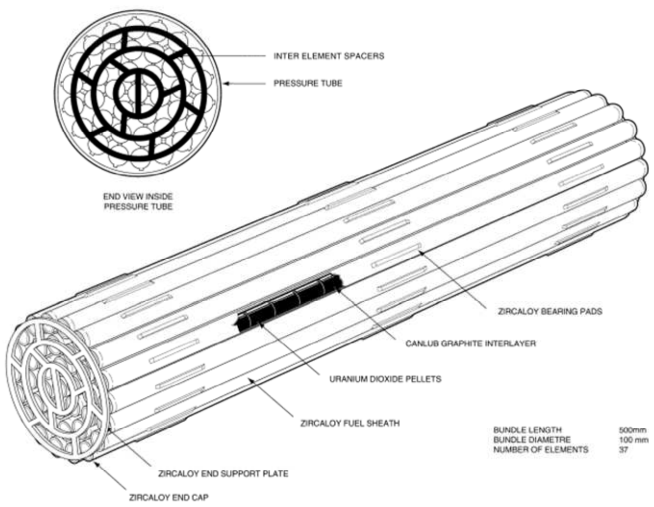

Figure 1. CANDU Fuel Bundle

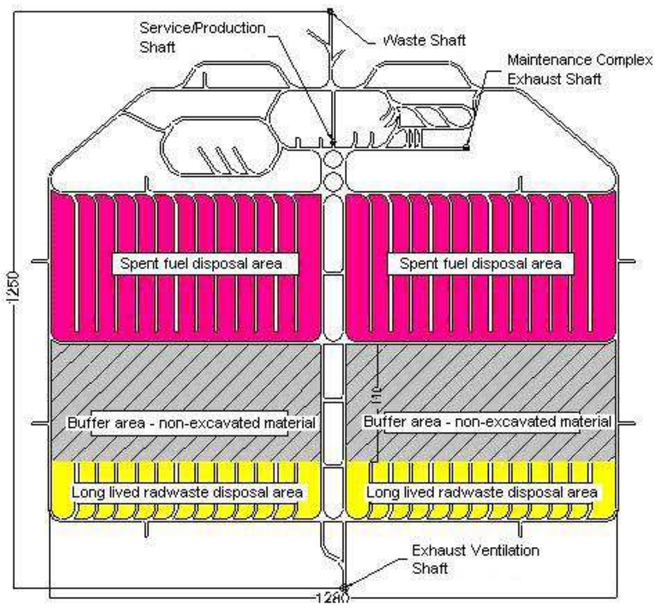

Figure 2. Generic concept of the SF and LL-ILW

CANDU spent fuel bundles (Figure 1) with 0.5m lenght, 0.1m in diameter and a weight of 24 Kg consist of 37 fuel rods of Zy-4 containing natural UO_2 pellets.

The only conceptual design of the repository available at this moment was developed for preliminary discussions and cost estimations. It is based on a multi – barrier approach and on the Canadian concept (Figure 2). The basic elements are:

- copper/steel double-shell containers for spent fuel encapsulation (each container accommodating 360 bundles [1]) as illustrated in Figure 3.
- reinforced concrete containers for LL-ILW
- buffer material surrounding the spent fuel/LL-ILW package (bentonite)
- seals and plugs to isolate disposal galleries from the transport and access galleries (bentonite/concrete)
- backfill material to fill the transport and access galleries (bentonite /sand mixture);
- buffer area between spent fuel and LL-ILW (non-excavated host rock)
- host rock – assumed to be either granite or clay.

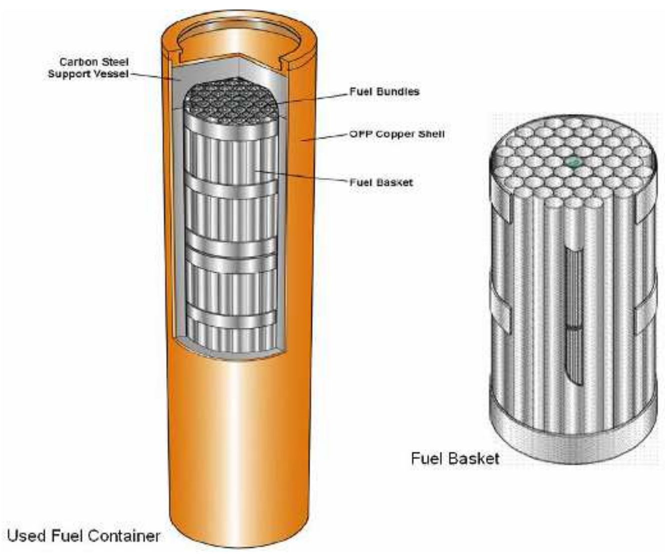

Figure 3. Disposal canister for CANDU SF

The host rock for geological disposal is not yet established. However, 6 geological formations were identified as potentially favourable to host the geological repository, namely: green schist, granite, basalt, clay, salt and volcanic stuff.

An Underground Research Laboratory is foreseen to be constructed in advance after the host rock is selected.

No detailed design and characteristics of the waste packages, waste forms, and engineered barriers are currently available.

2 C-14 Inventory

A preliminary estimation of the inventory of C-14 is given in **Table 2-1**. The main waste streams significantly contributing to the C-14 source term are:

- CANDU Spent fuel
- Non fuel contact spent resins from the moderator purification system and primary heat transport system
  - C-14 in non-fuel contact resins was estimated based on Canadian literature data for CANDU 6 rector [2].
- Activated materials from re-tubing and decommissioning (pressure tubes, Calandria vessel, Calandria tubes, cal end-feedings, etc)
- Graphite from the Thermal Columns of TRIGA 14MW and VVER research reactors

There are no measurements or estimations for C-14 content in the spent fuel, exchange resins, or activated materials arising from Cernavoda NPP; the values considered at this moment are conservatives. They are based on Canadian literature data [2, 3] and represents ORIGEN simulation results [3] for the activated materials and experimental measurements for SIERs.

Nitrogen impurity level in Zy-4 in the fuel bundles ranges between 10-40 ppm (as measured on the Zy-4 used in CANDU fuel fabrication in Romania). There are no available data for N2 content in UO2 ¬and Zy-2, and Zr-2.5%Nb.

C-14 in irradiated graphite from the thermal columns of TRIGA research reactor operating in the Institute for Nuclear Research Pitesti was estimated in CARBOWASTE project based on ORIGEN simulations coupled with experimental measurements [4]. C-14 amount and inorganic and organic content in the irradiated graphite of the thermal column of VVR-S research reactor were measured by IFIN HH [5].

Excepting the estimations for the C-14 in TRIGA and VVR-S reactor graphite for which previous work has been done [4], all other values have a high uncertainty.

C-14 speciation was not determined or considered for any of waste types mentioned above.

Table 2-1 Inventory of C-14 for the main waste streams in Romania (2 CANDU units, operating 40 y, including re-tubing and decommissioning and research reactor irradiated graphite)

Waste type	Amount	C-14 activity (Bq/Kg)	Uncertainty wrt C-14	Comments
Fuel Bundles* 2 CANDU units, 40y Burn up 7500Md/tU	~486500 FB (~9340.8 tU)	5.7 E+6 to 2.03E+7 (Bq/KgU)	High	Measured/ Estimated (ORIGEN simulations)
Ion Exchange Resins resins	272 m^3	~3.45E+09	High	Measured (on moderator SIERs)
pressure tubes: -Zr-2.5%Nb	92.720 tonnes	~1.51E+09 (Bq/kg metal)	High	ORIGEN simulations
Calandria tubes - Zy 2	34.96 tonnes	Not available	-	
Core internals (end-feedings, etc.) – stainless steel	1420.288 tonnes	Not available	-	
Graphite from Thermal Column TRIGA reactor	~1.5 m^3 (2.5 tonnes)	Ranging from 1.1E+7 to 1E+4 (Bq/kg graphite) (total 4 to 5E+10)	Moderate	Measured
Graphite from Thermal Column VVER reactor	5.3 tonnes	10^{10} – 10^{11} Bq	High	Measured Anorganic componds – 18 GBq Inorganic componds – 9 GBq

3 Safety concept

A systematic safety concept of the geological disposal in Romania is under development, according to the national standards and regulations, and based on the European experience acquired in more advanced programs. The safety functions considered for the generic concept of geological disposal in Romania are [6]:

-*physical containment: ensured by a waterproof barrier able to isolate the radioactive waste from groundwater during the first phase after repository closure; no radionuclides release can occur from the waste form as long as this barrier is effective; it prevents dispersion of radionuclides during the transient initial phase of the repository when re-saturation, heat release, strong radiation, pressure rebuilding occur.*

-*slow release: after container failure, when groundwater comes in contact with the conditioned waste, leaching of radionuclides from the waste matrix starts in combination with the degradation of the waste matrix; various physico-chemical processes, such as corrosion of and lixiviation from the waste matrix, precipitation, sorption or co-precipitation strongly limit the radionuclide releases into the surrounding layers;*

-*retardation: the radionuclides dissolved in the groundwater that is in contact with the waste will start to migrate through the bentonite buffer and the host formation; because of the very low groundwater fluxes in potential host formations, this transport will be very slow; furthermore, many radionuclides will be sorbed onto minerals of the buffer and the host formation; retardation delays the releases and drastically limits the amounts of radionuclides that are released into the biosphere per unit of time;*

-*dispersion and dilution: once the long-lived radionuclides leave the repository's barrier system, they are released into the overlying or surrounding aquifers and eventually into the accessible environment; the dispersion and dilution processes in* *the aquifers and surface waters will further reduce the radionuclide concentrations in the waters that are directly accessible by man.*

4 Treatment of C-14 in safety assessment

There is no safety assessment evaluation performed yet, even at generic level, for deep geological disposal as part of a national program. Since the updated waste management strategy and the implementation program for geological disposal will be finalized by May 2015, a systematic development of a generic safety case, including a preliminary safety assessment will be produced.

5 Key issues and priorities

In this context, the key issues are to build up a generic model of the near field and a performance assessment using data available before the start of this project to be used as a baseline for the evaluation of improvements brought by data achieved under CAST by the RATEN ICN experimental work. In particular we will look for reducing uncertainties in C-14 inventory and estimation of C-14 speciation impact on near-field performance assessment.

The steps considered are:

- Define the expected-evolution scenario including C-14 release and migration as organic and inorganic species;
- Build a generic conceptual model for performance analysis of the near-field (including parameterisation of the safety barriers);
- Define C-14 source term based initially on the available literature data, transport model in the near-field, including different behaviour of C-14 organic and inorganic species;
- Identify and assess the uncertainties related to C-14 inventory and speciation expected to be reduced by the outcomes of the CAST project.

- Calculate C-14 impact on the near-field releases with new experimental data produced in CAST

- Assess uncertainty reduction in C-14 inventory and impact of C-14 speciation.

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CArbon-14 Source Term

CAST

GRS contribution to D6.1

André Rübel

Date of issue of this report: 28/10/2015

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1 Geological Repository Concept

The strategy of the site selection and licensing procedure for a nuclear waste repository for high-level waste in Germany is under discussion at the moment of writing. While salt has been regarded as main option for the host rock for deep geological disposal, the three host rock types salt, clay and crystalline rock are to be considered equally in the future. Although there has been research on repository concepts in clay and crystalline rock, the repository concept for geological disposal in salt host rock clearly is the most advanced at this time. Therefore, this document describes the repository concept developed for the Gorleben site.

The Gorleben salt dome has been investigated as potential site for a repository for high-level waste since the end of the 1970s. Until now, it has been made neither a decision in favour of Gorleben as repository site nor a final statement on the actual suitability of the site. A site-specific research project, the Preliminary Safety Analysis for Gorleben (VSG), has been conducted from July 2010 to March 2013 to sum up the results of the Gorleben investigation achieved so far, to update the concepts like for emplacement, repository layout, sealing and performance assessment and to compile remaining open questions. The repository layout and the sealing concept for the Gorleben site developed during the VSG project were adapted to the site-specific geological boundary conditions.

The following description of the disposal concept refers to the repository concepts developed within the scope of the VSG [Bollingerfehr et al. 2011] and [Bollingerfehr et al. 2012]. The emplacement fields for spent fuel and HLW are located in the north-eastern part of the repository. Two emplacement concepts are considered for the repository in a salt dome. These concepts are either drift disposal (concept B) or vertical borehole emplacement (concept C) of disposal containers.

Drift disposal concept

The emplacement fields are tailored in such a way that they are completely embedded in the main salt (zsHS) of the salt dome. The repository layout took into account the known and expected geologic situation at the emplacement level (870 m below surface) of the salt dome. Two main transport drifts are the northern and southern boundaries of the twelve emplacement fields (East 1 to East 12). Each emplacement field consists of a crosscut and several parallel emplacement drifts in which the waste containers are emplaced on the floor. After the containers have been emplaced, the void spaces of the emplacement drifts are backfilled with dry crushed rock salt. There is no requirement to seal each single emplacement drift. Optionally, the emplacement field is sealed with a 10-m-long sorel concrete (MgO) plug at both ends of the crosscuts for operational reasons. Sorel concrete consists of magnesium oxide as adhesive cement and crushed salt as aggregate. These plugs have no specified requirement for the post-operational phase. The main transport drifts are backfilled with crushed salt as well, but with a water content of 0.6%wt, to accelerate the compaction process.

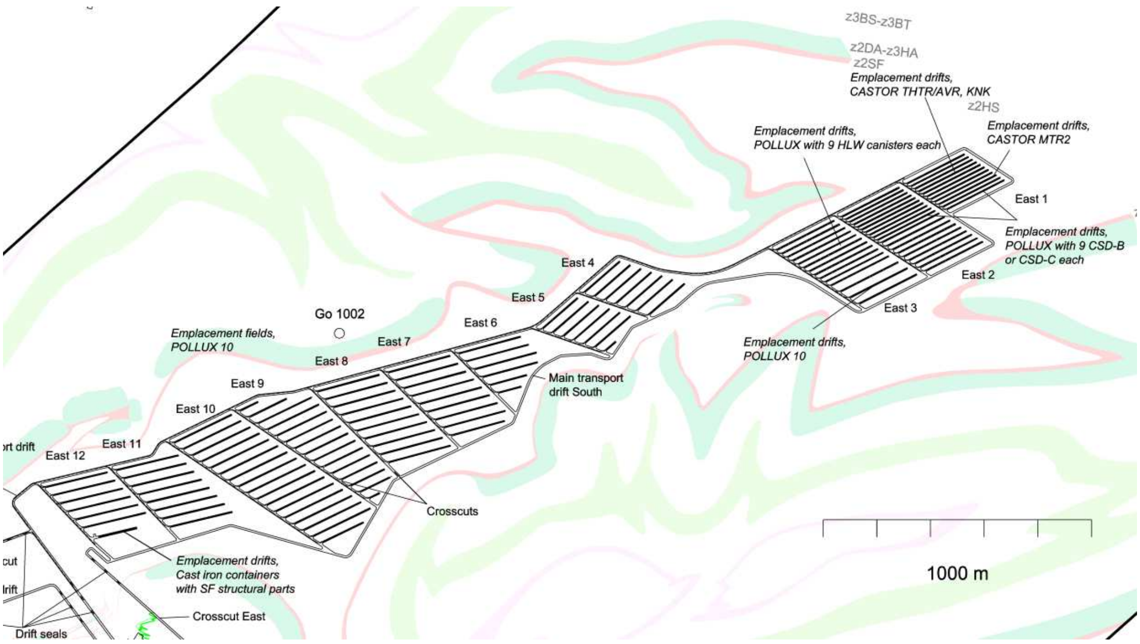

Figure 1: Layout of the north-eastern part of the repository [Bollingerfehr et al. 2012]

Drift seals are located close to the infrastructure area in the vicinity of the two shafts. Each drift seal consists of two 50-m-long sealing elements made of MgO-based concrete and three support elements. The total length of the drift seal is about 150 m. The infrastructure area is backfilled with non-compactible serpentine gravel to allow brines and gases potentially coming from the shaft to accumulate. The shafts are both backfilled and sealed over a length of nearly 600 m with a sequence of three sealing elements and multiple static abutments.

Borehole disposal concept

For the borehole disposal concept, two main transport drifts are the northern and southern boundaries of the emplacement fields. The main transport drifts are interconnected by the access drifts. In each access drift, boreholes with a depth of up to 300 m are drilled into the host rock. The boreholes are supported by steel liners to provide the possibility to retrieve waste canisters during the operational phase. The void spaces between the steel liner and the containers are backfilled with quartz sand. After one borehole is filled, it will either be sealed with a steel lid or with an MgO-plug for operational reasons. These plugs have no safety function during the post-operational phase. The backfilling and sealing of the drifts and shafts and the backfilling of the infrastructure area are comparable with the drift disposal concept. The locations and the layout of the drift and shaft seals are also identical.

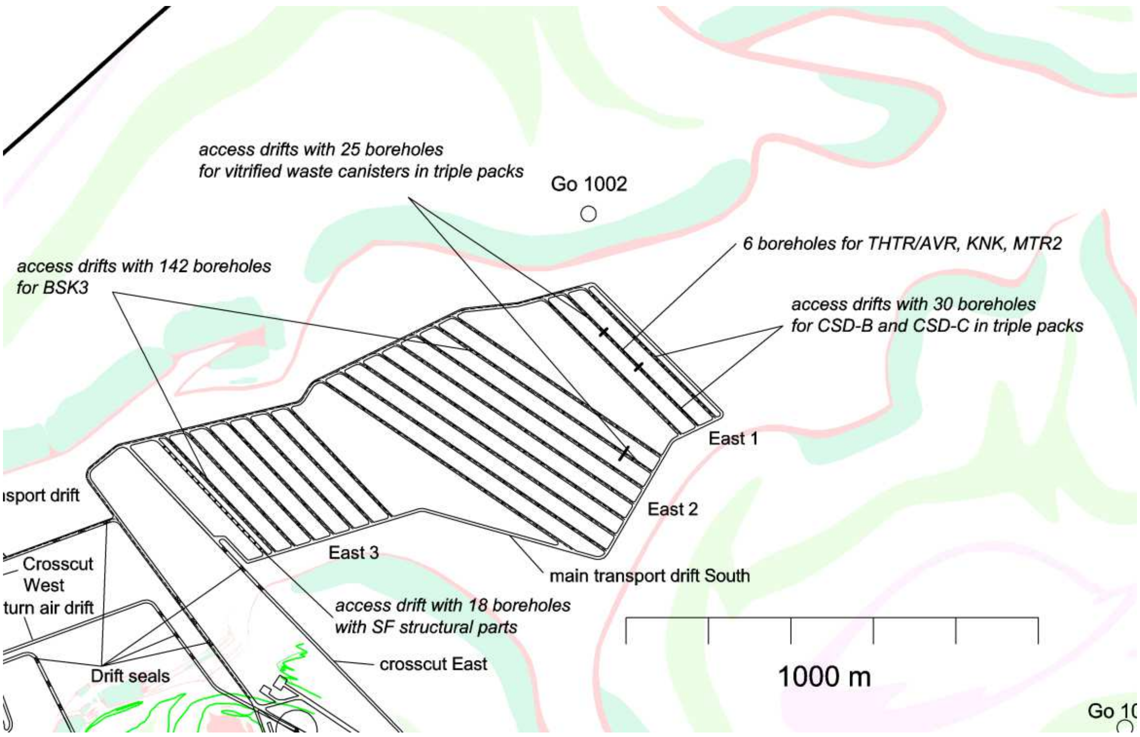

Figure 2: Layout of the north-eastern part of the repository [Bollingerfehr et al. 2012]

2 C-14 Inventory

The inventory described in this chapter refers to a future repository for heat generating waste in Germany. Waste types with negligible heat generation which are foreseen to be disposed of in the already licensed repository Konrad are not considered here. The data given was collected in the German research project Preliminary Safety Analysis for Gorleben (VSG). It considers the disposal of the following heat generating waste types:

1. Spent fuel elements (SF) for direct disposal from pressure water (DWR), boiling water (SWR), water-water (WWER) and research reactors,

2. Vitrified reprocessed spent fuel (CSD-V) mainly from reprocessing in France and Great Britain,

3. Vitrified effluents and sludges from reprocessing (CSD-B) and

4. Compacted Zircaloy hull material and metal parts from disassembled SF-elements (CSD-C).

The absolute amount of the wastes to be disposed of in the future that fall into these four categories are fairly well known due to the phase out from nuclear energy in Germany. The expected inventories are given in [Peiffer et al. 2011]. The radionuclide inventory of the spent fuel given in table 1 was calculated using the OREST-code assuming a burn-up of 50 GWd/t_{hm} for SWR fuel elements, and 55 GWd/t_{hm} for DWR fuel elements with a potential Nitrogen impurity of 30 ppm. Information on other impurities, the uranium enrichment of the fuel, the chemical composition of the hull material and information on the burn-up calculations, like neutron fluxes are also given in [Peiffer et al 2011]. The inventory of the CSD-C waste was obtained from the same OREST simulations with assumed Nitrogen impurities in the Zircaloy hull material of 45 ppm [Hummelsheim & Hesse 2001]. Nitrogen impurities in the metal parts are not given in the documentation of the calculations. The inventory of the CSD-V type of waste was also received from OREST calculations regarding DWR fuel elements with a burn-up of 33 GWd/t_{hm} and the same values for nitrogen impurities as for the spent-fuel given above. This inventory used in VSG for

CSD-V does not correctly take into account the actual distribution of C-14 in the different waste streams during the reprocessing at La Hague. This is why the inventory is expected to be by far too high. A more realistic estimation comes to the conclusion that the 14C inventory per CSD-V canister delivered to Germany from La Hague can be estimated to range between the minimum value of 84.6 MBq and the maximum value of 517 MBq with an expected, average value of 238 MBq [Meleshyn & Noseck 2012].

The type of packaging is not yet decided and depends on the repository concept and host rock type. The drift disposal concept (concept B) of the VSG assumes the use of POLLUX containers which can hold up to 30 SWR fuel elements with 0.177 tons heavy metal (t_{hm}) each or 10 DWR fuel elements with 0.52 t_{hm} each.

As an option, the additional storage of waste types with negligible heat generation in a separate second repository wing with a distance between both wings of about 400 m was examined within the scope of the VSG (concept A). The waste types considered in this option are:

5a. wastes containing graphite which mainly stem from neutron reflector shields from the high temperature rector (AVR),

5b. depleted Uranium tails which stem from the production of fuel elements and

5c. other, non-specified wastes which are not applicable to be disposed of in the future Konrad repository.

The waste categories listed under items 5 are on the one hand still ill-defined in terms of waste amount, packaging and chemical form and on the other hand, it is not yet decided whether those types of waste will be deposited in a future repository primarily constructed for heat generating waste at all. Therefore, these waste types are not considered in the following and the data given in table 1 only refers to the given list items one to four. The full nuclide vectors of the given waste types and additionally for the spent fuel from six research reactors are also listed in [Peiffer et al. 2011].

The uncertainty of the C-14 inventory of spent fuel and hull material directly results from the uncertainty in the assumptions of the burn-up calculations. The assumed Nitrogen impurity of 30 ppm in the spent fuel used for the calculations of the numbers given in table 1 is an upper bound of the expected range. The lower range can be assumed to be 4 ppm resulting in an uncertainty of the resulting C-14 inventory of a factor 3.6 [Hummelsheim & Hesse 2001]. Impurities of Nitrogen in Zircaloy-4 were assumed to be in the range of 45 to 75 ppm. The numbers in table 1 refer to the latter value. No information is given on the bandwidth of the other assumptions used in the burn-up calculations or of the resulting uncertainty of the calculated activities.

No information is available on the chemical form of the C-14.

Table 1: Inventory of C-14 for the main heat generating waste families stemming from the use of power reactors

Waste type	**Package number**	**C-14 activity**	**Uncertainty**
SF (SWR) UO2	14 350 fuel elements	37.7 GBq/t_{hm}	Moderate
SF (SWR) MOX	1 250 fuel elements	21.9 GBq/t_{hm}	Moderate
SF (DWR) UO2	12 450 fuel elements	37.3 GBq/t_{hm}	Moderate
SF (DWR) MOX	1 530 fuel elements	23.1 GBq/t_{hm}	Moderate
SF (WWER) UO2	5 050 fuel elements	10.2 GBq/t_{hm}	Moderate
CSD-V	3 735	17.9 GBq/CSD-V	Moderate
CSD-B	308	not listed	High
CSD-C	4 104	13.8 GBq/CSD-C	Moderate

3 Safety concept

The safety concept for a repository in salt was defined in the VSG and is based on the guidelines in the German regulations as given in the “Safety Requirements Governing the Final Disposal of Heat-Generating Radioactive Waste” [BMU 2010]. The safety concept defines the following three guiding ideas [Mönig et al. 2012]:

- A containment of the radioactive waste in a defined rock zone around the waste should be achieved to the greatest possible extent.

- The containment should be effective immediately after closure of the repository and be ensured durable and free of maintenance by the repository system.

- The immediate and durable containment in a defined rock zone around the waste should be achieved in the first instance by hindering or at least limiting the influx of brines to the waste.

The first two objectives are of general nature. They are applicable to all potential future repositories for heat generating waste in Germany. The third objective is a specific safety objective for a repository in salt. Accordingly, the main safety strategy is to prevent the contact of the waste with external waters. This approach on the one hand limits mobilisation of radionuclides from the waste and on the other hand prevents a connected transport pathway for dissolved radionuclides from the emplacement areas to the biosphere for the reference period.

This objective is achieved by the properties of the salt barrier. First of all by the undisturbed part of the salt host rock formation and second by the crushed salt, which is used to backfill the mine openings in the emplacement areas and access drifts. The crushed salt backfill is expected to be compacted over time by convergence of the host rock to achieve a sufficiently high hydraulic resistance to avoid inflow of brines into the repository. Plugs and seals must provide their sealing function during the early post closure phase, until the compaction of the backfill is adequate and the permeability of the backfill is sufficient low. Research during the VSG project indicates that this is achieved already after a few thousands of years, at the most. According to the regulations, the waste containers for heat generating wastes must be designed to avoid the release of radioactive aerosols for a period of 500 years.

4 Treatment of C-14 in safety assessment

The German safety requirements [BMU 2010] allow the possibility to perform a simplified long-term radiological statement that is achieved without modelling the dispersion of substances in the adjoining rock and the overburden. In this case, the radionuclide fluxes are calculated at the boundary of the so called isolating rock zone[1] (IRZ). The IRZ is part of the repository system and could be regarded as an envelope boundary around the whole repository mine. A maximum allowed release of radioactive substances from the IRZ of 0.1 person-millisieverts per year for a reference group of ten persons is defined for probable developments to comply with the safety requirements. For less probable developments a criteria of 1 person-millisieverts per year is defined. To neglect dispersion, dissolution and retention in the overburden for the simplified long-term radiological statement could be called a conservative approach. Especially since the IRZ can be quite close to the emplacement areas of the waste with additional low permeable rock formations around the IRZ.

The results from the safety assessment performed within the VSG are presented as a safety indicator called RGI (Radiologischer Geringfügigkeitsindex) representing the relative radionuclide outflux compared to the regulatory criterion given above. An RGI of 1 denotes for a radionuclide release equal to the maximum value allowed in the safety requirements. If the RGI is lower than 1, the regulatory criterion is achieved.

Since the salt host rock itself is impermeable for radionuclide flow, the RGI indicator is calculated from radionuclide fluxes in the mine drifts, at the position where the envelope of the defined isolating rock zone intersects the mine drifts. For the case of VSG, this is at the outer face of the drift seals in the access drifts, which is still inside the repository mine and on the repository depth.

[1] In German called einschlusswirksamer Gebirgsbereich (ewG). The VSG project used the term containment providing rock zone (CPRZ) in English.

Radionuclide transport simulations have been performed in the VSG for the reference scenario, for parameter variations representing probable future developments and for deterministic parameter variations to represent alternative less probable scenarios. No probabilistic assessment was performed in the VSG.

4.1 *The reference scenario*

The simulations for the preliminary safety assessment for Gorleben (VSG) were performed separately for the dissolved radionuclides with the single-phase transport code Marnie and for the radionuclides in the gas phase with a modified version of the two-phase flow code TOUGH2 [Larue et al. 2013].

In the reference case, the sealings behave for at least 50 000 years as designed and the porosity of the salt grit backfill in the drifts reduces down to 1% within a few hundreds to a few thousands of years. The according permeability of the salt grit is estimated to be $k = 3\cdot10^{-20}$ m², which is sufficiently low to reduce the inflow along the drifts into the mine to a value that the repository mine is still not fully saturated at the end of the reference period of one million years. No advection, but only diffusive transport is effective for the radionuclides dissolved in the liquid phase for such conditions. No dissolved radionuclides are released from the isolating rock zone during the whole reference period. Consequently, the contribution of dissolved radionuclides to the RGI is zero for the reference scenario.

For the simulations of the gas transport pathway, a lifetime of 500 years is assumed for the waste containers, with the exception of four containers which are assumed to have initial defects already at the time of emplacement. As a conservative assumption, it is assumed that the volatile radionuclides can be instantaneously released from these containers.

For the release of C-14 from the spent-fuel containers, the instant release fraction (IRF) of C-14 is assumed to be 10% for the UO_2 fuel matrix, 10% for the zircaloy and 20% for the metal structural parts. For the CSD-C containers from reprocessing, no IRF of C-14 was assumed due to the acid treatment of the material which removes the oxide layers. No bandwidths have been assumed for those values. No information is available if those are realistic or rather conservative values.

C-14 from the IRF in the matrix and in the metal parts is assumed to be released not before a contact of external water with the waste. However, the 10% of the C-14 in the IRF of the zircaloy disposed with the spent fuel is assumed to be in oxide layers and is assumed to be released instantaneously as CO_2-gas after container failure, even without a contact of the waste matrix with external waters. This assumption is based on [Smith et al. 1993] and subsequent publications and is clearly a conservative assumption.

A flow of non-radioactive gases in the mine is caused from the beginning of the post-closure phase by the displacement of air from the mine. This is due to the convergence of the salt host rock and the decreasing porosity in the salt grit. Additionally, a gas flow also results from hydrogen production caused by iron corrosion by the small amount of water initially emplaced with the containers and the salt grit backfill. External waters that might reach the emplacement fields closest to the shaft can potentially lead to a more significant corrosion and gas production, however to late times which are not relevant for the C-14 release.

The C-14 instantaneously mobilised from the IRF of the zircaloy is released from the four initially defect spent fuel containers directly at the beginning of the post-closure phase and further on is transported along with the non-radioactive gases through the unsaturated drifts to be released through the drift seal from the IRZ. Depending on the chosen parameters, this flux of C-14 yields an RGI up to a value close to 1 already 50 years after emplacement (see figure 3). This early peak is only related to the small number of containers assumed to already have a defect at the time of emplacement. An additional peak of the RGI indicator value occurs after about 10 000 years. This peak results from the large number of containers failing after 500 years. It is a conservative assumption that the other containers fail after 500 years. The reason for the small number of containers with initial defect yielding a relatively high indicator value compared to the large number of containers failing after 500 years is that the most relevant driving mechanism for the gas flow – which is the compaction of the salt grit and the resulting displacement of air – is decreasing rapidly with time.

The parameter variations which are plotted in figure 3 account for the uncertainty of the location of the initial defect containers in the mine, the corrosion rate of metal parts and the humidity in the containers.

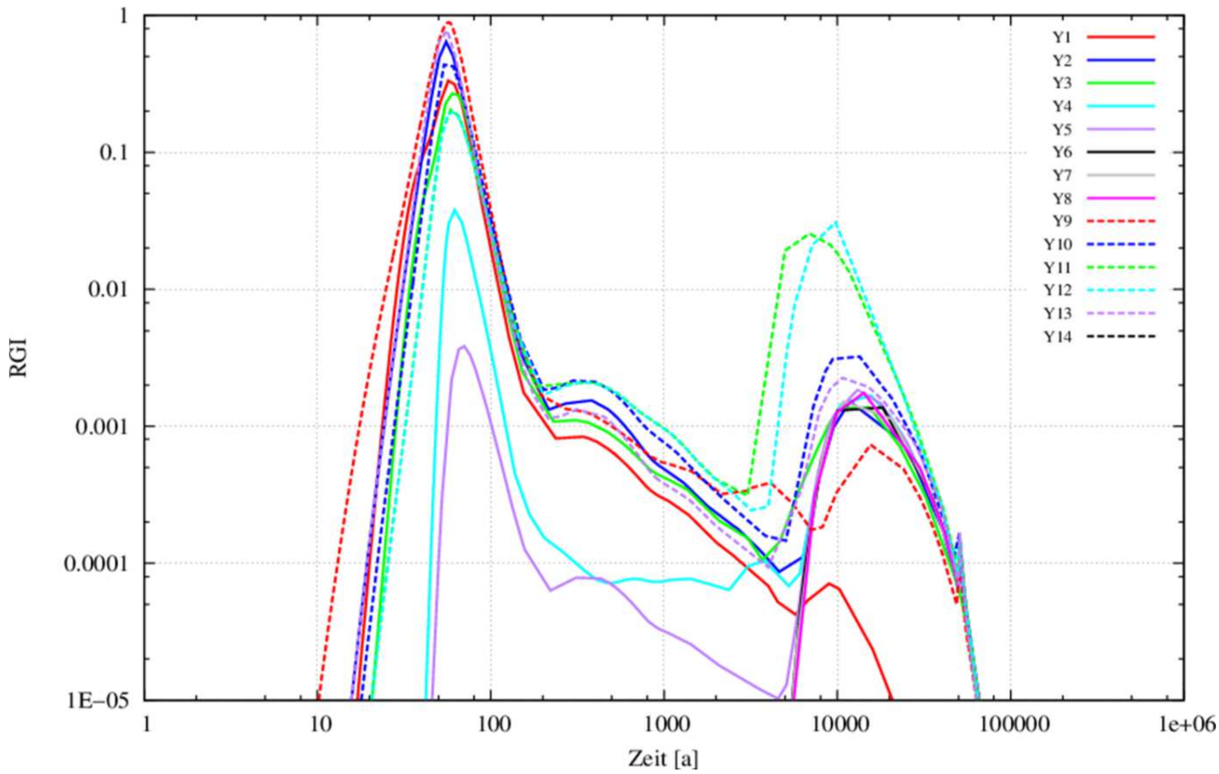

Figure 3: RGI resulting from the C-14 flux out of the spent fuel containers calculated at the isolating rock zone boundary for different variants [Larue et al. 2013]

4.2 Parameter variations for alternative scenarios

Alternative scenarios were examined using parameter variations with less probable values for the varied parameters. The alternative scenarios regarded the following cases: higher number of initially defect containers, higher rates for corrosion and gas production, the early failure of shaft or drift sealings and a higher porosity and permeability of the salt grit in the access drifts.

For the fluid transport pathway, most cases show no outflow of radionuclides out of the IRZ boundary. In the case of higher salt grit porosity, a transport of radionuclides in the fluid phase occurs out of the IRZ boundary, but C-14 shows no relevant contribution to the very low dose.

Alternative scenarios for the gas pathway show a similar behaviour as in the reference case. The curves for the RGI indicator show an early peak that stems from the C-14 release from initial defect containers and a second peak that stems from the large number of containers failing after 500 years.

5 Key issues and priorities

The safety assessment simulations summarized in the previous section which were performed as part of the Preliminary Safety Analysis for Gorleben have shown C-14 in the gas phase to be the most relevant radionuclide with calculated values for the radionuclide flux close to the regulatory limit. This shows the need for additional research and development to reduce potential uncertainties and conservatism. The calculated C-14 fluxes highly depend on the assumptions regarding time and chemical form of the C-14 release. Therefore, the highest priority is to reduce the uncertainty on the release behaviour of C-14. This is mainly related to three questions:

1. Which is percentage of C-14 which can be released into volatile form?
2. Which is the temporal behaviour of the release?
3. Is water necessary to transfer C-14 into volatile form or does this occur also without the presence of humidity?

An additional area of investigation is the optimisation of the repository design to delay a potential release of volatile C-14. A possible option would be construction of gas storage areas inside the IRZ.

Sensitivity analysis should be performed to investigate the most sensitive parameters on the C-14 dose contribution to help the design optimisation.

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CArbon-14 Source Term

CAST

ENEA contribution to D6.1

Barbara Ferrucci – Riccardo Levizzari – Alfredo Luce

Date of issue of this report: 08/06/2015

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1 Geological Repository Concept

During the past period of more than 15 years (since the 1990s), ENEA and some Universities that now belong to the Italian CIRTEN consortium (Interuniversity Consortium for Technological Nuclear Research, established in 1994) have managed a broad R&D program on radioactive waste management and disposal; some research activities have also been carried out in the frame of different EC framework programs before Horizon 2020.

In the same time SOGIN S.p.A. (the main Italian WMO) has been entrusted for the decommissioning of all the Italian NPP and fuel cycle facilities as well as for the radioactive waste management and disposal.

Based on past studies, SOGIN carried out a selection of suitable sites for the realization of a waste repository; this selection led to the choice of a salt formation in the south of Italy as a candidate site to host the national repository for LLW, ILW and HLW (2^{nd} and 3^{rd} category of Italian radioactive waste classification) [ENEA-DISP, 1987][1]. This choice was supported by a government decree on November 14th, 2003, but the Italian government was forced to immediately withdraw the decree after strong protests of local populations.

Presently the geological disposal is no longer on the agenda and the Italian strategy is to realize a surface repository for LLW and, in the same site, make a temporary storage for high level and long lived waste, waiting for future decisions.

Within the CAST project, the development of a conceptual model of a hypothetical Italian geological repository and its following implementation in simulation code is useful to further improve the knowledge about requirements in safety assessment and to support future R&D program on radioactive waste disposal.

[1] Italian radioactive waste classification: first category, for waste which decays, in a period of a few months to a maximum of a few years, to negligible values of radioactivity; second category, for waste which decays, in a period of a few decades to a few hundreds of years, to some hundred Bq/gr; third category, for waste which decays, in a period of more than thousands years and beyond, to some hundred Bq/gr.

Due to the lack of choice about the geological disposal, site specific data for safety assessment studies referred to the real Italian geological disposal are not available. Part of Italian past work have been focused on the clay formations of Pliocene age, occurring over large zones of basin structures, especially in Italian eastern area. An analysis conducted during the last months has evidenced that data and information about these geological contexts, although representative of many local environments of Italian eastern basins, are too generic. On the contrary, geological data from the site of the above mentioned salt host rock are site-specific and moreover they preliminary proved this could be a favourable solution to host a geological repository; then they have been chosen to proceed with the studies within the CAST project, creating a preliminary, but only theoretical, conceptual model of a deep repository for HLW.

It is important to underline that this is only a hypothetical study, because the mentioned site is no longer considered suitable for RW disposal for reason of social acceptance.

Regarding the stratigraphy, from soil surface, the site is featured by quaternary alluvial sediments for about 60 meters in thickness, beneath which a powerful series of Plio-Quaternary sedimentary rocks develops, until 1180 m depth (Figure 1).

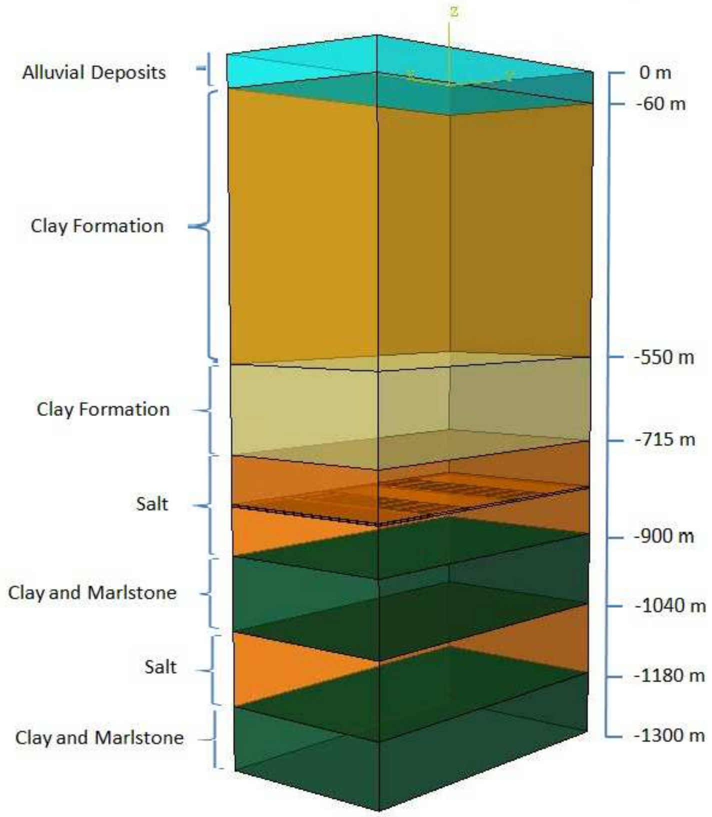

Figure 1: Stratigraphy of the potential site.

Within this succession, the favourable rock considered in the hypothetical study is constituted by a salt body, with thickness of about 200 m; this is the most superficial one, at about 720 m depth. The hosting rock can be considered quite homogeneous, although information about possible inclusions of other sedimentary material (clay, silt, etc.) could be obtained with direct investigations, nowadays not available. A second salt body is 130 meters thick, at about 1050 m depth from soil surface. The two salt bodies are separated by a layer of marl and clay, about 140 m thick. The formation containing the two salt bodies is superimposed on a formation of clay and marl of Miocene age, which extend from a depth of 1180 meters up to about 2000 m. Deep salt deposits are a promising solution for the disposal of HLW, particularly for self-sealing property due to their plastic feature, high thermal conductivity, very low permeability and relatively easy minability.

From the hydrogeological point of view, the presence of a powerful thickness of argillaceous materials to the roof and at the base of rock salt deposits, seems to exclude the presence of groundwater circulation, so salt deposit was assumed to provide little or no mechanism and pathway for water flux. The same existence of salt rock is an evidence of the absence of local circulating groundwater, otherwise salt minerals would have been dissolved. Only site-specific and detailed hydrogeological studies could confirm this hypothesis, with a direct evidence.

Since at this time no detailed project about geological repository is available in Italy, the hypothetical conceptual model has been developed as those of other countries, utilizing available literature data, i.e. from the US Waste Isolation Pilot Plant (WIPP); it consists of both surface and underground facilities, connected by vertical shafts. The underground layout is shown in Figure 2, referred to the closed repository (all waste is embedded into modules and packages, positioned within storage rooms).

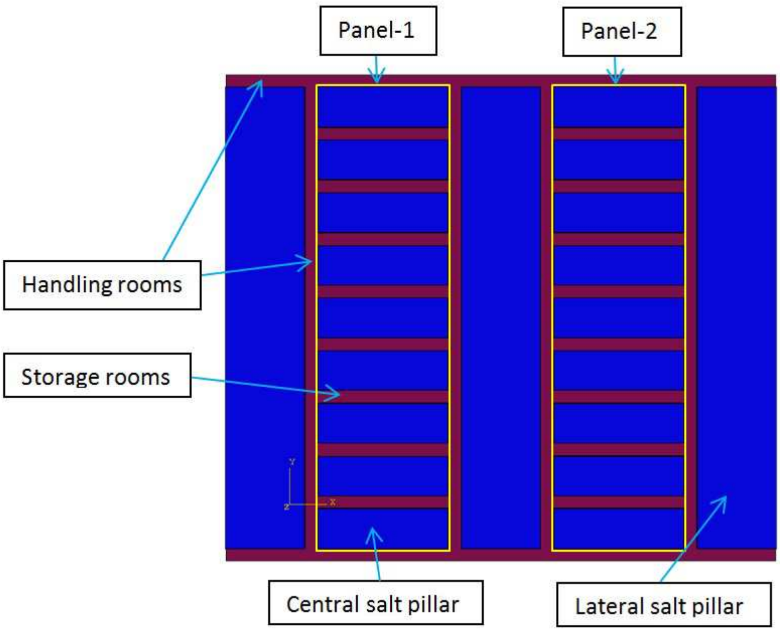

Figure 2: Preliminary layout of geological repository (underground facility).

At the moment, it's provided that the storage rooms are grouped in two panels, each of which consist of eight rooms (about 10 m wide, 100 m long and 4 m high), separated by a 30.5 m wide pillars (central pillar, in the Figure 2). The two panels are separated by 61 m wide pillar (lateral pillars, in the Figure 2).

2 C-14 Inventory

The following Table 1 gives a preliminary estimation of the inventory of waste containing C-14, to be placed in a geological disposal. The inventory does not contain the residues coming from the reprocessing of irradiated fuel sent abroad; this waste results from the irradiated fuel sent abroad for reprocessing and to be returned to Italy from UK and France (3rd category vitrified waste).

Waste listed in table are classified as long lived not only for the content/activity of C-14 but because of the high content/activity of other radionuclides; because of this, they are classified as 3rd category in Italian radioactive waste classification and for this reason they are destined to geological disposal.

Origin	Materials	Volume (m^3)	Inventory of C-14 (GBq)	Quality of information Uncertainty
GCR-Magnox Reactor (*Latina* NPP)	Graphite	3.30+E3	2.83E+04	Estimated Moderate
Nuclear Power Plants	Resins, metals	n.a.	3.30E+03	Estimated High
Medical, industrial, research	Conditioned sources	172.60 (863 drums 200 dm^3)	0.42	Estimated Moderate
	Not treated sources	6.78 (113 metallic drums 60 dm^3)	92.80	Estimated Moderate
	Cemented sources	113.60 (284 metallic drums 400 dm^3)	106.28	Estimated Moderate
	Solid treated sources	42.00 (105 drums)	11.00	Estimated Moderate
	Not treated liquid waste	4.19 (192 plastic 20 dm^3; 2 metallic drums 120 dm^3; 1 metallic 110 dm^3)	4.05	Estimated Moderate
	Not treated solid waste	15.06 (251 metallic drums 60 dm^3)	2.36	Estimated Moderate

Table 1: Inventory of Italian ILW and HLW-LL containing C-14.

At this stage of the project, no information about specific activity is available, as well as detailed information for a better distinction among the waste families. Volume data of graphite are referred to not-conditioned waste. The high uncertainty for data about resins and metals (“quality of information” in the above table) is due to the worst characterization of waste.

3 Safety concept

Although in Italy specific and detailed safety assessment for geological disposal has not been developed, the well-known safety criteria and functions can be applicable to the context of a hypothetical Italian geological repository. These can be implemented in the present conceptual model for the following simulation about safety, to preliminary evaluate the radiological impact of C-14 in the post-closure phase. In simulations, two scenarios will be examined: a reference base scenario, with normal evolution of the repository and external environment, and a disturbed scenario, referred to a huge worsening of the safety performance of engineered barriers and an unexpected presence of discontinuities in hosting rock, creating preferential pathways for radionuclides migration. The description of scenarios that are to be simulated in preliminary safety analysis are described in following paragraph § 4.1, § 4.2.

Safety in the long term must be guaranteed by a comprehensive approach ensuring the isolation of radioactive waste from external phenomena, the containment of radionuclides and the retardation of their migration toward far-field and biosphere, using the concepts of multi-barrier: engineered and geological barrier. This combination ensures a specific containment and the defence in depth, with a level of isolation effective for different periods of time (§ 3.1 for EBS), limiting the possible release and migration of radionuclides and isolating waste from surface phenomena [Frullini et al., 2011].

3.1 *Engineered Barrier System (EBS)*

The components of the EBS are: waste form, waste package, possible repository modules, crushed-salt backfill, panel closures and shaft seals.

For what concern the waste form, and with particular reference to simulations supporting future safety analysis, three types of solid waste forms are assumed to be in the repository (not all containing C-14): cemented waste, borosilicate glass waste, metallic waste. As part of the considerations for the modelling of the radionuclides release, waste form containing C-14 are characterized by three main properties: the degradation/dissolution rate, the retardation factor and the dilution factor of the embedded radionuclides. Since we will assume the release rate of radionuclides coincides with the degradation/dissolution rate of the waste form, it plays a crucial role in the evolution of selected scenarios for simulation of radionuclides release and migration. For what concern the graphite, at the moment no decision has been assumed about its conditioning and its final form.

At the moment, waste are considered to be packaged in metal drums and corrosion resistant containers, chosen on the base of the type and activity of waste, acting as barrier during the elevated-temperature phase that lasts a few hundred years. For this purpose the containment must have high integrity in salt-rock and salt-brine environment; resistance to corrosion must be guaranteed during the emplacement operations and in the post-closure phase, at least for 1,000 years. For example, in studies carried out about corrosion, stainless steel and carbon steel are promising material to guarantee safety, especially during the thermal phase [King, 2007]. In this phase, brine migration processes can be relevant and it's fundamental to guarantee the capability of isolation until the complete reconsolidation (at least 200 years) of the crushed salt backfill around the waste package, as better explained hereinafter [Clayton et al., 2012], [Sevougian, 2012]. As conservative assumption, in simulations supporting future safety assessment studies, waste package will not be considered: they are assumed to fail immediately (at time zero) after the closure of the repository; the release-rate of the C-14 and other contaminants will coincide with waste corrosion rate, that could be enhanced when in contact with concentrated brines at elevated temperatures, with gas generation in chemically reducing conditions.

The typology and necessity of a final module to contain waste package has not yet been decided. At the moment, one possible solution is a “multi-use container”, designed to isolate the 2nd category of waste (LLW and ILW), containing a number of waste packages (from 2 to 8) in the repository. It's realized with reinforced concrete to provide mechanical resistance, durability and radionuclide containment for at least 300 years; the volume of each module is 10.8 m^3. In simulations, modules, as the entire waste package, are not considered for conservative reasons.

Waste packages/modules are arranged next to each other and piled one on another in the rooms; void spaces will be backfilled with crushed salt excavated in the repository, to protect waste against corrosive agents in the mid and long term and to create a continuum barrier with the surrounding rock, against others physical and chemical degradation phenomena. Over a period of time following the emplacement, the confined space around the waste disposal area will be slowly closed by creep deformation of salt rock and the crushed salt backfill will undergo consolidation. This would results in close contact of the waste container with the consolidated salt rock (consolidation of salt backfill will take place in the first 200 years) [Clayton et al., 2012], [Sevougian, 2012]. During this period a linear decrease of both the permeability and the porosity of the backfill is considered. After this period the same characteristics (physicals and chemicals) for both the surrounding salt rock and the reconsolidated backfill are considered in simulations.

After the repository closure, drift and shafts could provide the primary pathways for radionuclide release and transport from the repository. The EBS system must also be effective for this problem, with appropriate sealing solutions, to reduce the inflow of groundwater toward the repository and to reduce degradation of waste and leaching of radionuclides. The sealing system provides containment for different periods of time; in particular, the concrete or asphalt components can provide the short term isolation (1,000–2,000 years), while crushed salt provides the long term barrier capability, consolidating to the surrounding rock, reducing its porosity and permeability, guarantying the isolation of the system for the entire lifetime of the repository.

3.2 *Geological barrier*

Salt deposits provides the capacity of radionuclides containment in the long term, to prevent them from coming in contact with the biosphere. The depth of the repository (at about 720 m depth) ensures a good safety margin with respect to the surface events and provides a barrier to the degradation agents that could reduce the effectiveness of engineered barriers. For what concern human intrusion (e.g. salt mining) it’s not considered in present safety analysis.

The salt rock formation is consistent with the necessity to isolate and contain waste from external phenomena as to retard the migration of radionuclides toward biosphere. This is due to low porosity and permeability of rock matrix and to the absence of groundwater circulation. Moreover the presence of argillaceous rocks at the roof and at the base of salt deposit prevents the migration of radionuclide towards biosphere, because of sorption phenomena, practically permitting migration only by diffusion. At this depth the circulation of water is supposed to be strongly limited or totally absent, reducing the possibility of radionuclides migration by advection. Based on the data available today, the lateral extension of the pile clay-salt-clay is supposed to be at least 1000 m.

The extension of Excavation Damaged Zone is considered limited within some meters around the repository structure and will partially reduce in time due to the self-sealing capacity of salt.

Other considerations on the safety performance of hosting rock will be developed taking into account Italian past studies about natural analogues and international studies about repository development in salt rock [Benvegnù et al., 1988], [Lombardi, Polizzano, 1988], [Sevougian et al, 2012].

4 Treatment of C-14 in safety assessment

Given that in Italy detailed studies about safety assessment of geological repository have never been performed, nowadays no information is available about treatment of C-14. In the simplified simulations that is going to be developed for the CAST project, the first evident assumption useful to understand dynamic of C-14 regards the fact that the bulk of this radionuclide is related to activated material; as a consequence of this, its dissolution induce release and migration of the radionuclide, especially in gaseous form.

The most important common aspects of considered scenarios in the future simulations:

- C-14 is homogeneously distributed only in the waste type containing the radionuclide;
- release rate of radionuclides coincides with the degradation/dissolution rate of the waste form;

- only a fraction of the C-14 will transfer to the brine, while the remaining will transfer to the gas phase (methane and carbon dioxide) or remain in solid phase;
- the transfer rate of C-14 is correlated with the degradation/dissolution rate of the waste form and with the Instant Release Fraction;
- crushed salt backfill around the waste packages is completely reconsolidated after 200 years and at this time it results in a dry intact salt material (during the first 200 years, the permeability and porosity of the backfill decrease up to the values of the surrounding salt rock);
- waste packages are conservatively assumed to fail immediately at the time of repository closure (i.e., at time zero) and the beginning of degradation in the waste form is conservatively assumed occurring from the beginning of the analysis.

4.1 *Reference case*

Radionuclides are released from the repository by a sequence of processes that could occur in a salt repository without any incidental events. The case assumes that backfill and shafts provides the primary pathways for radionuclide release and transport from the repository. Furthermore, due to the very low permeability of the salt rock, the transport of contaminants away from the repository through the hosting rock is supposed diffusion dominated.

4.2 *Disturbed case*

A second scenario is considered, assuming a huge worsening of original performance of engineered barriers, creating fast pathways for radionuclides to the far-field, especially through the sealed shaft to the surface. In this case, because of the presence of a disturbed zone overlying the repository, a series of fracture will also be expected, and an increase of permeability and porosity of surrounding rock will be modelled. This scenario will also be accompanied by an increase in the degradation/dissolution rate.

Other alternative *sub-scenarios* will be analysed during simulations, taking into account the current needs and what emerges from future considerations. In particular, a preliminary analysis about the most sensitive parameters, whose variation induces a large variation in the radiological impact of C-14, will be performed.

5 Key issues and priorities

Following priorities have been identified for the subsequent activities:

- quantitatively define the most important technical aspects for repository safety analysis;
- improve the conceptual model of repository for the subsequent implementation in the safety analysis;
- reduce the aspects of uncertainty about waste inventory data, in reference to their content of C-14.

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CArbon-14 Source Term

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RWMC contribution to D6.1

Tomofumi Sakuragi and Hiromi Tanabe

Date of issue of this report: 25/05/2015

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1 Geological Repository Concept

"All of the spent fuels are reprocessed" is a basic policy of nuclear fuel cycle in Japan. The TRU waste is arising from fuel reprocessing, MOX fuel fabrication and dismantling of these nuclear facilities. The electric power utilities and JNC (now JAEA) are TRU waste producers and have responsibility to manage it. In 2000, they published a summary document entitled "Progress Report on the Disposal Concept for TRU Waste in Japan (TRU-1)" [JAEA and FEPC, 2000], and then in 2005 followed by "Second Progress Report on Research and Development for TRU Waste Disposal in Japan (TRU-2)" [JAEA and FEPC, 2005]. The reports addressed the disposal of TRU waste and the possibilities for disposing of this waste under typical geological conditions in Japan (below 300 m). It also highlighted technological issues to be addressed in future research and development programs. The grouping of the TRU waste for deep geological disposal is shown in Figure 1. C-14 is contained in various radioactive wastes such as Hull & end waste (Hull waste) and Bitumen solidified waste of TRU wastes, irradiated core components of LWR, and so on in Japan. However this report deals only C-14 contained in the Hull waste.

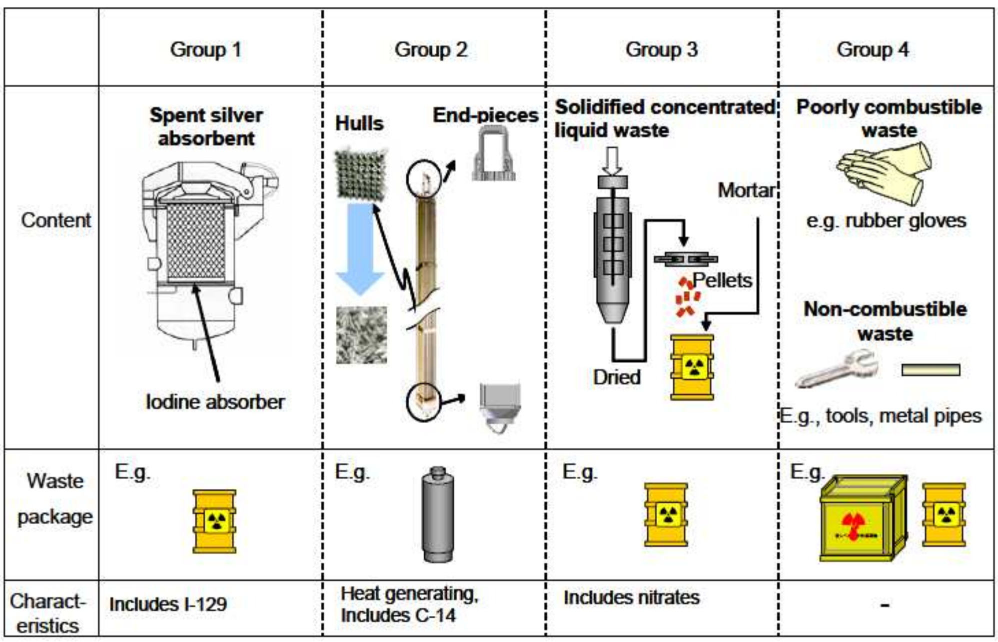

Figure 1: Grouping of TRU waste in Japan.

The basic concept for the TRU repository in Japan is shown in Figure 2. Since the heat generation is small, TRU wastes for deep geological disposal can be emplaced together in tunnels with large cross-sections (diameter of 12 m). However, there is a wide variety of waste materials such as metal, cement, nitrates and organics. For this reason in the basic TRU-waste repository layout, each of the waste groups will be emplaced in separate disposal tunnels. In other words one disposal tunnel or drift will contain waste from one group only. Moreover the design specification of each disposal tunnel will depend on the characteristics of the waste group and the surrounding host rock. Since no candidate site exists in Japan yet the generic datasets for hard rocks (HR) (e.g. crystalline rocks) were used in numerical analyses to model the performance of the EBS designs. Owing to the larger concentration of highly soluble and low sorbing radionuclides in TRU-waste in Group 1 and Group 2, it was shown that disposal tunnels for these wastes required a buffer consisting of compacted bentonite and sand mixture to maintain a low ground water flow in the repository. On the other hand, disposal tunnels for Group 3 and Group 4 did not require a bentonite buffer.

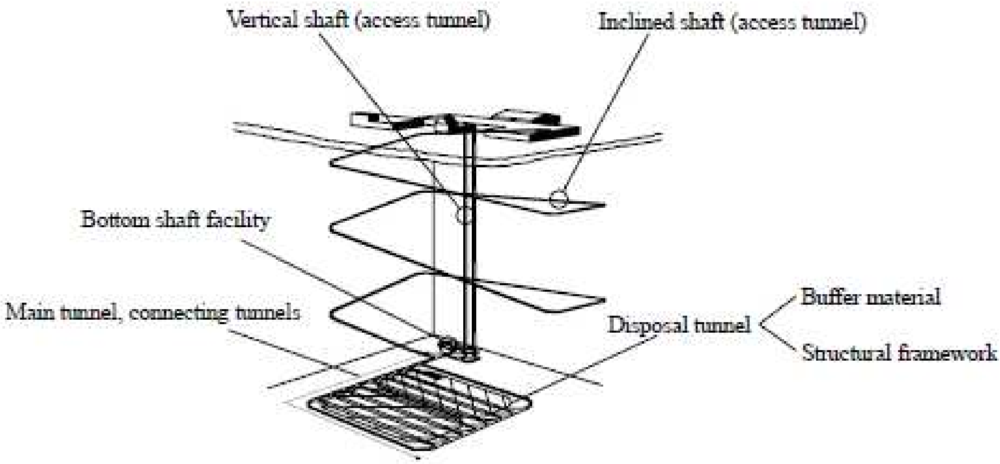

Figure 2: Basic concept for the TRU repository in Japan.

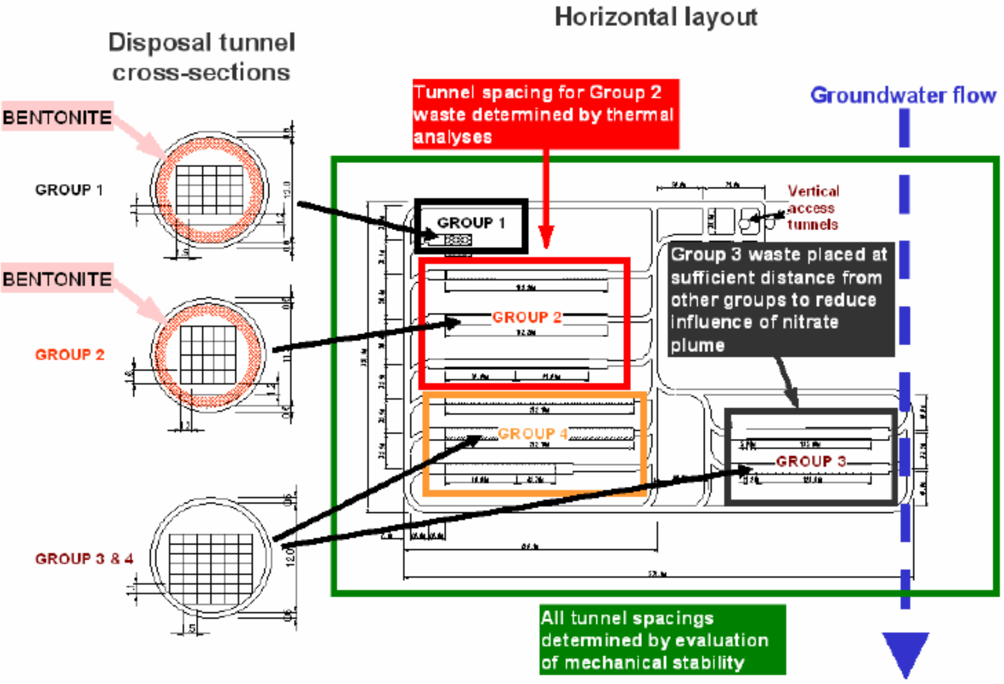

Figure 3: A layout of TRU waste disposal facility.

2 C-14 Inventory

The C-14 inventory for respective groups of the TRU waste is shown in Table 1. The Group 2 of hulls and endpieces waste contains large amount of C-14. The detailed descriptions for the hull waste are given below.

The TRU-2 report estimated the total C-14 inventory in hull and end-piece wastes corresponding to reprocessing 32,000 tons of uranium from the commercial reprocessing plant (JNFL, Rokkasho, Japan) to be 4.4×10^{14} Bq by using an activation calculation of ORIGEN-2. (The rest of the little was from JAEA) A typical PWR fuel assembly with a 45GWd/tU burnup (Enrichment 4.5%, Specific power 38MW/MTU), the maximum permissible burnup for the Rokkasho reprocessing plant, was assumed as a conservative assessment.

The assumed materials in the waste are Zircaloy-4, stainless steels (302, 304L), nickel base alloys (Inconel -600, -718 and –X750), hull oxide (surface zirconium oxide onto fuel rods) and fuel residue. The respective inventories are shown in Figure 4. The C-14 in hull oxide was assumed the abundance of 13%, based on a previous finding of a thick oxide layer of 80 µm on a single PWR cladding specimen and of a C-14 activity measurement [Yamaguchi et al., 1999].

The nitrogen impurities in the materials used above calculation are given below.

- Zricaloy 4: 40 ppm
- Stainless steels: 500 ppm
- Inconels: 60 ppm
- UO2: 0 ppm

These values for metals are judged from several specification data.

Table 1: C-14 inventory of TRU wastes in the TRU-2 report. Gr.2 inventory includes wastes from commercial (JNFL) and JAEA reprocessing. The percentage represents the C-14 ratio in each group.

TRU wastes	Gr.1	Gr.2	Gr.3	Gr.4
Inventory (Bq)	0	5.0×10^{14}	5.7×10^{12}	8.5×10^{11}
Major wastes	-	JNFL & JAEA; Hull & ends waste (100%)	JAEA; Bitumen solidified waste (MA*) (75%), $NaNO_3$ solidified waste (MA) (19%), Bitumen solidified waste (LA*) (4%)	JNFL; Ash melt + Water in hull can (68%), Hull can (21%), Water in hull can (7%)

MA: Low-level radioactive liquid waste with relatively high radioactivity
LA: Low-level radioactive liquid waste with low radioactivity

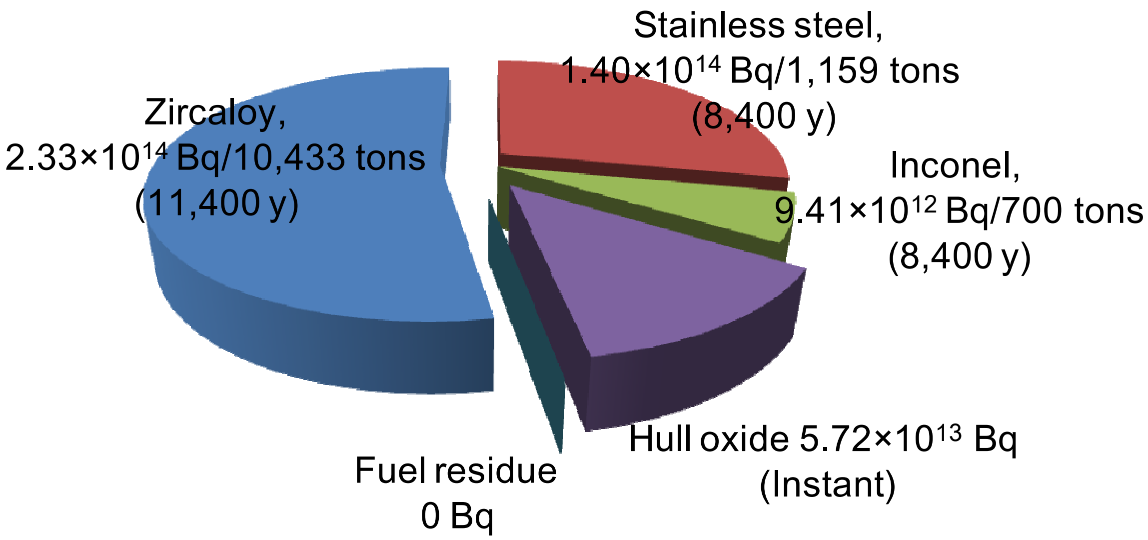

Figure 4: C-14 inventory in group 2 (hulls and endpieces) corresponding to commercial 32,000 MTU reprocessing. The values in parentheses represent the assumed leaching period in safety assessment.

Some conservative assumptions and preliminary estimates were used in the above calculation. For example, the total amount of radionuclides generated from a single type of fuel assembly (45 GWd/tU for a PWR unit), and the thickness of the Zircaloy oxide film on the hulls (80 μm) were both overestimated. The second assumption in particular has an effect on exposure dose evaluation (see Figure 9). The recent study has been re-evaluated with the C-14 inventory in the Group 2 based on the more realistic information for fuel types, burnup conditions and oxide thickness [SAKURAGI et al, 2013]. Although the total inventory was the almost same, the distribution of the C-14 inventory to hull oxide, which was estimated under the assumption of instantaneous radionuclide release in the safety assessment, decreased from 5.72×10^{13} Bq (13% of the total) to 1.30×10^{13} Bq (2.9% of the total).

Chemical speciation of C-14 after released from the wastes is assumed all aqueous organics (specific compounds are not known).

3 Safety concept

The overview of radionuclides (C-14) transport from disposal facility to biosphere in the aqueous media is shown in Figure 5. The safety function can be classified into three parts. The first is the waste, the second is the engineered barrier and the third is natural barrier systems.

C-14 in Hull waste exists in metals and oxide film of Zircaloy. C-14 in metals is considered to be released to the environment congruently with the corrosion of metals. C-14 in the oxide film is considered to be released to the environment instantaneously (see Figure 4).

In the engineered barriers, transportation of released C-14 in liquid phase is retarded by absorption reaction with cement material of filler and migrates into the buffer material. At present, C-14 transport in the structure is ignored. Diffusion is the dominant transport mechanism for C-14 in the buffer material and absorption of C-14 by buffer material is not considered for conservatively. Figure 6 shows the basic concept of the engineered barrier system.

C-14 which penetrate into the buffer material are rapidly mixed with groundwater flowing through the excavation disturbed zone and flows into cracks in the host rock. In the excavation disturbed zone, retardation effects such as sorption are not considered. In the reference host rock, C-14 is considered to transport by advection/dispersion in cracks, diffusion (matrix diffusion) into the rock matrix from cracks and sorption onto the surface of mineral grains in the rock matrix. Figure 7 shows the conceptual illustration of the radionuclide transport in the natural barrier (host rock).

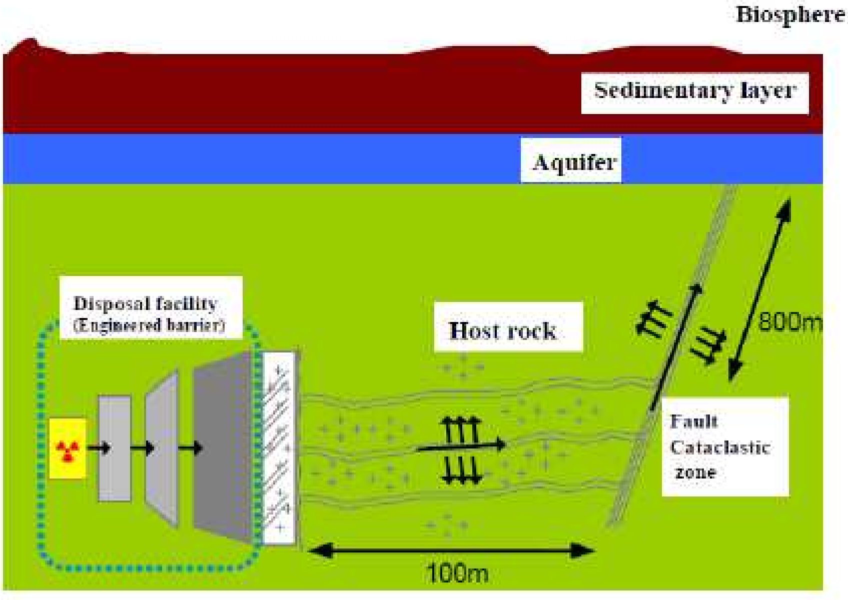

Figure 5: Overview of radionuclide transport pathway.

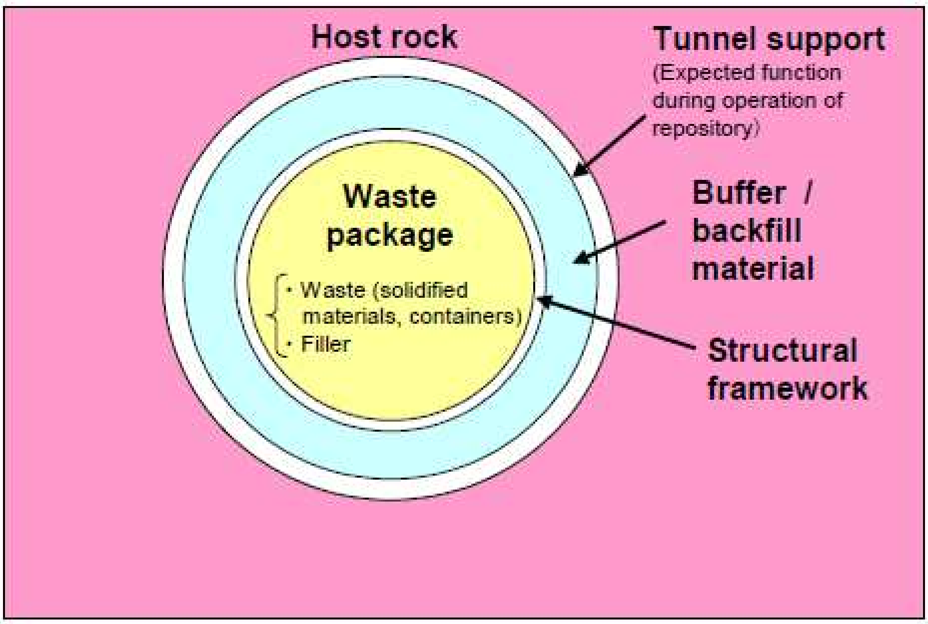

Figure 6: Basic concept for the engineered barrier system.

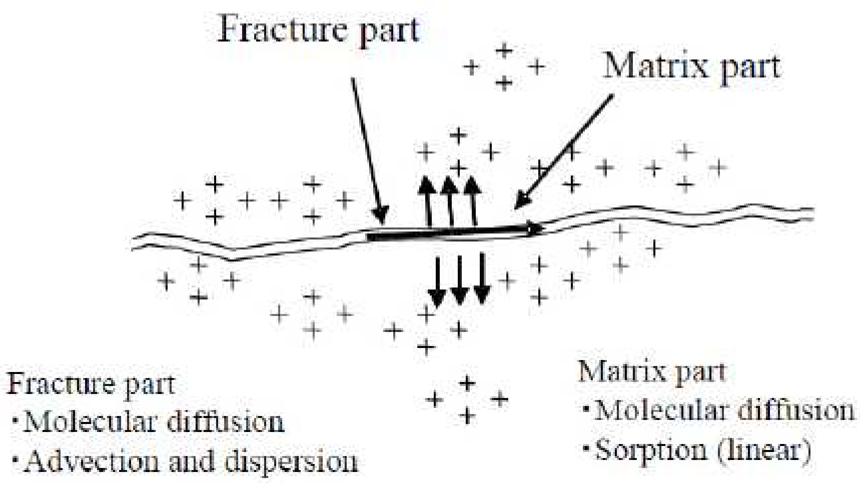

Figure 7: Conceptual illustration of the radionuclide transport in the natural barrier (host rock).

4 Treatment of C-14 in safety assessment

Since no candidate site exists in Japan yet the generic datasets for a geological environment (Table 2) and migration parameters for radionuclides (Table 3) were used in performance assessment [JAEA and FEPC, 2005]. .

Table 2: Boundary conditions and parameters in the transport model for the natural barrier

Table 4.5.2-12 Summary boundary conditions in the Reference Case model

Parameter	Host rock (one-dimensional multipathway model)	Fault located downstream of facility (one-dimensional parallel-plate model)
Rock type	Granite (acidic crystalline rock)	
Groundwater	Freshwater	
Hydraulic gradient	0.01	
Radionuclide release rate input	Migration from engineered barriers	Migration from host rock
Migration distance (m)	100	800
Transmissivity (m^2/s)	Log-normal distribution (log-mean -9.99, log-standard deviation 1.07, maximum and minimum value for transmissivity: 10^{-7} and 10^{-13})	10^{-7}
Darcy flow velocity of groundwater in fracture ($m\ s^{-1}$)	$0.5 \times \sqrt{(\text{transmissivity})} \times (\text{hydraulic gradient})$	
Dispersion length (m)	10	80
Proportion of fracture surface from which radionuclides can diffuse into the matrix (–)	0.5	
Diffusion depth (m)	0.1	
Porosity (–)	0.02	
Dry density (Mg/m^3)	2.64	
Effective diffusion coefficient(m^2/s)	3×10^{-12}	
Sorption distribution coefficient	Table 4.5.2-11	

Table 3: Parameters used for the C-14 transport model

Elements	Solubility (mol/L)	Distribution coefficients (m^3/kg)			Effective diffusion coefficients (m^2/s)	
		cement	bentonite	host rock	cement	bentonite
Carbon (Organic)	Soluble	0.00025	0	0.0001	8×10^{-10}	4×10^{-11}

The biosphere model considers a plain topography and a fresh water system which is evaluated by applying the same compartment model. The dose conversion factors for farming, freshwater fishing and marine water fishing exposure groups are estimated. Dose conversion factors for C-14 are summarized in Table 4 [JAEA and FEPC, 2005]. From this, the farming group is identified as the dominant group. These dose values are used in the radionuclide transport analysis. In the calculation of dose conversion factors, the effects of naturally occurring stable isotopes are not considered.

Table 4: Dose conversion factors

Table 4.5.2-13 Dose conversion factors ((Sv/y)/(Bq/y))

Radionuclide	Farming group	Freshwater fishing group	Marine fishing group
C-14	6.4×10^{-17}	3.5×10^{-17}	2.4×10^{-17}

The dose is calculated by multiplying the release rate to the biosphere (via the fault) by the effective dose conversion factor in the biosphere. The result for each group and radionuclide are shown in Figures 8. The safety standards in foreign countries and natural background radiation levels in Japan are also shown. The waste group which has the largest effect on dose is Group 1, with a maximum value of about 2 µSv/y per 10,000 years. The second largest dose is given by Group 3 because the influence of nitrates in Group 3 was considered. According to Figure 8, the dominant radionuclide after disposal and up to 10^7 years is I-129. This is followed by C-14, Se-79 and Tc-99. In particular, Se-79 and Tc-99, which belong to Group 3, control dose due to the influence of nitrates.

For Group2, the dose from respective components is also shown in Figure 9. Zircaloy gives the largest dose due to large amount of its inventory. Although the inventory of hull oxide is small the dose from hull oxide is not negligible because it is assumed to be released instantaneously.

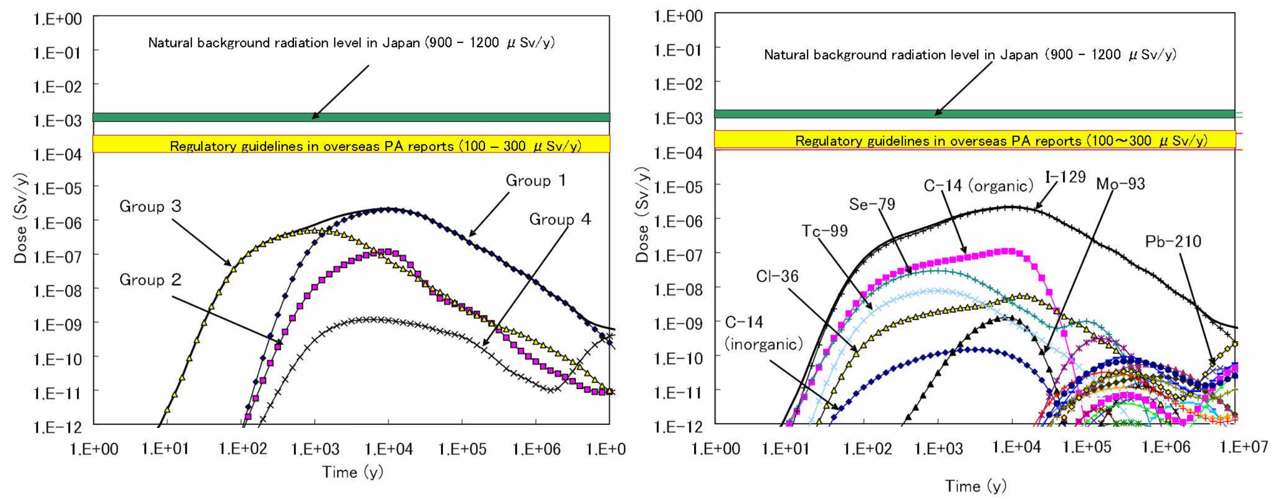

Figure 8: Results of dose assessment for each waste group (left) and for each radionuclide (right).

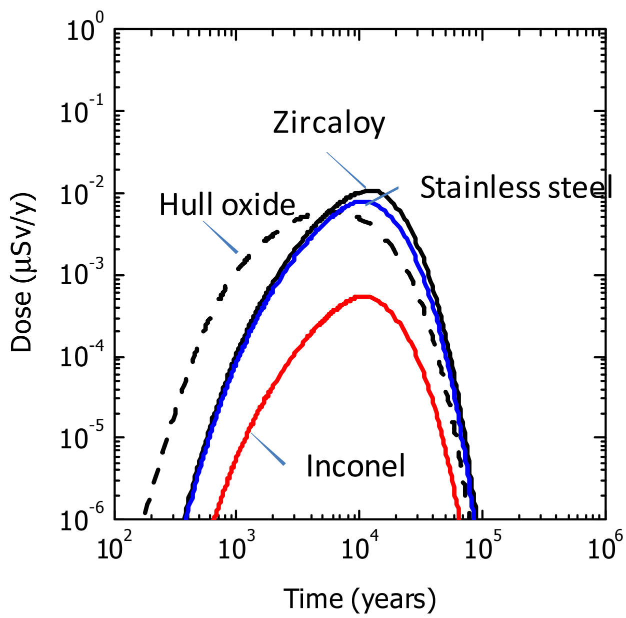

Figure 9: Results of dose assessment from respective components of hull waste. The detailed data are obtained from Sakuragi et al. [SAKURAGI et al., 2013].

5 Key issues and priorities

Key parameters to evaluate transportation of C-14 of Hull waste are as follows;

(1) Inventory in Zircaloy metal and Zircaloy oxide, stainless steels end-peace, Inconel spacer, etc.,

(2) Long term release property from metal and oxide,

(3) Chemical form in underground water, and gas phase,

(4) Distribution coefficient to various cements and rock,

(5) Long term corrosion behaviour of Zircaloy metal, stainless steels end-peace, Inconel spacer, etc.,

(6) Long term stability of hydrogen absorbed in Zircaloy metal.

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Fortum contribution to D6.1

Olli Nummi

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1 Geological Repository Concept

In Finland, the nuclear power companies are responsible for the management and disposal of the radioactive waste produced in their nuclear power plants (NPPs). The owners of the Loviisa and Olkiluoto NPPs have formed a joint company, Posiva, to implement their spent fuel disposal in Olkiluoto at the depth of approximately 400 m. The operational and decommissioning wastes from NPPs, classified as low and intermediate level waste (LILW), will be disposed of by the companies themselves in separate repositories at the power plant sites in Loviisa and Olkiluoto at the depth of 60-110 m. For more details of the radioactive waste handling in Finland, see e.g. [STUK 2014]. The focus here, however, is solely on the Loviisa LILW repository.

The Loviisa NPP is located in southern Finland, approximately 80 km East of Helsinki. The power plant is owned and operated by Fortum Power and Heat Oy. The plant consists of two VVER-440 units commissioned in 1977 and 1980 and the planned operating time for both units is currently 50 years. The on-site repository for the operational waste has been excavated in crystalline bedrock at the approximate depth of 110 m at the Hästholmen island and it has been in operation since 1998. The repository is dedicated for the waste from Loviisa NPP only. The caverns for the decommissioning waste will later be licensed and excavated as an extension of the repository as shown Figure 1.

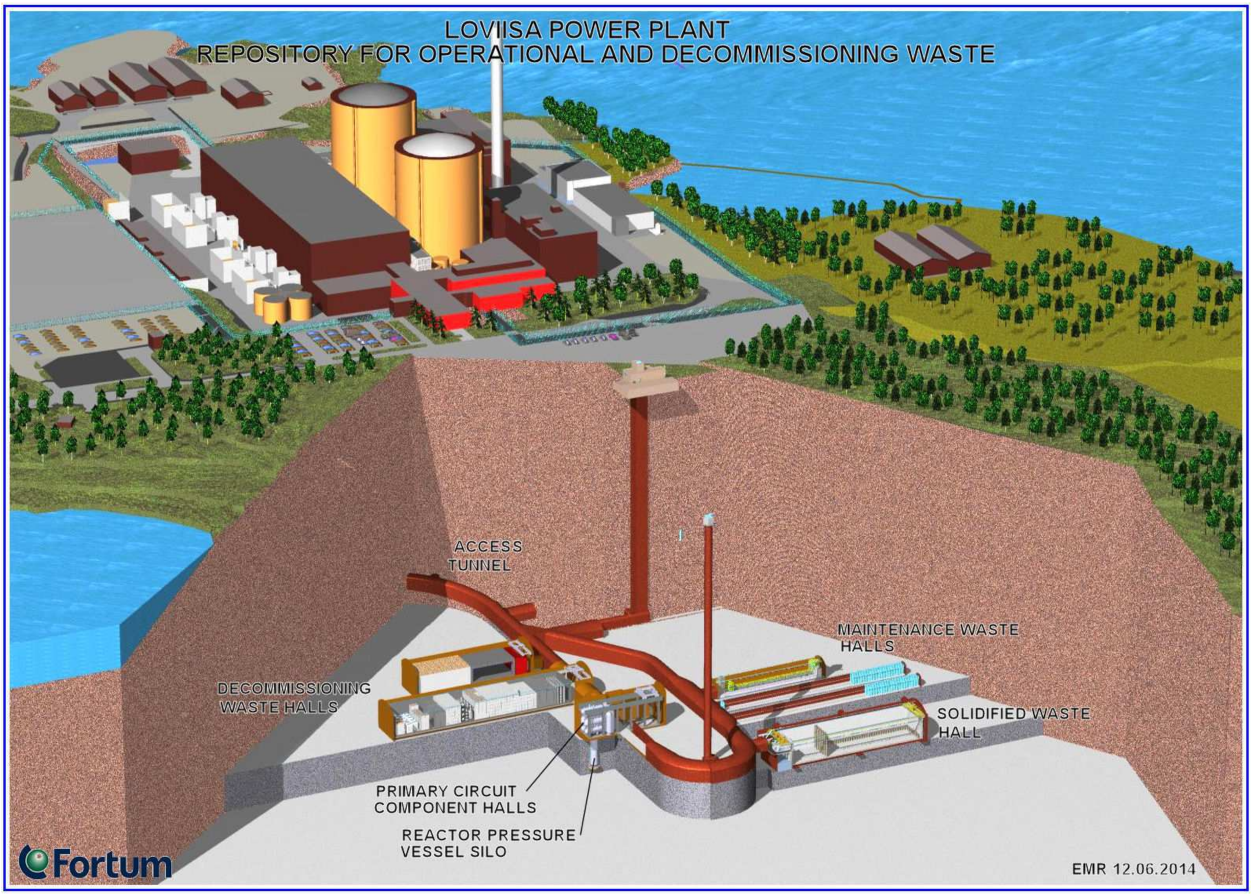

Figure 1: The LILW repository located at Loviisa NPP site. The existing waste caverns of the operational waste are located on the right side of the connection tunnel, whereas the future decommissioning waste caverns are located on the left.

The wastes will be placed in separate waste caverns. The operational wastes will be placed in

- Halls for maintenance wastes (two of which are in operation) containing dry maintenance wastes compressed in metal drums disposed as such.
- Hall for solidified liquid waste containing solidified liquid waste products cast inside concrete containers emplaced inside a concrete basin containing majority of C-14 in the operating wastes.

The decommissioning wastes will be placed in

- Reactor pressure vessel silos containing reactor internals packed inside the reactor pressure vessels. The reactor internals contain the majority of C-14 in the decommissioning wastes. The pressure vessels are surrounded by concrete walls.

- Hall for primary circuit components containing e.g. steam generators and pressurizers disposed as such.
- Halls for decommissioning wastes containing miscellaneous decommissioning waste such as activated concrete, small components etc. packed in concrete or wooden boxes. Almost all the wastes will be emplaced inside separate concrete basins, but low level wastes will also be placed outside the basins.

At the time of the closure, the crushed rock backfilling and concrete plugs at the entrance of the waste caverns and along the access tunnel will be set in place. These will limit the groundwater flow in the repository, protect waste and the waste packages from mechanical loads and prevent human intrusion.

The bedrock in the area is mainly rapakivi granite, of which the most common variation is pyterlite. The rock between the main fracture zones is fractured cubically, and via this fracture system the repository is connected to the sea. As to sorption of radionuclides, rapakivi granite can be considered quite favourable. Radionuclides dissolved in groundwater diffuse to the micropores in the rock matrix and are retarded due to sorption in the rock material.

There are two distinct groundwater zones at the repository area, common to the coastal area. A lens of fresh water accumulated from infiltrated rain floats on top of the saline ground water. The salt content of the saline groundwater is higher than that of the current seawater, which indicates that it mainly resides from the most saline period of the Baltic sea, thus having aged for 6000 - 10 000 years.

The evolution of the site is mainly driven by the glaciation-related crustal uplift which gradually changes the surface environment and the groundwater flow field and composition. The land uplift rate in the Hästholmen area in the recent years has been about 2 mm/a [Eurajoki 2006]. As a result of the uplift, the Hästholmen island will be connected to the mainland and the current sea bays located west and east of the island will eventually be transformed into lakes. As a result of the uplift the boundary between the fresh and saline groundwater regimes deepens and the groundwater type at the repository depth changes from saline to fresh water. The groundwater flow field will also change due to the effects of the land uplift.

2 C-14 Inventory

The C-14 inventory is estimated based on measurements and activation calculations and it is summarized in Table 1.

The C-14 inventory of solidified ion-exchange resins has been determined based on measurements. The C-14 activity in carbonate form was first determined using acid treatment after which the residual C-14 activity was measured by incineration. The exact composition of the residual C-14 is not known, but it is most likely organic. The main uncertainties in the inventory are due to uncertainties in the measurements of C-14 in the ion-exchange resins and variability between the samples. The amount of C-14 in other solidified liquid wastes and dry maintenance wastes is very low compared to ion-exchange resins.

In the decommissioning wastes, the C-14 inventory is estimated using activation calculations and based on operation experiences. The reactor pressure vessel and its internals, as well as the surrounding structures, mainly serpentine concrete and ordinary structural concrete are activated in the neutron irradiation and, hence, they contain most of the C-14 activity. The activation calculations have been carried out using MCNP calculation and nitrogen impurity content of 0.04 - 0.14 % in steels were used [Eurajoki & Ek 2008]. The main uncertainties in the C-14 activation calculations are due to material properties, especially the nitrogen content in the steel components. The chemical form of the C-14 released from the activated metal parts includes a high level of uncertainty and the subject is discussed in more detail in the safety case context below.

Table 1: Total realistic C-14 activity inventory resulting from two reactor units in different waste types at the time of the closure. In other waste types, C-14 activity is significantly lower.

Waste type	Waste packages	Average C-14 activity (GBq/kg)	Total C-14 activity (GBq)	Assumed speciation of C-14
Solidified ion-exchange resins	Concrete container placed inside a concrete basin	0.001	500	15 % organic 85 % inorganic
Reactor internals	Reactor internals packaged inside reactor pressure vessel	0.4	53 800	Organic or gaseous
Reactor pressure vessel	Disposed as whole and surrounded by a concrete wall	0.003	350	Organic or gaseous
Other decommissioning wastes	Concrete and wooden packages partly placed inside a concrete basin	0.0001	142	Organic or gaseous
Total			**~55 000**	

3 Safety concept

The safety concept of the LILW repository is based on multiple safety functions. These are the limited release of activity from the waste and the waste packages as well as slow groundwater flow rates inside the repository caverns. However, for the low-level waste (dry maintenance waste, part of the decommissioning waste) the safety functions related to the closure structures and natural barrier system (host rock) are considered to be sufficient.

The vast majority of the C-14 is bound to the activated steel and its release is governed by the steel corrosion. The waste packages are chosen based on the waste characteristics and its activity inventory. Additional barriers, such as the concrete around the reactor pressure vessels and concrete basins, limit the release of activity and protect the integrity of the waste packages. The concrete placed inside and outside the reactor pressure vessel creates a high-pH, alkaline environment which reduces metal corrosion rates of the reactor internals and the pressure vessel wall.

The backfilling and closure structures limit groundwater flow and minimize the effect of possible rock movements for the waste packages. The closure structures consist of concrete plugs, which effectively limit the groundwater flow between the repository caverns and the tunnels. The bedrock provides a stable barrier between the repository and the biosphere and it can be assumed to stay stable over relevant time-scales. The activity migration through the bedrock is governed by the slow groundwater flow rate in the bedrock fractures and possibly retarded by radionuclide diffusion and sorption into rock matrix. The retardation is insignificant for organic C-14, but has an effect for the migration of inorganic C-14.

4 Treatment of C-14 in the safety assessment

Separate safety cases have been compiled for operational [Eurajoki 2006] and decommissioning [Eurajoki 2008] wastes. In both safety cases a set of scenarios have been defined based on different assumptions regarding future evolution. The base scenario describes the expected evolution of the disposal facility and the disposal site. In the variant and disturbance scenarios alternative lines of evolution have been considered. The focus here is on the scenarios, which directly affect the release and migration of C-14 and thus all the scenarios considered in the safety cases are not discussed.

The key issues related to C-14 in all the scenarios are the release rate and the release form. The release rate from the solidified liquid waste was determined based on the expected performance of the waste matrix, concrete waste packages and concrete basin surrounding the packages. C-14 is released from the waste matrix and the waste packages via diffusion, and from the concrete basin via advective groundwater flow through the basin.

In activated metals, the release rate of C-14 is assumed to be solely governed by the corrosion rate of the steel, which is significantly affected by the steel type and prevailing pH conditions. The corrosion rates of stainless steel were assumed to lie between 0.01 - 1 μm/a depending on the pH conditions and the degree of conservatism. The diffusion out from the metal was also considered, but its contribution for the overall release rates was considered low during the time periods relevant for C-14. The reactor internals were divided into layers with specific activity concentrations in order take into account different activation rates at different depths. The instant release fraction from steels was assumed negligible.

Three main variants for the speciation of the C-14 were considered: carbonate (inorganic), organic gaseous substance and organic soluble substance. A literature survey [Kuitunen 2007] revealed that the most common carbon species released in the repository conditions is carbonate which precipitates almost completely in the alkaline repository conditions due to the low solubility of the calcium carbonate. On the other hand, there are experimental indications of both gaseous and soluble organic substances, but the applicability of those experiments to the repository conditions may be questionable [Kaneko et al 2003, Deng et al. 1997]. Furthermore, the C-14 speciation may change due to reactions either inside the repository or while migrating along the bedrock fractures.

Due to the related uncertainties, C-14 is conservatively assumed to be released in organic form from the activated metals in the base scenario and in gaseous organic form in the variant scenario. The C-14 is assumed to be released in organic form from the other decommissioning wastes in all the scenarios. In solidified liquid waste the proportion of C-14 released as organic and inorganic is based on measurements.

In all the scenarios, no solubility limits are applied for C-14 and no sorption is assumed for the organic C-14, whereas moderate sorption of C-14 into concrete and rock (K_d = $1 \cdot 10^{-3} \ldots 1 \cdot 10^{-4}$ m^3/kg) was assumed. The sorption retards the release of inorganic C-14 from the concrete waste packages and furthermore its migration in the repository caverns and bedrock.

4.1 *The base scenario*

The base scenario describes the most likely set of events affecting the disposal site. The C-14 released from the activated metal is assumed to be in organic form reflecting the uncertainty related to the C-14 speciation. The C-14 released from the solidified liquid wastes is divided between inorganic (85 %) and organic (15 %) species based on the measurements.

From the waste caverns the released C-14 migrates into biosphere along the fracture network in the bedrock. The groundwater analysis indicates that contaminated water from the repositories flows into the direction of a sea bay, which will be transformed into a lake thousands of years after the closure due to the land uplift. In the sea scenario the main pathway into the humans is the ingestion of fish, but in the lake scenario also irrigation water and drinking water for livestock are considered. The concentration ratio for C-14 in fish was chosen from the literature to have a relatively high value, whereas in reality the concentration ratio depends on the overall carbon concentration in the sea or lake.

4.2 Variant scenario: RPV gas scenario

In the reactor pressure vessel (RPV) gas scenario, the C-14 released from the activated metal is in gaseous form. Gas is assumed to migrate immediately to the primary circuit component hall located above the reactor silos, where it is assumed to be dissolved into groundwater. From there C-14 migrates along with the groundwater flow similarly as in the base scenario.

4.3 Disturbance scenario: drilled wells

The drilling of the wells is assumed to occur after administrational control period ceases 200 years after the closure. The well locations are chosen according to the characteristics of the ground surface, in such a way that they intersect large fracture zones. Drilled wells affect the groundwater flow field depending on their size and location. In the well scenario, the major pathways in the biosphere model are drinking water for humans and livestock as well as irrigation of field and garden.

4.4 Resulting dose rates

In the base scenario, where the contaminated water flows into the direction of the sea or lake the doses are generally lower than in the scenarios with drilled well and, therefore, they are not discussed here in detail. The focus here is on the doses arising from the disturbance scenario with drilled wells. The well probability of 10 % is included in the expectation value of the results and the expectation value can be used as a comparison to the regulatory requirements [STUK 2013].

In Table 2, the doses arising from the use of contaminated water from a small drilled well for household purposes are presented. The realistic and conservative calculation cases differ by their selection of parameters, examples of which are groundwater flow rates, distribution coefficients, steel corrosion and concrete degradation rates. The realistic case mainly uses best estimate values, whereas in the conservative case estimated upper or lower limit values are applied. The well locations are different in the cases of solidified waste cavern and decommissioning waste silos and hence the resulting annual dose rates are not combined. Also the release mechanisms of C-14 from these wastes are different. Usually, C-14 is the most dominating nuclide, partly due to its related uncertainties [Eurajoki 2006, 2008].

Table 2: Expected dose rate from C-14 in the small wells, including well probability of 10 %. In case of solidified waste hall, a different well is chosen. The sum overestimates the results as the maxima do not necessarily occur simultaneously. The regulatory constraint for expected dose rate is 0.1 mSv/a.

	Repository cavern	**Speciation of released C-14**	**Maximum expected dose rate (mSv/a)**	
			Conservative calculation case	**Realistic calculation case**
Operational waste	Solidified liquid waste hall	Organic (15 % Total C-14)	1E-02	7E-05
		Inorganic (85 % of total C-14)	5E-04	5E-06
	Sum		**1E-02**	**8E-05**
Decomm. waste	Reactor pressure vessel silo 1	Organic	8E-04	4E-05
		Gas	1E-02	2E-03
	Reactor pressure vessel silo 2	Organic	1E-03	4E-05
		Gas	1E-02	2E-03
	Decommissioning waste halls	Organic	5E-03	4E-03
	Sum	**Organic**	**7E-03**	**3E-03**
		RPV Gas + DW halls organic	**3E-02**	**8E-03**

The temporal behaviour of the annual doses are presented in Figure 2 indicating that maximum dose rates caused by C-14 do not occur simultaneously from the operational and decommissioning wastes. The effect of different release mechanisms can also be seen in Figure 2. From the reactor pressure vessel silos, the C-14 is released from the pressure vessel at almost constant rate during the first thousands of years after which the effect of radioactive decay can be seen. The sharp increase roughly 30 000 years after the closure is caused by the full corrosion of the pressure vessel wall causing a sudden release of activity initially released inside the pressure vessel from the reactor internals. On the other hand, the dose rate caused by the C-14 in the solidified waste product is strongly dependent on the performance of the concrete basin and the concrete containers. In the base scenario visualized in Figure 2, the degradation of concrete basin initiates 500 years after the closure and it is fully degraded 2000 years after the closure. These phenomena can be seen in the dose rates caused by the organic and inorganic C-14.

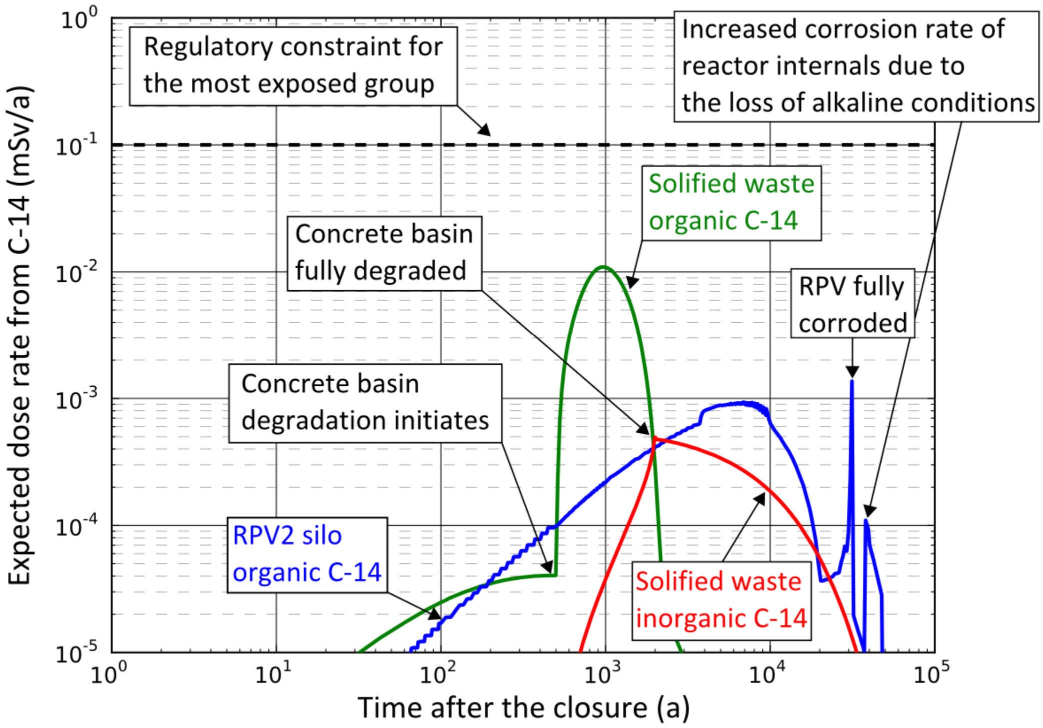

Figure 2: The doses arising from solidified waste cavern and reactor pressure vessel silo 2 (RPV2), calculated using conservative parameter values. The dose rates from the both reactor pressure vessel silos are almost identical and hence only RPV2 results are shown. The well probability of 10 % has been taken into account in the expected dose rate.

5 Key issues and priorities

Currently, C-14 is one of the most dominating nuclides causing the radiation dose after the closure of the Loviisa LILW repository. However, the uncertainties related to C-14 speciation can be considered significant and hence conservative assumptions have been used in the safety assessments. The reduction of key uncertainties related to C-14 leads to less conservative assumptions lowering the dose rate and to a more balanced safety assessment. The key uncertainties are listed below.

- The speciation and inventory of C-14 in activated metal parts in reactor internals
- The release rate of C-14 due to corrosion from the metal parts

- The speciation and inventory of C-14 in the ion-exchange resins
- The treatment of C-14 in the biosphere.

The uncertainties related to the first three items above are expected to be reduced based on the results of the CAST project. These include experimental results as well as deeper understanding of the subject.

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CArbon-14 Source Term

CAST

LEI contribution to D6.1

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1 Geological Repository Concept

The main (but not only) C-14 source is irradiated graphite from Lithuanian nuclear power plant at Ignalina site. Irradiated graphite from RBMK[1]-1500 reactor is classified as long-lived low level waste (LLW-LL). After dismantling such wastes (including irradiated graphite) will be stored in the steel containers at an interim storage facility until the final decision on their final disposal option will be made [POSKAS, 2012]. Currently waste acceptance criteria for the near surface repostory (NSR) do not foresee disposal of graphite waste in the NSR in Lithuania thus its final disposal option is remaining and open.

Lithuania is in the generic (early conceptualisation) stage for long-lived waste and spent nuclear fuel (SNF) disposal. Thus, there is no final decision on the long-lived intermediate and low level waste disposal option, disposal container or host rock yet. However, with implementation of the EC directive 2011/70/EURATOM, the plans and important milestones are under development as it must be reported to EC till August 2015.

For the preliminary analysis of radionuclide release and transport, the following concept was considered. The long-lived low level waste could be disposed at the same repository at a certain distance from SNF emplacement tunnels according to proposed generic repository concept of RBMK-1500 SNF disposal in crystalline rock in Lithuania [POSKAS, 2006]. Waste emplacement tunnels could be app. 16 × 16 m in cross-section. Within this study, the cementitious grout (Nirex Reference Vault Backfill (NRVB)) was assumed as proposed in NIREX concept (United Kingdom) to fill void regions within the tunnels after the emplacement of the LLW-LL.

According to proposed generic repository concept metal containers could be used for the disposal of LLW-LL. As the dimension of the waste package has not been developed in Lithuania yet, thus the Generic Specifications [NIREX, 2007] developed in UK were analysed and the information on the waste package was used in terms of geometrical data

[1] "RBMK" is a Russian acronym for "Channelized Large Power Reactor" which is water cooled graphite-moderated reactor.

(Figure 1). The height of the metal container is 2.2 m, the length is 4.013 m, the width is 2.438 m, thickness is 0.002 m (Fig.2) based on [NIREX, 2007, TOWLER, 2010] .

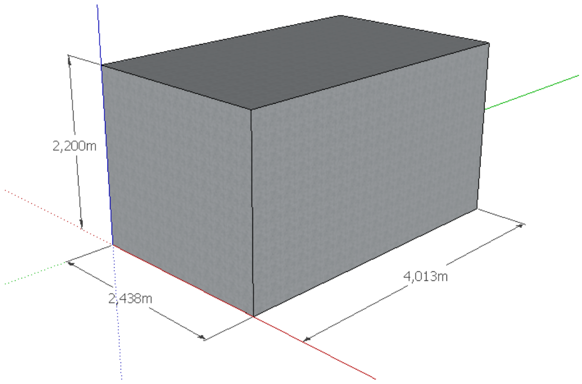

Figure 1: Steel container considered for the analysis of C-14 migration from RBMK-1500 graphite waste

For the possible waste grouting a cementitious material (encapsulant) was assumed. Its thickness inside the container was considered to be 0.15 m. Total internal volume of the container of these dimensions is 18.9 m^3. Taking into account the packing efficiency of 0.67 total internal volume to be occupied by waste 12.7 m^3 [TOWLER, 2010].

The disposal tunnels were assumed to be constructed in crystalline host rocks. The tunnels will be oriented in such way that it would lead the radionuclides released to be transported along the disposal tunnel to take more time for chemical interaction with backfill material. Containers stack is of app. 10 m height and of 13 m width. Waste containers would be emplaced in these tunnels with a spacing of 0.9 m. Within the tunnel of 16 × 16 m dimensions, 4 containers in horizontal and vertical directions could be stacked (Figure 2). The upper part of the tunnel has to be sufficient for the container handling equipment.

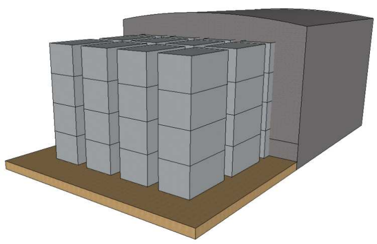

Figure 2: Graphite waste disposal tunnel considered for the modelling

In case of RBMK-1500 graphite disposal in the crystalline rocks in Lithuania the far field would be constituted by the crystalline rocks covered by sedimentary rocks. Hydrogeological cross-section of Lithuania is presented in Figure 3.

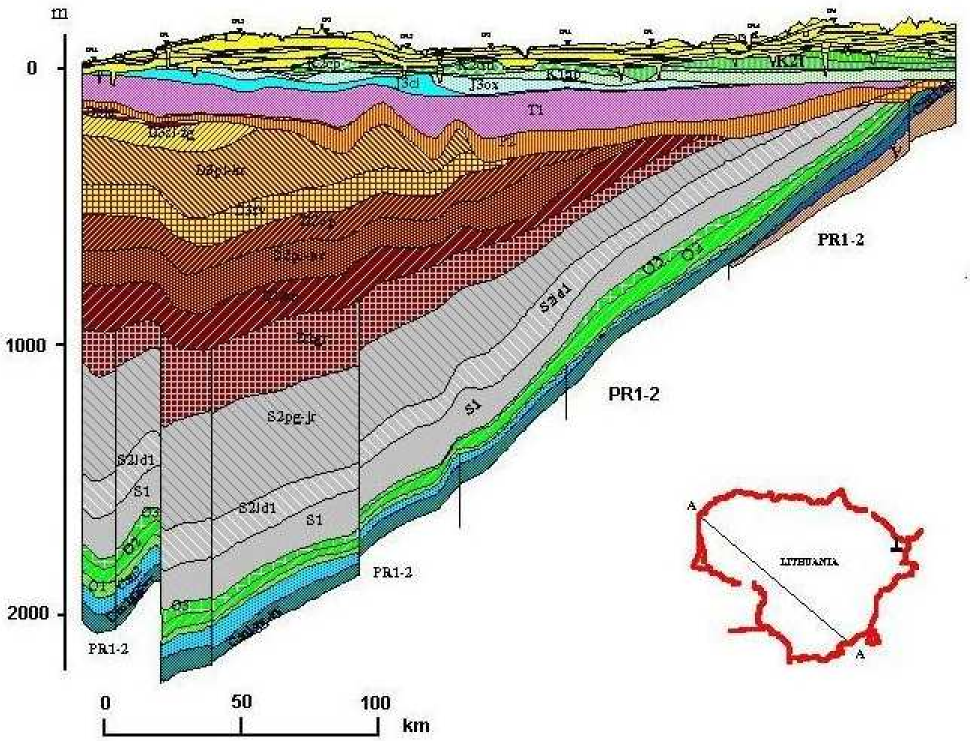

Figure 3: Hydrogeological cross-section of Lithuania; crystalline basement is marked as “PR1-2” (Authors: L. Kilda, S. Šliaupa, J. Lazauskienė)

In the southern Lithuania crystalline rocks occur at the depths ranging from 210 m to 700 m, while in most of Lithuania territory the depth of the basement exceeds 700 m, reaching 2300 m in the west. The crystalline rocks underlie the sedimentary rocks of different hydrogeological properties (forming aquifers and aquitards). For the radionuclide transport analysis, the repository was assumed to be located in the crystalline rock at the depth of 500 m under app. 400 m thick sedimentary rocks cover [NARKUNIENE, 2013].

2 C-14 Inventory

For Lithuania, the main C-14 source is irradiated graphite from Lithuanian nuclear power plant (NPP) at Ignalina site. Two units of Ignalina NPP were equipped with RBMK type reactors containing graphite as a moderator and reflector. After the dismantling of both reactor cores app. 3800 tonnes of graphite blocks and sleeves will be accumulated [POSKAS, 2012]. Graphite rings/sleeves are placed around fuel channels and ensures thermal contact between graphite blocks and fuel channels. The graphite blocks are of GR-280 type and the graphite sleeves are of GRP-2-125 type. Different graphite grades mean different impurities in it, different inventories and different behaviour. This was one of the main message out of the CARBOWASTE project and this message should be spread as wide as possible.

The C-14 activity depends on the neutron flux (location in the reactor core), operating power history, initial concentration of impurities, amount of cooling gases, etc. Numerical modelling of C-14 inventory was performed taking into consideration a possible C-14 generation from C-13 and N-14 in RBMK graphite. Taking into account maximal initial nitrogen impurities (70 ppm (weight)) in graphite matrix and nitrogen present in all (open and closed) graphite pores, the estimated maximum activity of GR-280 type graphite (blocks) is up to 9.9×10^5 Bq/g (activity of blocks is higher than of graphite sleeves) [NARKUNAS, 2010].

Up to now there are only few measurements performed regarding C-14 inventory in RBMK graphite besides numerical estimations [REMEIKIS, 2010]. Short leaching test for RBMK graphite was performed in the frame of CARBOWASTE project too [DRUKTEIKIENE, 2013].

3 Safety concept

The safety concept has not been developed as the safety assessment according to IAEA's definition [IAEA, 2011] has not been carried out for irradiated graphite in Lithuania. As it has been noticed there is no decision on graphite final disposal on national level in Lithuania. Thus neither safety concept nor repository concept is developed and approved and no safety assessment is carried out.

4 Handling of C-14 in safety assessment

As mentioned above, full scale safety assessment has not been developed yet. LEI analysed the C-14 migration from irradiated graphite disposed of in a generic geological repository [NARKUNIENE, 2013] under expected conditions. The relation between treatment and direct disposal of graphite on repository performance was analysed by the way of radionuclide transport modelling from geological repository, as the radionuclides eventually released from the irradiated graphite waste could be transported to the surface and lead to human exposure [NARKUNIENE, 2013].

C-14 migration in the near field and far field was modelled with the source term based on LEI results on RBMK-1500 inventory, illustrative rates for representation of possible differences on non-treated/treated graphite and conceptual models developed. Treatment (crushed, powdered graphite) leads to removal of rapidly released fraction of inventory – represented by leaching rate over time. The importance of waste leaching rate was studied within the context of different performance of engineered barrier (in terms of sorption, limited solubility) and considering possible graphite encapsulation in cementitious material (Table 1).

There were no results from leaching tests for RBMK-1500 graphite available during the study. Due to lack of empirical data, the illustrative leaching rates (Table 1) were selected in this study to represent possible differences on non-treated/treated graphite for the analysis of C-14 migration from the graphite disposed of and to evaluate the impact of these differences on the radionuclide flux to geosphere. Treatment leads to removal of rapidly released fraction of inventory, this effect is reflected by annual leaching rate over time.

The importance of waste leaching rate was analysed within the context of different performance of engineered barrier considering three different cases each supplemented by a number of variants (Table 1) [NARKUNIENE, 2013].

Table 1: Analysed cases and variants based on leaching rate and cementitious barrier performance

Cases		
Case 1: K_d=0, no solubility limitation		
Case 2: K_d=0, solubility limit=0.01 moles/m^3		
Case 3: K_d=0.2 m^3/kg, solubility limit=0.01 moles/m^3		
Variants based on annual fractional leaching rate of C-14 from graphite (for each Case):		
Variant A	1.83×10^{-5} (1/year)	Experimentally measured rate [LIMER, 2010]
Variant B1	<10 yr. - 0.1 (1/year) >10 yr. - 0.01 (1/year)	Corresponds to not treated waste, higher leaching rate [LIMER, 2010]
Variant B2	<10 yr. - 0.1(1/year) >10 yr. - 0.001 (1/year)	Corresponds to not treated waste, lower leaching rate [LIMER, 2010]
Variant C1	0.1 (1/year)	Corresponds to treated waste, highest leaching rate [LIMER, 2010]
Variant C2	0.01 (1/year)	Corresponds to treated waste, lower leaching rate [LIMER, 2010]
Variant C3	0.001 (1/year)	Corresponds to treated waste, the lowest leaching rate [LIMER, 2010]
Variant D	instant release from waste	Barrier function of solid waste matrix disregarded

Porous medium (continuum approach) was applied for modelling the groundwater flow and contaminant migration in the larger scale model, comprising the first fractured natural barrier and the subsequent ones (crystalline and sedimentary rocks) (Table 2).

In this case, the analysis was performed for the radionuclides released from the crystalline rocks and being transported upward to the groundwater discharge area. The conceptual model (Figure 4) of the repository is based on the conservative assumption that the repository would be located in the area where the groundwater flow is upward, thus the distance to the surface is shorter than could be expected during the designing of real repository. No sorption was assumed for geological layers. Transport in the biosphere was not analysed in LEI study.

Table 2. Hydrogeological data of natural barriers

Layer no.	Depth (m)	Hydraulic conductivity (m/s)	Porosity (m^3/m^3)	Description
1	0–100	$1.2 \cdot 10^{-5}$	0.25	glacial loam
2	100–200	$2.3 \cdot 10^{-5}$	0.35	sand
3	200–250	$5.8 \cdot 10^{-5}$	0.5	anhydrite
4	250–270	$1 \cdot 10^{-6}$	0.5	limestone
5	270–290	$5.8 \cdot 10^{-5}$	0.05	sandstone
6	290–310	$1.16 \cdot 10^{-10}$	0.13	clay
7	310–410	$6 \cdot 10^{-6}$	0.05	sandstone
8	410–420	$1.16 \cdot 10^{-7}$	0.01	weathered crystalline rocks
9	420–520	$1.16 \cdot 10^{-13}$	0.0038	monolithic crystalline rocks

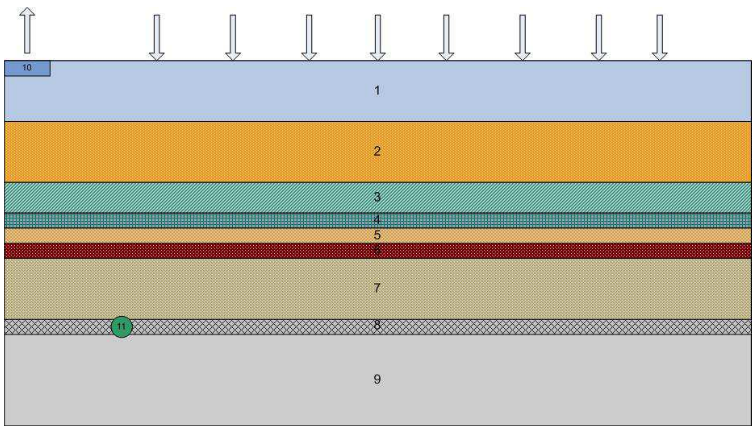

Figure 4: Far field model (1 – glacial loam, 2 – sand, 3 – anhydrite, 4 – limestone, 5 – sandstone, 6 – clay, 7 – sandstone, 8 – weathered crystalline rocks, 9 – monolithic crystalline rocks, 10 – groundwater discharge area, 11 – radionuclide flux from the crystalline rock)

Modelling of the radionuclide migration within the near and far field was performed using AMBER (UK) and TOUGH2 (USA) computer codes. For C-14 migration in the near field advective/diffusive transport model was assumed. Groundwater flow rate was calculated based on assumed prevailing head gradient and rock hydraulic conductivity. Analysis of C-14 migration in the far field was performed for dissolved radiocarbon (in liquid phase). Exact speciation during the transport along the emplacement tunnels was not considered in the near field model while chemical interaction (sorption, limited solubility) with engineered materials was taken into account by the means of linear sorption coefficient and solubility limit. In absence of knowledge over the C-14 speciation, no sorption/no solubility limit cases were considered to take into account possible radiocarbon speciation into species which are transported without retardation. Modelling was made for two options, using the reference near field model (for non-encapsulated waste) and alternative near field model (considering possible encapsulation in cementitious material).

Modelling results

Modelling results of C-14 transport with no interaction in the near field showed that the differences in leaching rates do not lead to the same differences in the peak flux to the geosphere (see Figure 5). The radionuclides released from graphite by the rates of the order of 1×10^{-2} – 1×10^{-1} 1/yr (variants B1, B2, C1, C2) were released to the geosphere at similar rate as in case of instant release. If the radionuclides were released at lower rates (or the order of 1×10^{-3}, 1×10^{-5} 1/yr. with RBMK-1500 inventory), the flux to the geosphere becomes more dependent on the leaching rate.

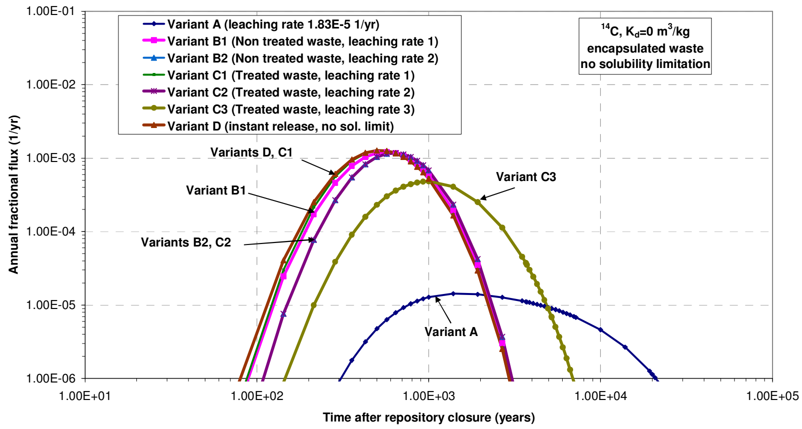

Figure 5: Release of C-14 to the geosphere from one disposal container with not encapsulated waste (results of Case 1, sorption and solubility limit disregarded)

Due to transport and chemical interaction with a tunnel backfill material (in terms of sorption and solubility), the C-14 flux to the geosphere was significantly decreased and the maximal fractional flux is in the range of 1×10^{-12}–1×10^{-11} 1/yr. (Case 3) in comparison to 1×10^{-5}–1×10^{-3} 1/yr. without consideration of these two processes. If we consider that the release of C-14 is solubility limited, then a plateau in the flux profile was observed (maximal flux observed of the order of 4×10^{-5} 1/yr., Case 2) for all leaching rates except the lowest one (1.83×10^{-5} 1/yr) also indicating the higher importance of sorption process in decreasing of the maximal flux from the near field.

The results for encapsulated and not encapsulated graphite are presented in Figure 6. As it could be seen considering sorption, the impact of waste encapsulation is significant and lead to the decreased C-14 peak flux by up to app. one order of magnitude in comparison to not encapsulated waste. If sorption and solubility processes do not occur, there is no significant effect on the maximum release rate from the near field (peak flux).

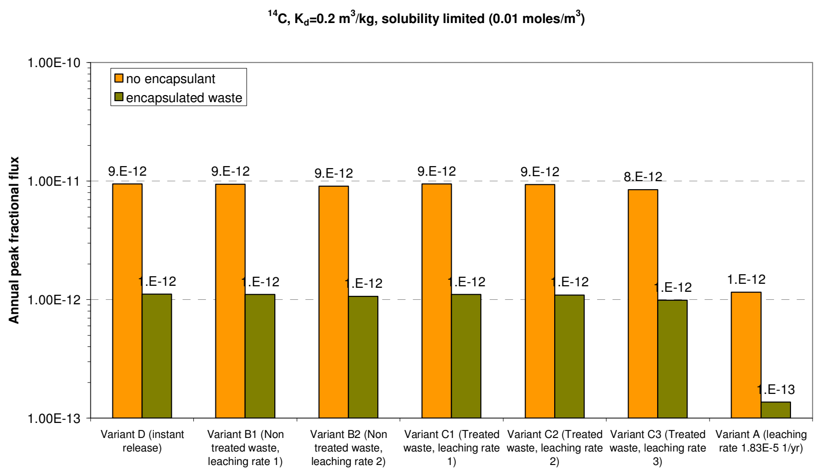

Figure 6: Comparison of peak release for not encapsulated and encapsulated waste (results of Case 3, considering sorption and solubility limitation)

Results on C-14 flux in Case 1 (sorption and solubility limit disregarded) and instantly released inventory from the waste matrix were transferred to the far field model built in TOUGH2. Modelling results (Figure 7) showed that the system of natural barrier contributes to the significant delay of release of radionuclides even with conservative assumption on engineered and natural barriers, and a significant decrease of it maximal peak flux rate by at least 5 orders of magnitude was observed.

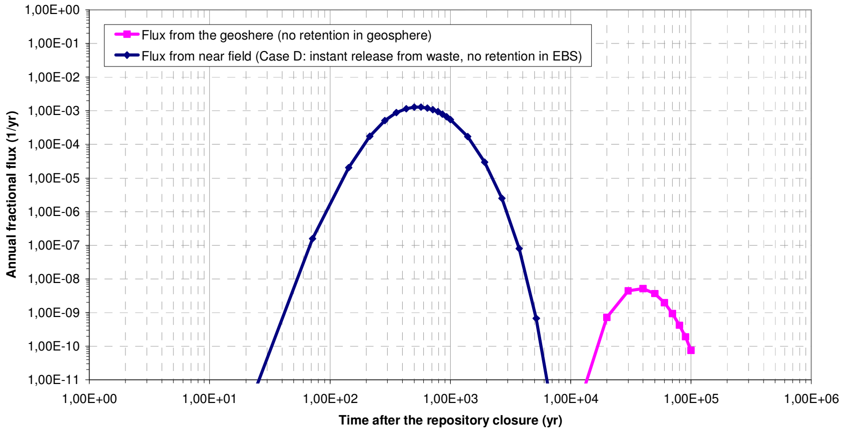

Figure 7: Release of C-14 from the backfill and geosphere

Peak fractional flux of dissolved C-14 from the near field and far field was compared to the transfer rate metric based on UK graphite inventory and generic biosphere model [TOWLER, 2012]. This metric corresponds to the transfer rate which could give a rise to the impacts from C-14 inventory of UK graphite around the regulatory guidance levels. Modelling results showed that for much lower amount of RBMK-1500 graphite than in UK case the peak fractional flux would be less than the transfer rate metric even with conservative assumptions on the near field and far field performance. The compliance with the metric (7×10^{-6} 1/yr.) would support the geological disposability of RBMK-1500 graphite in respect of C-14 migration in solution.

Hypothesis made in the analysis should be treated as “a priori” as the magnitude of chemical interactions, assumed groundwater flow conditions were not supported by reference site data. C-14 inventory used in the analysis should be treated as conservative as it was based on assumption of maximal initial nitrogen impurities in graphite matrix and nitrogen present in all (open and closed) graphite pores.

5 Key issues and priorities

The importance of waste leaching rate depends on several aspects: on the performance of the backfill and natural barrier system (on the scope of its impact on the attenuation of the radionuclide flux). The impact of the options (treatment vs no treatment of graphite) on the

C-14 flux to geosphere is not straightforward. While reasoning of option of treatment/not treatment the inventory, leaching rates, barrier performance and transport conditions need to be considered.

The analysis was performed with conservative assumptions on the inventory. The analysis based on the best estimates of C-14 inventory was not performed and the uncertainties related to the inventory have not been treated.

The uncertainty of RBMK-1500 graphite inventory is expected to be evaluated at least qualitatively within the CAST project. Uncertainty in leaching rate is expected to be bounded during CAST (based on other than RBMK-1500 graphite). This information will be used to update the C-14 transport analysis together with implementation of available Lithuanian site-specific information.

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CArbon-14 Source Term

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NRG contribution to D6.1

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1 Geological Repository Concept

In the Netherlands, extended (intermediate) surface storage of all radioactive waste in buildings has been chosen as the preferred policy. In due time, deep geological disposal in suitable host formations is envisaged for all categories of radioactive waste. Due to the relatively small amounts of radioactive waste, no separate disposal facilities for LILW and HLW are presently envisaged [MinEZ, 2011].

During the interim storage period the geological disposal is prepared financially, technically and socially in such a way that it can be implemented when deemed necessary. At some point during the intermediate storage period (until around 2100), a decision is foreseen with the options being (1) to continue the surface storage, (2) to realize the GDF, or (3) to use new techniques or management options that may have become available during the period of interim storage. If it is decided to construct a geological disposal facility, it should become operational after 2130 [Verhoef, 2014a].

COVRA, the Dutch waste management organisation, is considering the safety and feasibility of disposing of all radioactive wastes generated in the Netherlands in a geological repository. For a thorough substantiate of this topic the five-year research programme for the geological disposal of radioactive waste, OPERA[1], was established in 2011. The main objective of the OPERA research programme is to provide tools and data for the development of Safety Cases for Dutch repository concepts for radioactive waste disposals in two host rocks present in the Netherlands, Boom Clay, which is the primary focus of OPERA, and rock salt [Verhoef, 2011].

The disposal concepts for geological disposal in Boom Clay and rock salt are illustrated in Figure 1 [Verhoef, 2014b] and Figure 2 [Heijdra, 1997] respectively. The underground facilities contain four separate waste disposal sections: for vitrified HLW, for spent fuel from research reactors, for non-heat-generating HLW and for the disposal of ILW/LLW and

[1] OPERA: *OnderzoeksProgramma Eindberging Radioactief Afval*; Dutch acronym translating as *Research Programme into the Geological Disposal of Radioactive Waste*

depleted uranium. Figure 3 shows an artist impression of the HLW disposal in Boom Clay [Verhoef, 2011] and rock salt [Heijdra, 1997].

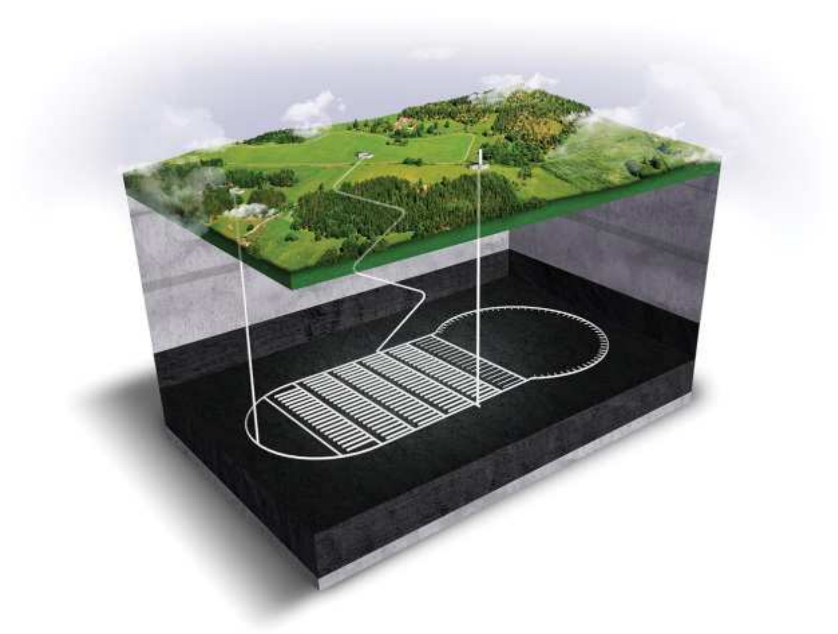

Figure 1 Reference design for the disposal in Boom Clay in the Netherlands

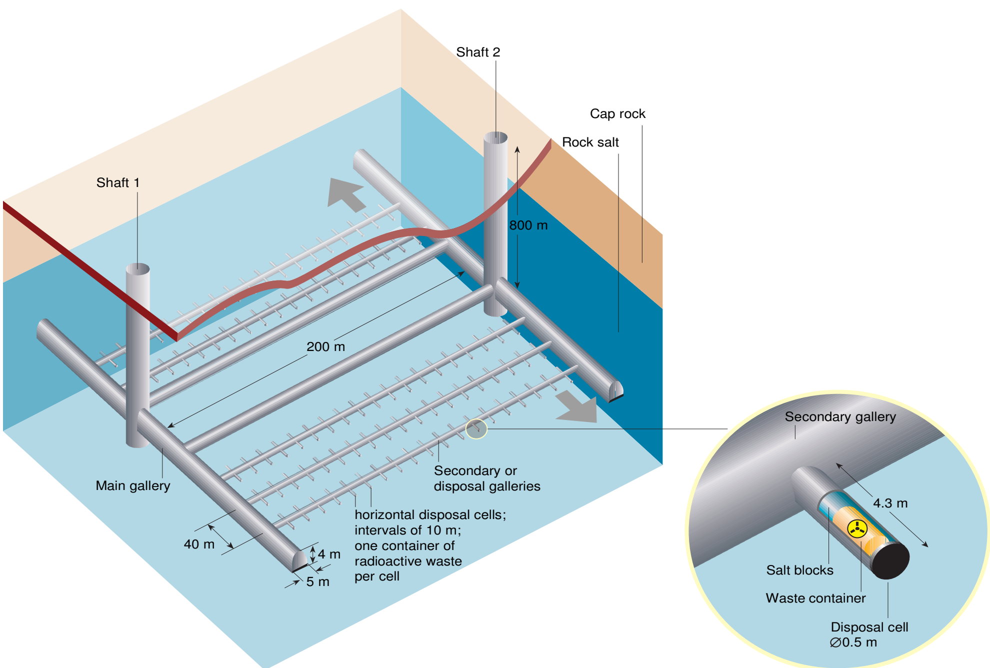

Figure 2 Concept for the disposal in rock salt in the Netherlands

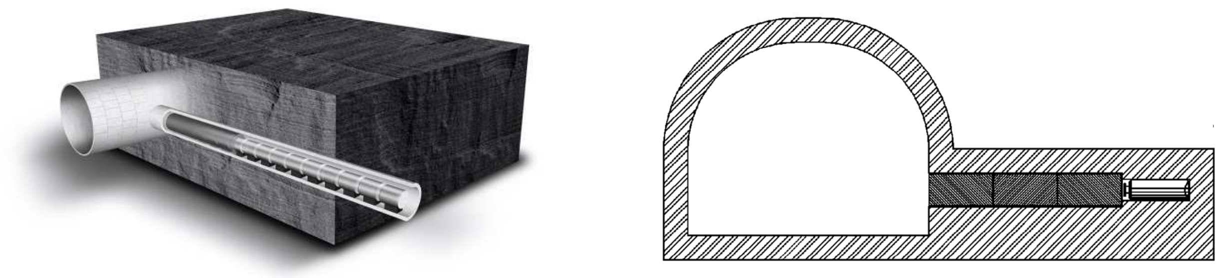

Figure 3. Artist impression of the HLW waste in clay (left) and rock salt (right)

In case of disposal in a low permeable Boom Clay formation, the vitrified heat-generating HLW and the spent fuel (from research reactors) will be packed in concrete supercontainers and placed in disposal drifts with a length of 45 m (see Figure 3, left). The heat-generating HLW section is located at the inside of the curved part of the repository (see Figure 1). This would allow a modular extension of the HLW section at the outside of the curved part. The other wastes are emplaced in 200 m long disposal drifts.

The disposal facility in rock salt contains short horizontal disposal cells drilled into the side walls of a gallery, each accommodating one HLW container (Figure 2, and Figure 3, right). The disposal cell is sealed off with compacted salt blocks.

Retrieval of already emplaced waste containers is considered technically feasible, and is enhanced applying a disposal concept with short, horizontal disposal cells and/or structural adjustments to maintain accessibility, e.g. linings or additional support to prevent borehole convergence and eventual collapse of the disposal drifts [CORA, 2001].

2 C-14 Inventory

The C-14 waste inventory consists of several parts, in analogy with the anticipated waste sections in the ultimate deep geological repository:

- Low and intermediate level waste;
- Non heat producing high level waste;
- Spent fuel and target materials from research reactors;

- Vitrified high level waste from reprocessing of spent fuel from the Dutch nuclear power plants.

The data presented below in Table 1 have been obtained within the OPERA research program [Hart, 2014a,b]. In OPERA Work Package 1, seven future nuclear energy scenarios have been defined and studied in order to estimate the amounts of waste by the year 2130, viz. the anticipated time of disposal. In three scenarios no new nuclear power plants (NPPs) are assumed, whereas in four scenarios it is assumed that new NPPs will be built, generating up to about 5000 MWe in the year 2040. For these last-mentioned four cases the estimated C-14 quantities largely depend on the applied future nuclear technologies. Consequently the calculated amounts comprise a significant uncertainty.

Table 1 Estimated inventories (Bq) of C-14 in the separate waste sections for the year 2130

Waste type	C-14 Activity (Bq)	Uncertainty
No New Nuclear Power Plants [Hart, 2014a]		
Low and intermediate level waste	2.25E+12 (min) 2.75E+12 (best est.) 3.30E+12 (max)	Significant; estimates based on extrapolations of LILW emplacement rate during last 30 years.
Non heat producing high level waste	2.07E+12	Significant; uncertain estimates of C-14 quantities in decommissioning waste
Spent fuel and target materials from research reactors	1.33E+13	Moderate; assuming deployment of one new research reactor
Vitrified high level waste from power plants, including hulls and ends (CSD-V + CSD-C)	1.72E+13	Moderate
Assuming New Nuclear Power Plants (5000 MWe by 2040) [Hart, 2014b]		
Low and intermediate level waste Non heat producing high level waste	Not estimated	Significant. Total C-14 quantities are judged to be considerably smaller than in SF/HLW waste (to be confirmed)
Spent fuel and target materials from research reactors	1.33E+13	Moderate; assuming deployment of one new research reactor
Spent fuel and/or vitrified high level waste from power plants, including hulls and ends	2.27E+13 (min) 4.45E+14 (max)	Significant. Specific activities for the different waste component parts were not estimated.

3 Safety concept

Since 1984, the policy in the Netherlands is that all hazardous and radioactive waste must be isolated, controlled and monitored (ICM) [VROM, 1984]. For a geological disposal this can be achieved by the following strategic choices:

(I) The facility will be constructed at sufficient depth to take into account the impact of surface phenomena. The host rock and geological environment should provide an effective containment of the emplaced waste and isolation from the biosphere. The depth of the facility should be sufficient to take into account the impact of surface phenomena such as glaciation.

(I) The facility will be constructed within a Tertiary Clay formation or Zechstein rock salt formation. Past and ongoing research in The Netherlands, Belgium and Germany indicates that, due the very low permeability, the plasticity and other favorable characteristics, these rocks can provide very long term containment, allowing the waste to decay while it is contained in the rock formation.

(C) The facility has to be designed, operated and closed such that the process is reversible and the waste is retrievable.

(M) Geological disposal planning will assume that surveillance and monitoring will continue for as long as deemed necessary.

At present, the Dutch safety concept for the geological disposal of radioactive waste in Boom Clay is being developed in the OPERA research programme [Verhoef, 2011]. The Boom Clay concept relies on the consideration of multiple barriers and safety functions that can be attributed to the subsequent barriers [Verhoef, 2011; Section 4.1]. This safety concept would in principle also be applicable for the disposal in salt-based repositories.

Multiple barriers

The principle of a multi-barrier system was already recognized in the Netherlands in the 1980's for the geological disposal in rock salt (OPLA, 1989; p. 35). The geological disposal concept in Boom Clay, adopted in OPERA, relies on a sequence of complementary and/or redundant barriers (defence-in-depth, see also Figure 4, right) (Verhoef, 2011a; p. 8). In principle, the multi-barrier systems adopted for the disposal in rock salt and Boom Clay do not differ significantly.

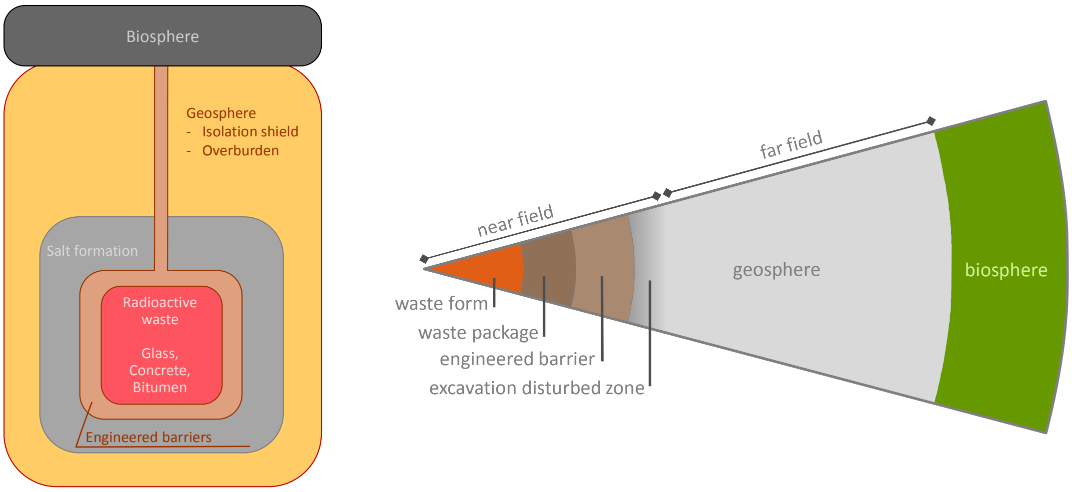

Figure 4. Principle of a multi-barrier system in salt formations (left), and in Boom Clay (OPERA, right)

Safety Functions

In OPERA, the safety functions as defined in [ONDRAF/NIRAS, 2009, Section 3.4.] have been adopted. Figure 5 gives a graphical presentation of the safety functions for the disposal in Boom Clay. The indicated timescale for the thermal phase applies to HLW. For the final disposal in rock salt no safety functions have yet been formulated.

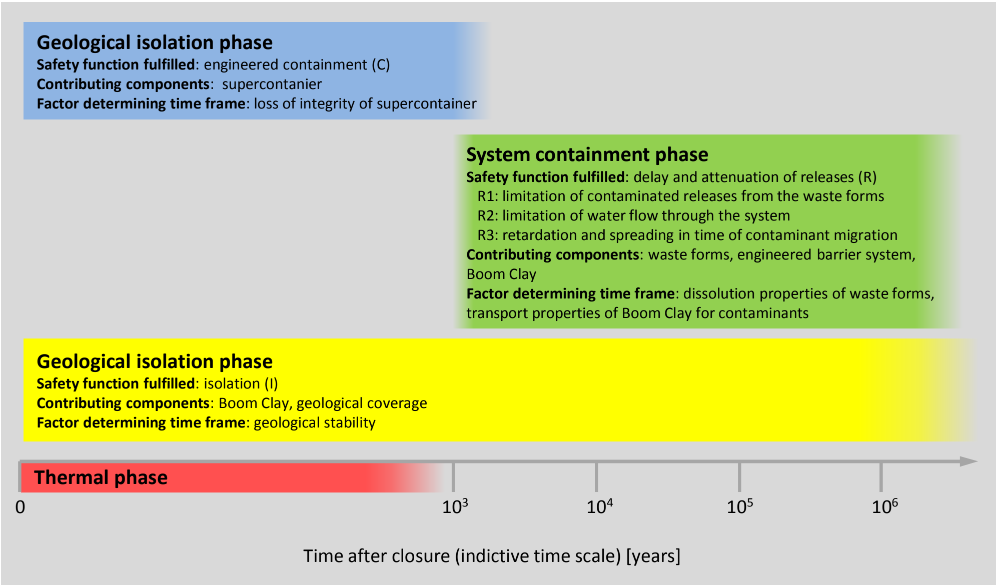

Figure 5. Safety functions provided by the main components of the disposal system in Boom Clay and its geological coverage

4 Treatment of C-14 in safety assessment

Safety assessments for the disposal in rock salt have been performed in the VEOS [Prij, 1989], PROSA [Prij, 1993], and METRO [Grupa, 2000] studies. Results from the OPERA safety assessment studies for the disposal in Boom Clay are not yet available. These studies will address the normal evolution scenario, and also some altered evolution scenarios.

4.1 *The normal evolution scenario*

The Normal Evolution Scenario (NES) is considered the most likely scenario. The NES assumes normally progressing and undisturbed construction, operation, "passive" operation, and closure of the facility. However, natural processes affect the facility's engineered barriers in the long term.

Under in situ conditions in Boom Clay, carbon can exist in three stable forms: HCO_3^-, CH_4 and organic derivatives. The bicarbonates (HCO_3^-) are the dominant species [ONDRAF/NIRAS, 2001]. All C-14 species will dissolve in the ground water, mostly as

HCO_3^-. The dissolved C-14 can migrate through the clay rock formation by diffusion only. The time to reach the overlying aquifer[2] by diffusion is on average about 100 000 years. Since the half-life of C-14 is 5700 years, this means that virtually all C-14 will decay in the clay host rock.

In the case of disposal in rock salt, the waste is completely enclosed by several hundred meters of dry rock salt. Consequently, all C-14 will decay in the facility. The displacement of air from the mine (potentially including C-14), caused by the convergence of the rock salt has not yet been taken into account [Grupa, 2000; p.11].

4.2 *Altered evolution scenarios*

The altered evolution scenarios include consideration of potential faults, abandonment and construction/design failures leading to early flooding of the facility. Although transport times are shorter in these scenarios, C-14 always gave a small contribution to the exposure compared to other radionuclides relevant for the long-term safety.

In all considered altered evolution scenarios C-14 is dissolved in groundwater. Relative little effort has been put in analyzing scenarios where C-14 may migrate as gas. The main potential sources for gas formation are metals (in the waste or package material) and organic wastes (in particular in LLW and ILW). The transport modes and rates in the 'gas-scenarios' depend on the total amounts of produced gas, the total gas production rates, the capacity of the disposal system to store and remove gas, and the response of the host rock to large, pressurized volumes of gas. These processes will be analyzed in OPERA in several scenarios.

[2] Features of the geosphere, including any aquifer systems surrounding the Boom Clay host rock are presently studied in Work Package 6 of the OPERA programme

5 Key issues and priorities

- A main safety issue connected to C-14 is its potential of transport in the gas phase, which may be enhanced compared to transport in the liquid phase.

- A priority research issue is to investigate the 'gas balance' in the repository , i.e. determining the gas production rates, including C-14 containing gases, and the gas storage and removal capacity of the repository system and the surrounding host rock, by physical and/or chemical processes.

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SURAO contribution to D6.1

Antonín Vokál

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1 Czech Geological Disposal Concept

The Czech Republic has no suitable salt domes or large clay soil areas so that only crystalline rocks can be considered for the construction of a deep geological repository. The Czech DGR concept assumes that waste packages containing spent fuel assemblies will be enclosed in steel-based canisters placed in vertical or horizontal boreholes at a depth of ~ 500m below the surface. The space between the canisters and the host crystalline rock will be backfilled with compacted bentonite which will make up the final engineered barrier (Fig 1). The reference canister design contained in the Czech DGR concept is composed of two layers, an outer layer of carbon steel which will corrode very slowly under anaerobic conditions and a second inner layer of stainless steel which will corrode at an almost negligible general corrosion rate and exhibit a low tendency to local corrosion under anaerobic conditions. The steel-based canisters considered in the Czech concept, as opposed to the copper-based canisters considered in the Swedish and Finnish concepts, have a number of advantages and disadvantages. The main disadvantage is that steel, unlike copper, is not thermodynamically stable in the reducing environment of geological repositories located 500 meters below the surface. The main advantages are that manufacturers have substantial industrial experience of the production of steel-based canisters thus leading to a considerably lower probability of initial defects in the canisters caused by human error during the production process, and that the price of steel canisters is much lower than that of copper canisters.

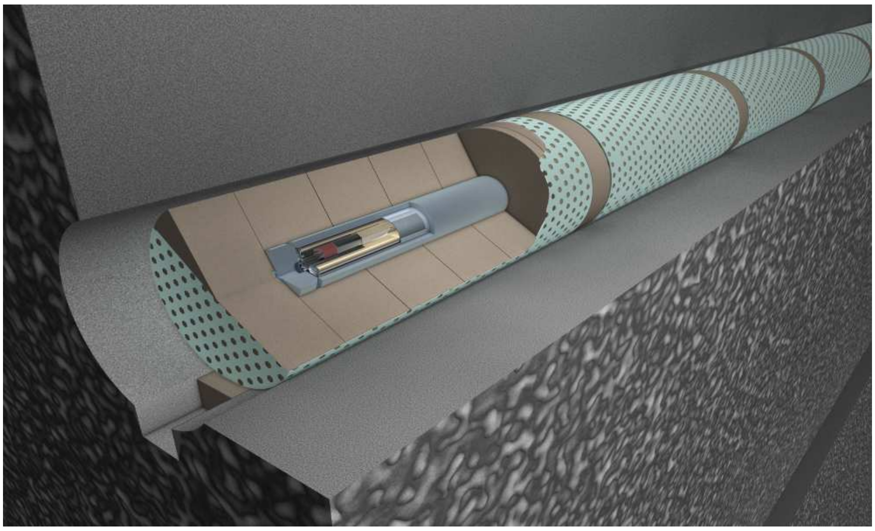

Figure 1: Visualisation of the Czech DGR concept using horizontal boreholes

Intermediate-level waste (ILW) with long-lived radionuclides, such as reactor core parts, that cannot be disposed of in the near-surface repositories available in the Czech Republic (Dukovany and Richard repositories) will be disposed of in a future deep geological repository. The ILW repository will be located at the same site as that for spent fuel assemblies; however the exact location has not yet been selected. The main requirement is that the repository areas set aside for spent fuel assemblies and ILW should not be able to affect one another. A scheme for both repositories is provided in Fig.2. The ILW will be emplaced in concrete canisters in specially excavated chambers that will be filled with bentonite based backfill (Fig. 3).

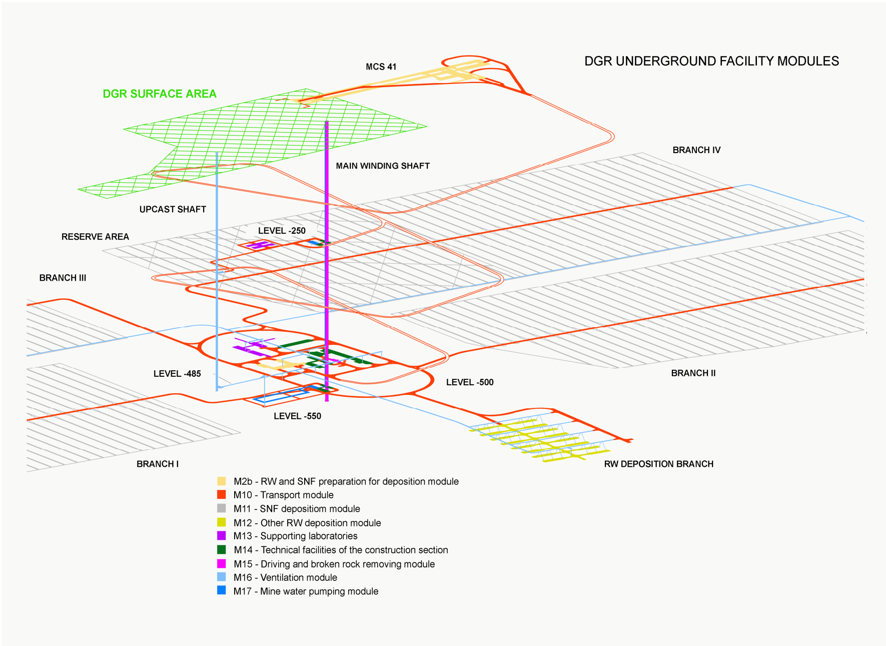

Figure 2: Scheme of the future Czech DGR for spent fuel assemblies and ILW

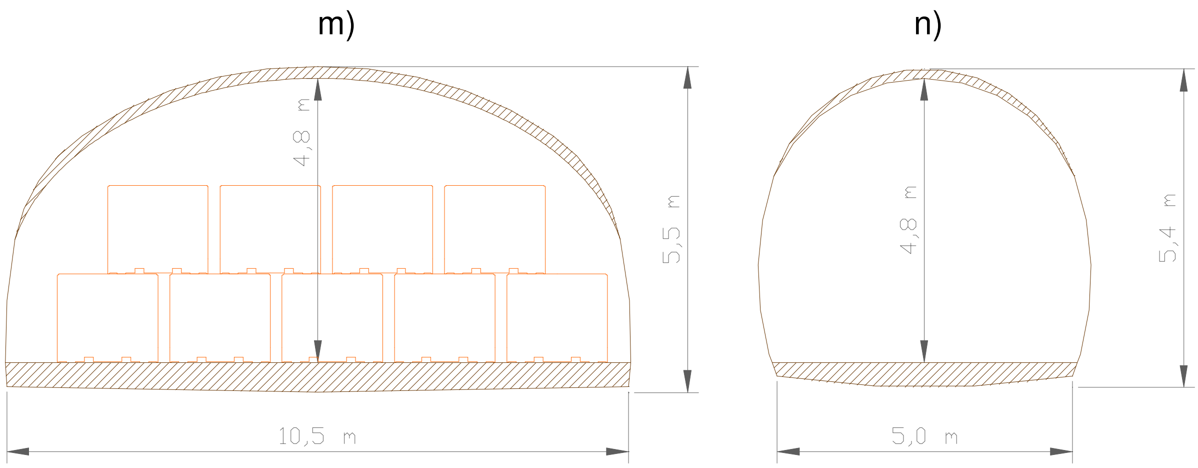

Figure 3: ILW disposal chambers

2 C-14 Inventory

DGR in the Czech Republic will involve all radioactive waste which is not acceptable to near-surface repositories, primarily spent fuel assemblies from two nuclear power plants located at Dukovany and Temelin. These nuclear power plants (NPPs) operate WWER 440 and WWER 1000 reactors, respectively. Currently, SF assemblies are stored in dry storage facilities located in the area of both NPPs in CASTOR-440/84 type approved casks or in pools at reactor sites. More than 7000 SFs from WWER 440 reactors and 300 SFs from WWER 1000 reactors are stored in this way. More than 13000 SFs are expected to be spent by 2045 at Dukovany reactors (under the assumption of extending lifetime of this NPP from 40 to 60 years) and 4600 SFs by 2042 at Temelin reactors. There is also considered in the Czech Republic to build new nuclear reactors at the sites of current NPPs to be in operation in time approaching the lifetime of current reactors.

The content of C-14 in spent fuel assemblies has been for preliminary safety cases estimated already in 1997 [Burian et al, 1997]. The inventory was calculated using ORIGEN code assuming average burn-up 42GWd/t_{hm} with enrichment of 3.6% for WWER 440 reactors and 50GWd/t_{hm} with average enrichment of 3.6% for WWER 1000. It was assumed [Burian et al, 1997] that potential nitrogen impurity in UO_2 matrix is 200ppm and in spent fuel structure materials 30ppm. The new C-14 inventory calculations were conducted in 2009 [Krupar, P., 2009] for SFs of new type TVSA-T (burn-up 60GWd/t_{hm}, enrichment 4.92%) intended for WWER 1000 reactors. The results are given in the Table 1.

Other sources of C-14 will come from decommissioning of internal, metal parts of reactors [Krupar, P., 2009] and ion-exchange resins (The reference option is, however, to dispose of ion-exchange resins in the near-surface repository at Dukovany). The expected inventory of C-14 of these waste streams based on measurements is given in the Table 1.

Table 1: Inventory of C-14

Waste type	Number of waste units	C-14 activity $[GBq/t_{hm}]$	Uncertainty
SF from WWER 440 reactors	20 250 SF assemblies	187	High
SF from WWER 1000 reactor	4600 SF assemblies	235	High
SF from WWER 1000 (new type)	8100 SF assemblies	166	High
Decommissioning waste from WWER 440 reactors	1996 waste packages	77	High
Decommissioning waste from WWER 1000 reactors	250 waste packages	148	High
Ion-exchange resins	4200 waste packages	0.13	High

3 Safety concept

The Czech safety concept is based on the KBS 3 concept developed in Sweden. The Swedish concept is based primarily on the role of the thermodynamic stability of copper overpacks in the reducing conditions of granite host rock 500m below the surface as well as on the almost totally impermeable compacted bentonite surrounding the canisters that protect them from corrosion. It is supposed that the corrosion rate of copper under repository conditions will be so slow that canisters may fail only after a period of 10^6 years; under certain very unlikely conditions connected with the erosion of the bentonite surrounding the canisters, it has been estimated that corrosion could lead to the penetration of a limited number of copper canisters within a safety assessment period of 10^6 years. The disadvantages of copper canisters consist of: difficulties concerning the welding of the lids which lead to the requirement for advanced technologies both for welding itself and welding control, and the considerably higher price of copper canisters compared to that of steel-based canisters. Hence the Czech safety concept opted for cheaper steel-based canisters instead of those based on copper. It is well known that steel is not thermodynamically stable under reducing conditions, but it is known that under anaerobic conditions its corrosion rate can be very slow if microbial corrosion is excluded (the same condition also applies for copper canisters) which is possible due to the compacted bentonite preventing the growth of microbes. In contrast to Sweden, the Czech Republic enjoys geologically more favourable conditions with respect to the corrosion of canisters since in general the groundwater contains lower concentrations of chlorides which make up the most aggressive agent in terms of the corrosion of stainless steel by groundwater at sites near to the sea. But it will be, however, necessary to prove that the lifetime of steel canisters will be sufficient for ensuring safety even in the case of disturbances, such as bentonite erosion, by the comprehensive research programme.

The number of canisters which will fail in one year is very important since, following the failure of a canister, the most mobile radionuclides, which will most probably be located in the gap between the spent fuel cladding and the uranium matrix, could be released immediately thus, in crystalline rocks, allowing rapid migration via fractures into the environment. However, the amount of such mobile radionuclides is small. According to the preliminary safety case prepared recently in the Czech Republic in connection with the updating of the Czech DGR reference design concept, the following radionuclides will be most critical in terms of repository long-term safety: ^{129}I, ^{36}Cl, ^{79}Se and ^{14}C. In the later stages of repository development, following the failure of all the canisters, ^{226}Ra and its daughter products (^{210}Po and ^{210}Pb) could also present a risk for people living in areas above the repository.

4 Treatment of C-14 in safety assessment

In the Czech preliminary safety case conducted in 2010 [Vokal et al, 2010], the total inventory of C-14 was estimated to be 1.65×10^{15} Bq [Burian et al, 1997], [Krupar, P., 2009]. The instant release fraction of C-14 was estimated at 10% [Johnson L et al., 2005]. The leaching rate from spent fuel matrix waste was estimated at 1×10^{-8} yr^{-1} based on literature data [SKB, 2006] and from metals 1×10^{-2} yr^{-1}. Conservatively, no solubility limit was considered for carbon in the repository. Diffusion coefficients for bentonite and granite were estimated at $1.2 \times 10^{-10} m^2 s^{-1}$ and $2.4 \times 10^{-14} m^2 s^{-1}$ respectively and sorption coefficients for bentonite, granite and soil on the surface $0.2 m^3 kg^{-1}$, 0.1and $0.99 m^3 kg^{-1}$, respectively [Vokal et al, 2010]. A transport of C-14 in form of a gas phase has not been considered.

The results of activity release from the total source term of SF and ILW to the geosphere considering estimated minimal lifetime of canisters 50 000 years and mean lifetime 110 000 years, are provided in Fig. 4. An estimate of effective doses considering following conversion factors in the biospheric calculations: for ingestion 5.8 x 10^{-10} $SvBq^{-1}$, for inhalation 5.8 x 10^{-9} $SvBq^{-1}$ [Resrad, 2007], [IAEA, 2010] are shown in Fig. 5. All calculations have been conducted using GoldSim module RT [GoldSim, 2010].

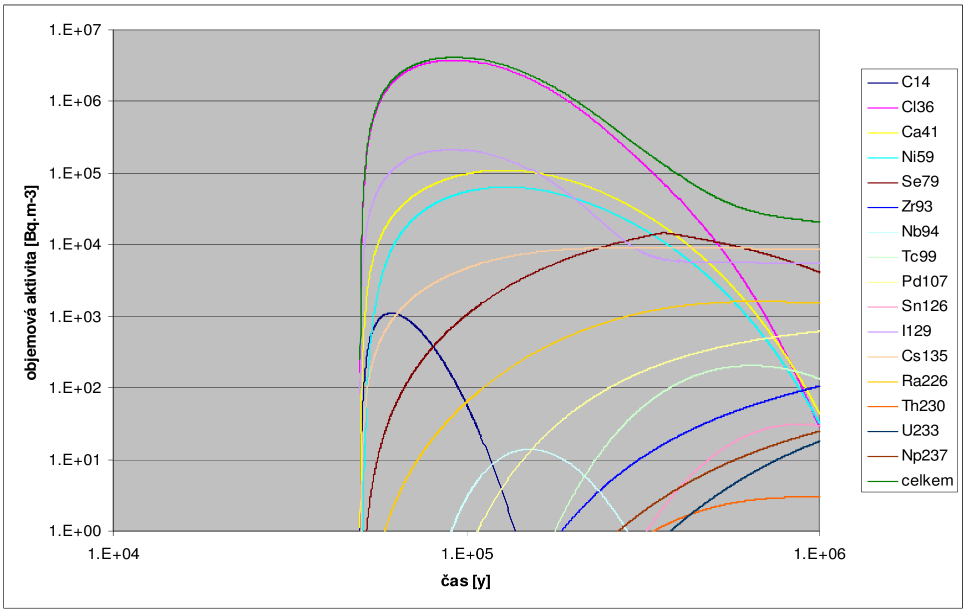

Figure 4: Release of radionuclides form source term to geosphere

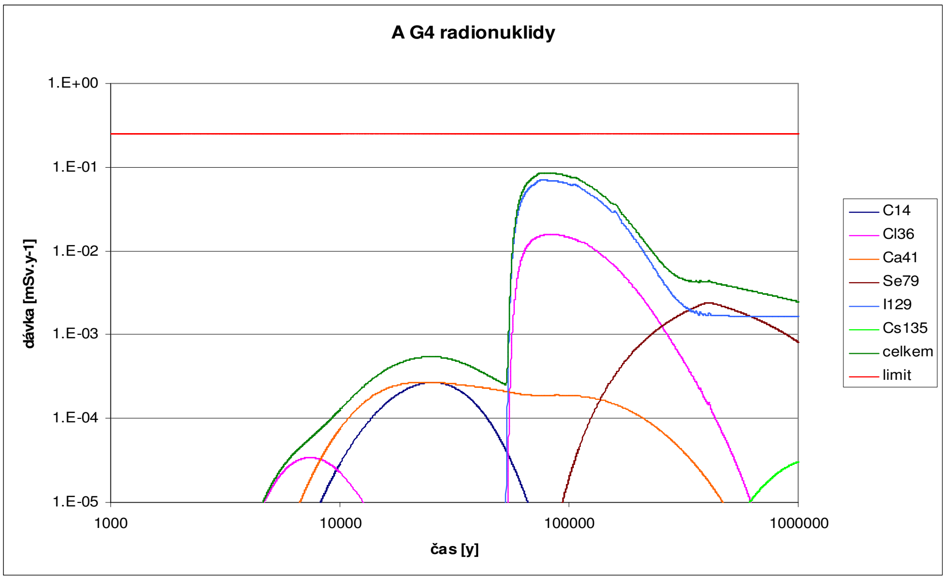

Figure 5: Estimate of effective doses with contribution of C-14

It can be seen that the contribution of C-14 to the effective dose is very small, but not negligible. C-14 could be released from the DGR in the first batch of radionuclides mainly from the waste from decommissioning reactors internals.

5 Priorities & Safety issues

Preliminary assessments were based on very conservative and uncertain data. The main priorities are therefore to reduce uncertainties and conduct sensitivity and uncertainty analysis in order to identify the most critical parameters for C-14.

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CArbon-14 Source Term

CAST

SKB contribution to D6.1

Klas Källström

Date of issue of this report: 18/05/2015

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1 Geological Repository Concept

The final repository for short-lived radioactive waste (SFR) located in Forsmark, Sweden is currently being used for the final disposal of low- and intermediate-level operational waste from Swedish nuclear facilities. SKB plans to extend the repository to host waste from the decommissioning of the nuclear power plants. The existing facility (SFR 1) comprises of five waste vaults with a waste disposal capacity of approximately 60,000 m^3, see Figure 1-1. The waste vaults are located about 60 meters below the seabed in Öregrundsgrepen in granitic rock. The planned extension (SFR 3) will connect directly to SFR 1 and the six new waste vaults. An overview of the repository after planned sealing and closure measures described is shown in Figure 1-2. It is required by the Swedish regulator that the safety analysis for SFR 1 must be updated every tenth year. In 2008 SKB handed in a updated Safety analysis of SFR 1 (SAR-08). In that analysis C-14 was the dose dominating nuclide, see Figure 1-3. The total dose from the main scenario was 12 µSv just below the requierments of 14 µSv to meet the risk criteria of 10^{-6}. In later years a project (PSU) has been ongoing in order to extend SFR 1 with six new rock caverns (SFR 3) to be able to take care of decommissioning waste from the Swedish reactors. For the planned repository for spent nuclear fuel there will be a total of $5.10 \cdot 10^{14}$ Bq of C-14 not further divided into inorganic or organic fractions. For the spent nuclear fuel safety assessments C-14 release is divided into an instant release factor (IRF) and a corrosion release factor (CRF). For the future repository of long lived low- and intermediate level waste (SFL) the C-14 content is estimated to be about $4.6 \cdot 10^{14}$ Bq where the major part $4.0 \cdot 10^{14}$ comes from induction of either carbon or nitrogen impurities in core components.

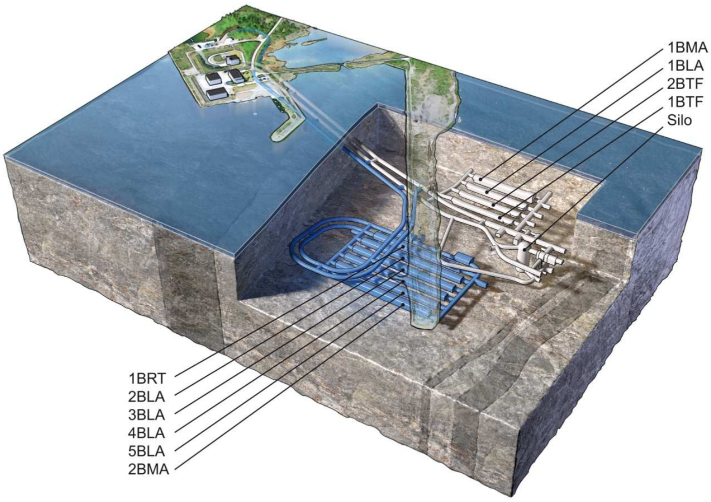


Figure 1-1. Schematic illustration of SFR. The grey part is the existing repository (SFR 1) and the blue part is the planned extension (SFR 3). (The waste vaults in the figure are Silo for intermediate-level waste, 1-2BMA vaults for intermediate-level waste. 1-2BTF vaults for dewatered spent ion exchange resins and incineration ashes.1-5BLA vaults for low-level waste and BRT vault for reactor pressure vessels).

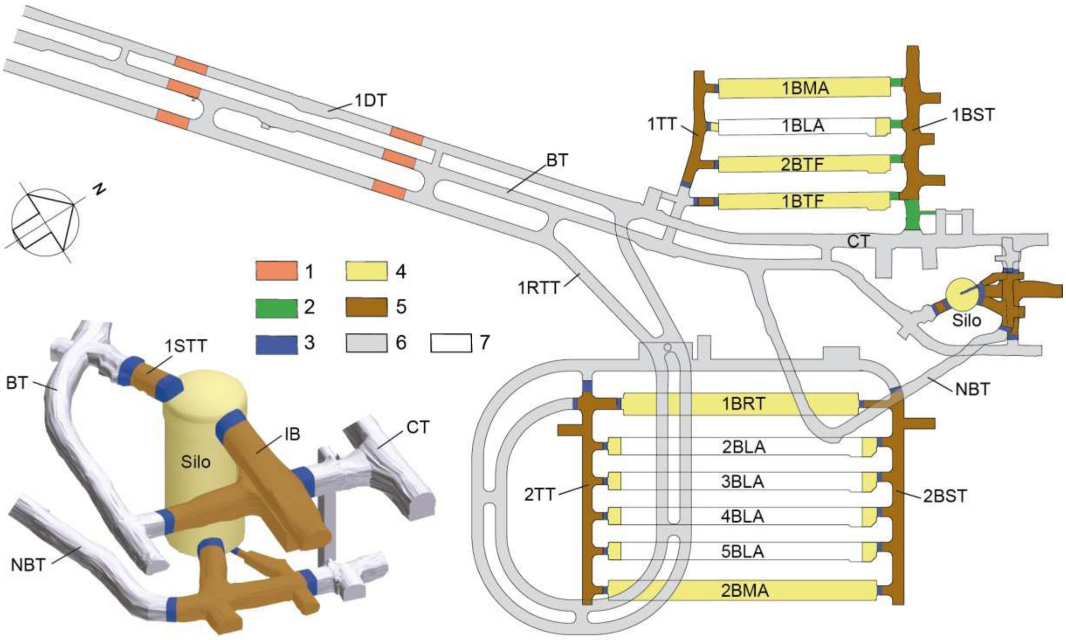


Figure 1-2. Overview of SFR after closure with detailed view of silo. Key to colors. 1 Tunnel plugs, 2. Transition material, 3. Mechanical plugs of concrete, 4. Backfill material made of MacAdam, 5. Low hydraulic conducting section made of bentonite, Tunnel backfill material of MacAdam, 7. Non backfilled waste vaults

Every waste type that is to be disposed in SFR must have an approved Waste type description that describes the whole handling sequence from production to final disposal of the waste. The C-14 waste is mainly deposited in the Silo, BMA and BTF caverns with the majority of the C-14 ending up in the BMA cavern. All these repository parts have concrete structures that serve as technical barriers for the long term safety. They serve both as sorption barriers, for the C-14 carbonate and as limiting the water flow through the waste. In addition the Silo has a bentonite barrier that serves both as a sorption and diffusion barrier. The actual resins that contain the C-14 are either embedded in cement or bitumen in the Silo and/or the BMA. For the BTF caverns the ion exchange resins are dewatered and placed in concrete containers. The actual cement waste form does provide an extra sorption barrier.

Grouting will be installed between the waste containers. This grouting will provide extra resistance to water flow through the waste as well as extra sorption capacity.

2 C-14 Inventory

SKB is responsible for estimating the C-14 content in SFR. For the activity originating from the spent ion exchange-resins (SIERs) this is done on a yearly basis by gathering information about the thermal energy produced every year for each reactor and multiplying this by the produced C-14 in the reactor. To correct for the actual activity that is deposited of in SFR a resin uptake factor is introduced. The uptake factor applied depends on from which reactor the resin originates from. The uptake of C-14 on the ion exchange resins from Swedish BWR reactors varies between 0.49 % to 3.8 % of the total produced C-14. For the PWRs the uptake is 3.6 % of the total produced amount. It is this factor that has been investigated during the last decade in the Swedish sampling and analyzing program. The total activity is the divided into inorganic and organic C-14 by multiplying with the fraction of organic C-14 as measured within the Swedish C-14 program. In addition to the SIERs C-14 is also present in reactor internals and decommissioning waste, biological shielding). The activity originating from these waste streams are calculated using the geometry of the parts and the neutron flux (modelled) it has received during the operational life time.

The total C-14 inventory in SFR is estimated to be about $6 \cdot 10^{12}$ Bq at the closure. The majority of the inventory originates from the SIERs.

The inventory of C-14 as used in the Safety assessment for SFR is given in Table 1-1. The C-14 activity for the majority of the waste are considered as moderate (within one order of magnitude) whereas the uncertainty associated to ion-exchange resins are considerate to be low.

Table 1-1. Inventory of C-14 for the main waste strems

Waste type	Package number	C-14 activity (GBq/package)	Determination method	Uncertainty wrt C-14
Resins	≈6000	≈1	Measurements and calculations	Low [±] 20 %
Reactor pressure vessels (neutron induced steels)	9	0.9	Modelling	Low [±] 2 i.e double or half the amount
Biological shield (neutron induced concrete)	>1000	≈0.001	Modelling	Low [±] 2 i.e double or half the amount
Industry and research	34	9	Estimation	High >10 i.e. 10 times higer or one tenth of the best estimate.

For the SIERs the inventory of the organic C-14 fraction varies between the different reactors. The content from the PWR reactors is about 30 % whereas from the BWR reactors the content varies between 1-5 % depending on the water chemistry and the construction of the reactor, forward pumping, down stream processing of the SIERs before waste solidification. The induced C-14 (steel and concrete) is assumed to be present as carbides in the steel which reacts with water upon corrosion and forms volatile short chained aliphatic hydrocarbon. The speciation of the induced activity in the concrete biological shielding has not been further investigated and is currently treated as non sorbing species as a pessimistic assumption.

3 Safety concept

In contrast to a KBS-3 repository for spent nuclear fuel, where total isolation of the waste is the most important overall safety principle, safety in SFR is based on the limited quantity of activity in the waste and retardation of releases of radionuclides by the engineered barriers and the geosphere.

3.1.1.1 Near field

The limited quantity of activity that may be deposited in SFR is a prerequisite for the safety of the repository, and the quantity of activity in the different waste parts has therefore been selected as a performance indicator.

The different engineered barriers in the repository constitute obstacles to the release of radionuclides. Nuclides that are enclosed in moulds and other closed packages must first diffuse out of the packages. In the repository parts that are designed as closed volumes, such as the silo, transport is further hindered by the low hydraulic conductivity of the outer walls. A property that all engineered barriers have in common is that they contribute to safety by means of the safety function “limited advective transport”, and they have a common safety performance indicator, hydraulic conductivity see SKB 2008 chapter 5 for a detailed discussion of the safety functions and safety indicators.

Sorption of radionuclides will occur in all repository parts where C-14 are deposited and in the surrounding rock. The greatest potential for sorption exists in the repository parts that contain cement. The safety function “good sorption” is therefore defined as sorption on cementitious material, which includes concrete walls, grout and waste packages. Many radionuclides sorb very strongly in bentonite, rock and aggregate. Credit is taken for this in the nuclide transport calculations, but it has not been defined as a safety function in SAR-08 because sorption on cement is assumed to dominate compared with sorption on other materials.

3.1.1.2 Geosphere

A low groundwater flow through the repository caverns is a prerequisite for its safe function. This is particularly true of BLA, which lacks other retardation functions.

The site of SFR was selected partly because of the low hydraulic gradient. The direction and magnitude of the gradient will change due to shoreline displacement, however. Today, when SFR is below the sea floor, the gradient is slightly upward-directed. After about 1,000–2,000 years the gradient is expected to increase and become more horizontal. In this later phase the gradient is also expected to be controlled more by the local, rather than the regional, topography.

3.1.1.3 Biosphere

A location beneath the Baltic Sea is considered to be favourable in the sense that no wells could be drilled down to the repository before the process of land uplift had reached the point where the repository is beneath land.

The absence of wells in the actual repository is therefore considered to be a safety function. The performance indicator is the location of the repository in relation to the shoreline.

4 Treatment of C-14 in safety assessment

4.1.1.1 The reference scenario

The engineered barrier system (EBS) contains large quantities of cementitious materials, whose properties evolve with time. The dissolved inorganic carbon in cement pore water is considered to be controlled by the solubility of calcite. However this is treated as a distribution coefficient (K_d) in the radionuclide transport calculations.

In the last safety assessment [SKB 2008] the assumed C-14 inventory on SIERs was taken from Magnusson et al. [2007] with the pessimistic assumption that 30 % of the total C-14 content was organic C-14. No other sources of C-14 than from the SIERs were considered. In Magnusson et al. [2007] the amount of C-14 based on a life-time of the nuclear power plants of 40 years is assumed to be 5.0 TBq. The annual production of C-14 is also calculated and added to correspond to 50 and 60 years of reactor life time. The total activity of C-14 was distributed within the different repository parts e.g. different rock caverns and the Silo according to the Co-60 distribution between the different repository parts. The long term doses due to release of radionuclides from the SFR 1 repository were dominated by C-14 especially organic C-14. Inorganic C-14 is attributed sorption (K_d between 0.2-0.4 m^3/kg cement). Organic C-14 is not retained upon sorbing species mainly due to lack of relevant experimental values and the assumed properties of low molecular weight organic acids that are highly soluble and poorly sorbing onto the cement phases.

Within the ongoing safety analysis (PSU) in the radionuclide transport calculations degradation of the engineered barrier system is modelled by increasing hydraulic conductivity. The degradation varies between each cavern due to differences in water inflow hence increased out transport of C-14 depends upon the water transport properties for each cavern. The Silo has the best engineered barrier system and will be diffusion controlled during the whole time period that is evaluated (100 000 years). For the repository parts that do not have bentonite as an barrier the concrete will degrade and the hydraulic conductivity will increase with time. Roughly after 20,000 years the hydraulic conductivity of the concrete will resemble the hydraulic conductivity of the backfilling in BMA. For the induced C-14 the release will follow the anaerobic corrosion rate under alkaline conditions (0.05 µm/year) until the pH has dropped below 10.5 where the corrosion rate is set to increase by a factor of about 50. For the rock cavern containing the BWR reactor pressure vessels this will happen roughly 20000 years after closure. Residual scenarios have been evaluated within the safety assessment. The one that affects C-14 migration is called no sorption in the near-field where the sorption term in the radionuclide transport calculations have been omitted. This is considered to be a highly pessimistic case and hence it is treated as a residual scenario in order to highlight the importance of the sorption as a safety indicator.

4.1.1.2 Biosphere model for C-14

The biosphere model previously used for C-14 is based on the specific activity approach, which is recommended by UNSCEAR and IAEA for dose calculations from releases of C-14 to the environment from nuclear installations. The most important assumption in these models is that the long-term behaviour of C-14 is modulated by the natural cycle for stable carbon (C-12) and that the isotopic equilibrium between C-14 and C-12 is achieved with a constant isotopic ratio (specific activity), i.e. the same specific activity (expressed in Bq/g C) will be observed in all parts of the biosphere. Furthermore, several realistic and conservative assumptions were made to obtain simple equations for calculating the specific activity of C-14. These assumptions are documented and explained in the background report to the C-14 model [Avila and Pröhl 2008].

The specific activity model for C-14 was used to calculate dose to humans in ecosystems than can receive releases from the geosphere during different periods: mire, forest, agricultural land, coastal area and lake. The specific activity model for C-14 was also used to calculate the dose from exposure as a consequence of use of a well for drinking water and for irrigation of vegetables. The exposure from C-14 in ecosystems downstream is always judged to be lower than the exposure from the ecosystem that receives the release from the geosphere. This is a direct consequence of the fact that isotope dilution is irreversible; in other words once C-14 has been mixed with a given amount of C-12 the concentration cannot increase again [Sheppard et al. 2006].

The following exposure pathways were included in the calculation of dose from C-14:

•Ingestion of contaminated food and water in both terrestrial and aquatic ecosystems,

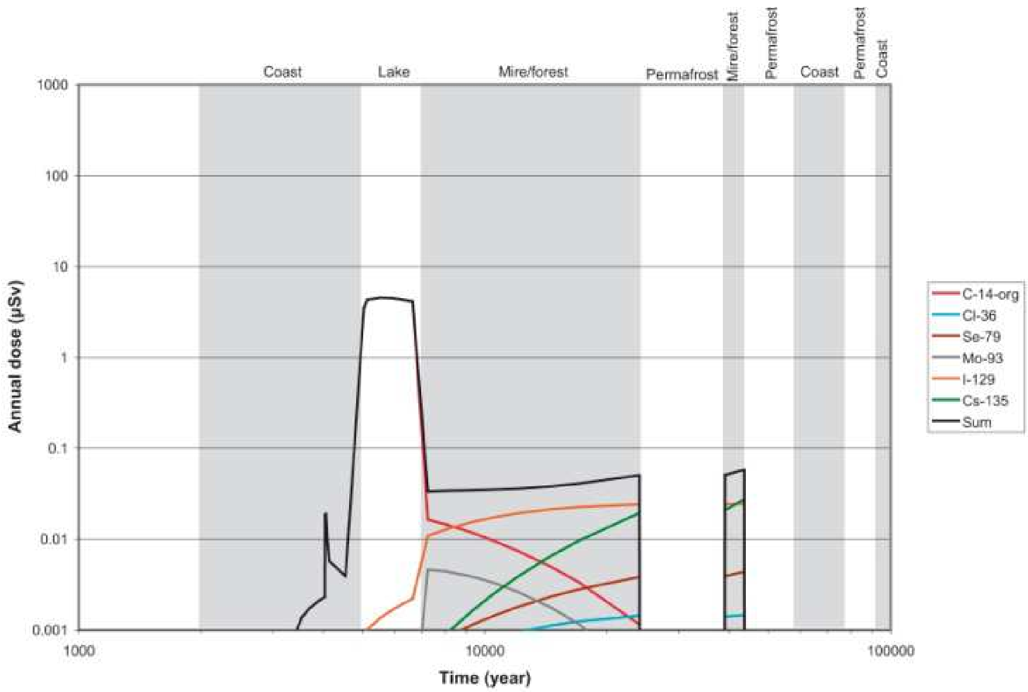
•Inhalation of contaminated air in terrestrial ecosystems.

Exposure by external irradiation has been neglected, as C-14 is a pure low-energy beta emitter.

4.1.1.3 Altered scenarios

The gas-driven advection case is based on the fact that gas generated in the repository due to corrosion and microbial processes gives an advective flow that drives out contaminated water to other parts of the system than in a situation with no gas.

Transport of radionuclides due to gas formation causing advective transport from the silo is not taken into account in the main scenario. In order to shed light on the importance of such transport, releases of radionuclides during a period of increased gas generation were calculated. The result was an early peak of the release of organic C-14 to the coastal area. The dose at the time of the release increased slightly, but the highest dose and the long-term dose were not affected. The case therefore did not lead to any difference in highest dose [Bergström et al. 2008].



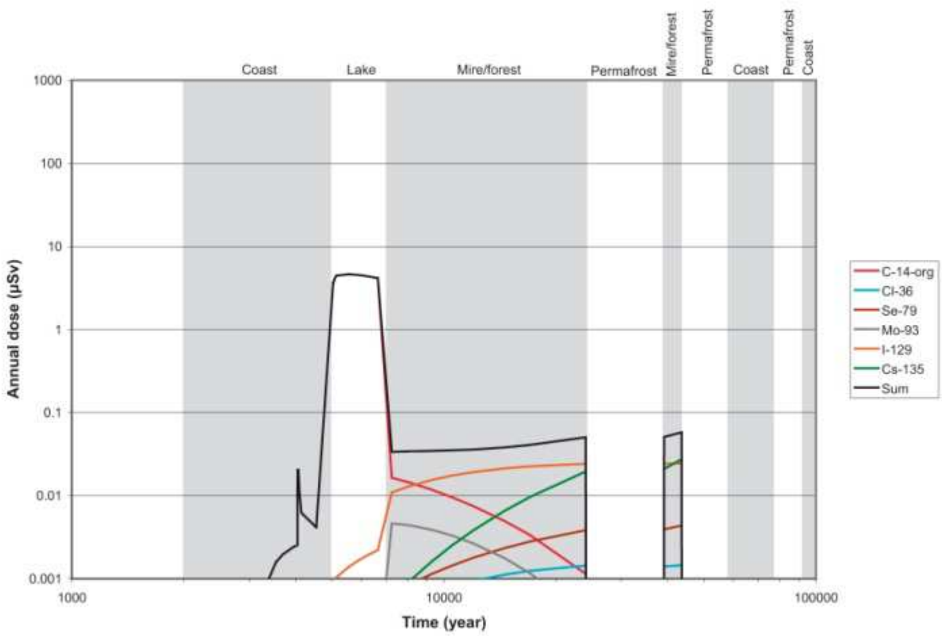


Figure 1-3. Calculated doses from the Silo in SAR-08 [SKB2008]. To the left doses from the main scenario. To the right doses from the gas driven advection scenario.

4.1.1.4 Sensitivity analysis

Calculations have been carried out with an elevated inventory, up to the total quantity of activity for which the repository is licensed. The activity of the individual radionuclides has thereby been scaled up by a factor of 7, causing the doses to increase by an equal amount, i.e. by a factor of 7, compared with the results for the main scenario's Weichselian, last glacial cycle, variant. Within the current safety analysis project the a case with higher inventory of C-14 due to the uncertainties of the inventory has been constructed.

5 Key issues and priorities

From the last safety assessment (SAR-08) it was concluded that C-14 is the main dose contributing radionuclide and that better understanding of the source term and the behavior in the biosphere has to be implemented for the upcoming safety analysis. Results from such investigations is implemented in the ongoing safety analysis project.

- The safety analysis is based on best estimates of C-14 inventory. SKB and the Swedish nuclear power plants need to better estimate the actual C-14 content, divided into inorganic and organic carbon that is actually deposited of in SFR.
- A better understanding of the organic content of the C-14 might lead to less pessimistic retardation factors (K_d) hence lowering the future doses.
- A better understanding of the behavior of C-14 in the biosphere should lead to less pessimistic assumptions.

Within the present safety analysis the best estimate of the C-14 content is used as a base case and is the foundation of all process understanding and the reference evolution. The inventory based on the upper limit in the C-14 content is considered a less probable scenario and it contributes to 5 % in the risk summation that is used to show compliance. The 5 % is a pessimistic value since it is based upon the assumption that all upper limit uncertainties will be realized.

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CArbon-14 Source Term

CAST

ENRESA contribution to D6.1

Cuñado Peralta Miguel

Date of issue of this report: 31/10/2015

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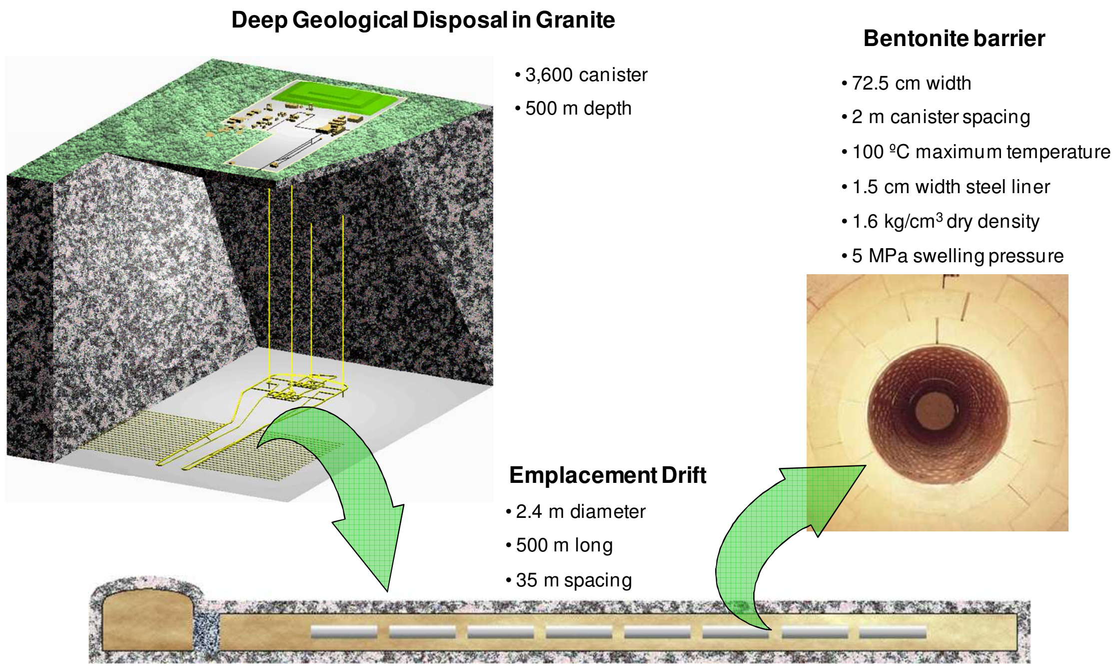
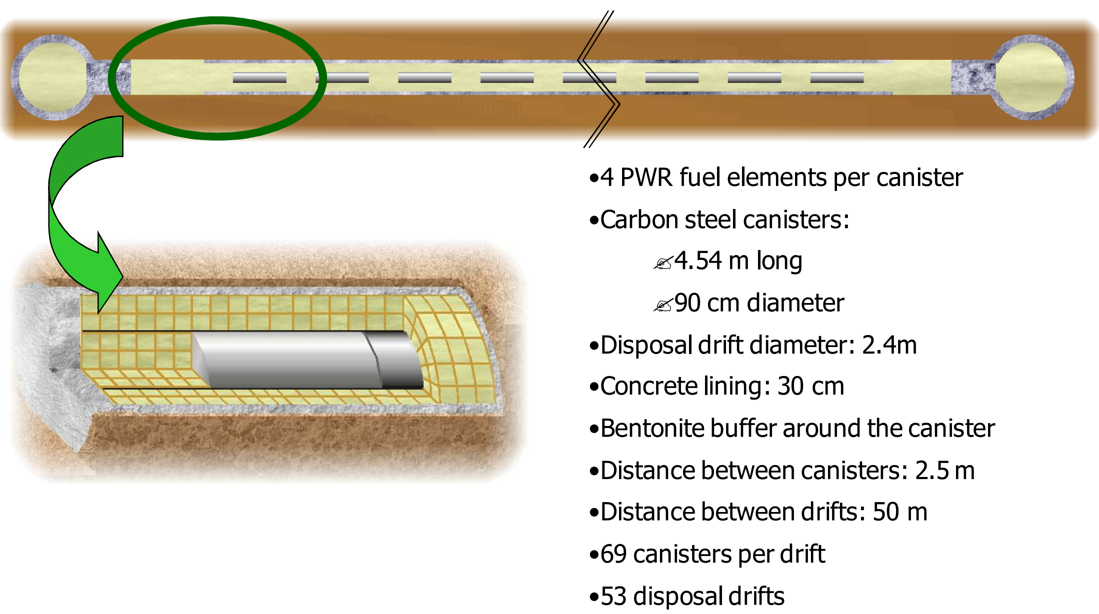
1 The Spanish Geological Disposal Concept



Repository in granite



Repository in clay

2 Inventory

The expected production of spent fuel during the operational lifetime of the Spanish NPPs is 20,000 fuel assemblies (FAs), 11,600 generated in Pressurized Water Reactors (PWR) and 8,400 generated in Boling Water Reactors (BWR).

To simplify the Safety Assessments in the first stages of the Deep Gelogical Disposal Project, a reference spent fuel assembly was defined. The reference fuel assembly is a 17x17 PWR AEF-ENUSA element, with 3.5% initial U-235 enrichment and 40 GWd/tU burnup. The initial uranium content of 461.41 kgU/FA. A total of 14,400 reference elements are roughly equivalent to the 20,000 FAs of different types expected to be produced by the Spanish NPPs.

The inventory of C-14 in the spent fuel was calculated using ORIGEN2.1. C-14 is created mainly by neutron activation of the nitrogen in the different components of the spent fuel. Nitrogen is not present in the specifications of the materials in the fuel, but small amounts will be present as impurities.

The table presents the nitrogen abundances considered in the ORIGEN2.1 source term calculations and the resulting C-14 activities per fuel assembly. Fission also produces some C-14, but the amount produced is much smaller, in the order of $2.41 \cdot 10^6$ Bq/FA.

Material	Initial nitrogen content	C-14 activity (Bq/FA)
Inconel-718	1300 ppm	$5.74 \cdot 10^{9}$
Stainless Steel SS-304	1300 ppm	$4.30 \cdot 10^{9}$
Zircaloy-4	80ppm	$8.14 \cdot 10^{9}$
UO_2 impurities	25 g/tU (upper limit)	$1.11 \cdot 10^{10}$
Total	--	$2.93 \cdot 10^{10}$

(N abundances taken from an example file of ORIGEN2.1 for a PWR fuel)

3 Safety concept

The long-term safety is provided by the joint function of the multibarrier system. In case of a granite host rock, the repository is located in an intact granite block without important conductive features, providing a considerable delay for the transport of any radionuclide dissolved in water. The bentonite buffer allows the transport of substances only by diffusion, a slow mechanism, and provides a strong sorption in pore spaces. Nevertheless, for the particular case of C-14, these two barriers are not so effective as for other nuclides since the carbon has a very low sorption coefficient in both materials. This is not the case for a repository hosted in a clay formation, where the clay layer has some hundreds of meters, causing a long travel time to the dissolved nuclides being transported through diffusion. This gives enough time for some nuclides like C-14 to decay. The canister keeps its contents for a minimum of 1000 years, thanks to a slow corrosion process. And finally, the fuel matrix provides contention through a very slow dissolution rate after the canister fails.

4 Treatment of C-14 in safety assessment

Source term

In the Safety Assessments for repositories in granite and clay the metallic components (that contain roughly 60% of the C-14 inventory of the FAs) are assumed to corrode relatively quickly, and C-14 is released congruently with metal corrosion.

In deterministic calculations, metals corrode at a constant rate during 1,000 years, while in probabilistic calculations a log-uniform distribution between 100 years and 10,000 years is used for the metallic components duration.

The C-14 produced in the UO_2 represents roughly 40% of the C-14 total inventory in a FA. A significant fraction is assumed to be present in the Instant Release Fraction (IRF) of the inventory that is released rapidly when water penetrates into the canister and contacts the spent fuel (no credit is given to the cladding as a barrier).

In deterministic calculations 6% of the C-14 inventory produced in the UO_2 is included in the IRF, and the remaining 94% is released congruently with UO_2 oxidation due to alpha radiolysis. In probabilistic calculations IRF(C-14) is represented by a uniform distribution between 3% and 12%.

Transport calculations for a repository in granite in the reference scenario

Only C-14 transport as a solute is considered. No calculations for C-14 transport as a gas have been performed.

Carbon is assumed to be transported through the bentonite as an anion. In the deterministic calculations there is no sorption of carbon on bentonite (Kd(C)=0), and in the probabilistic calculations Kd(C) is represented using a uniform distribution from 0 to 0.005 m^3/kg.

The relatively fast release of C-14 from the metallic parts of the FAs, and the fast transport through the bentonite (due to negligible carbon sorption) lead to significant releases of C-14 from the near field. The figure shows that C-14 is one of the radionuclides that produce the highest releases from the near field to the geosphere, especially in the first thousands of years.

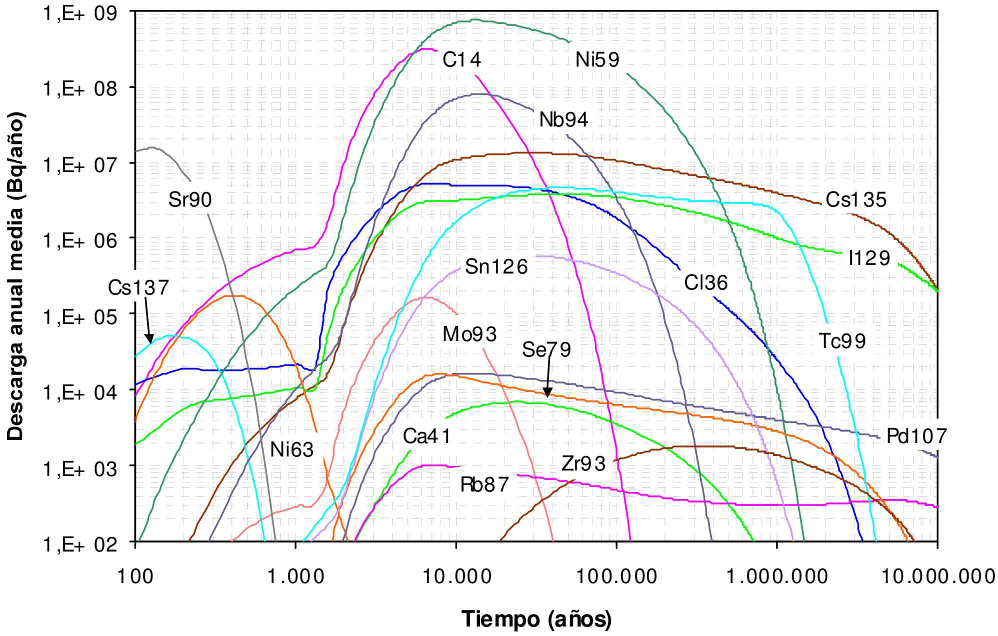


Figure - Mean Release Rates from the near field in the probabilistic calculations (reference scenario). Fission and activation products.Fig 6-17 de Enresa, 2000.

The values of Kd(C) in granite used in the calculations are always very small. In the deterministic calculations Kd(C)=0.001m^3/kg, and in the probabilistic calculations a log-triangular distribution is used, with minimum=0.0002m^3/kg, maximum=0.002m^3/kg and most likely value=0.001m^3/kg.

The small sorption on granite translate into a fast transport of C-14 through the geosphere. The figure shows that C-14 is one of the radionuclides that produce the highest releases from the geosphere, especially in the first thousands of years.

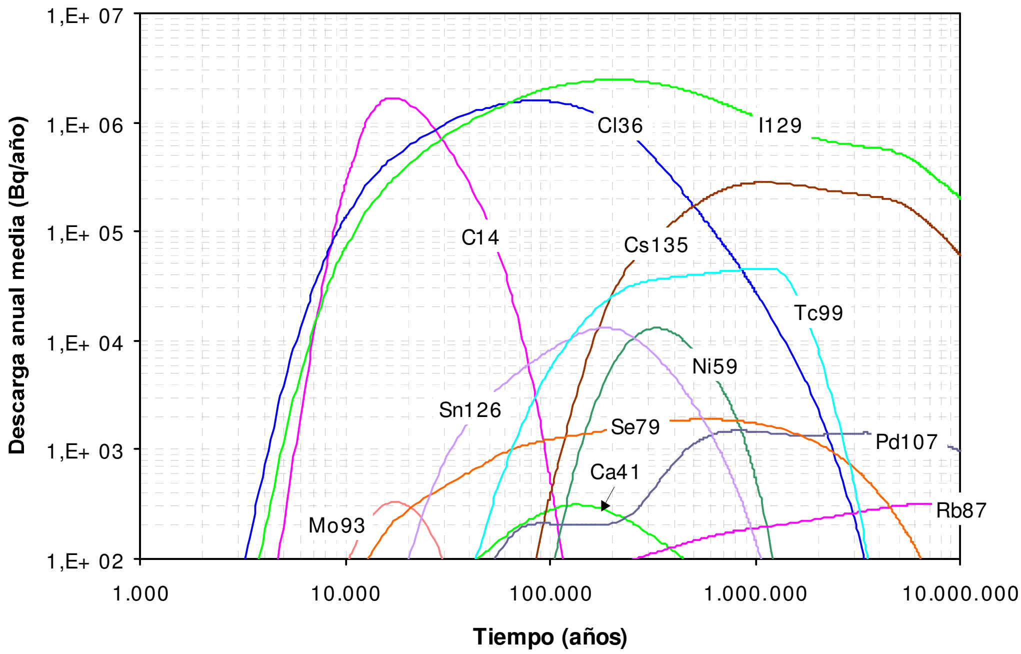
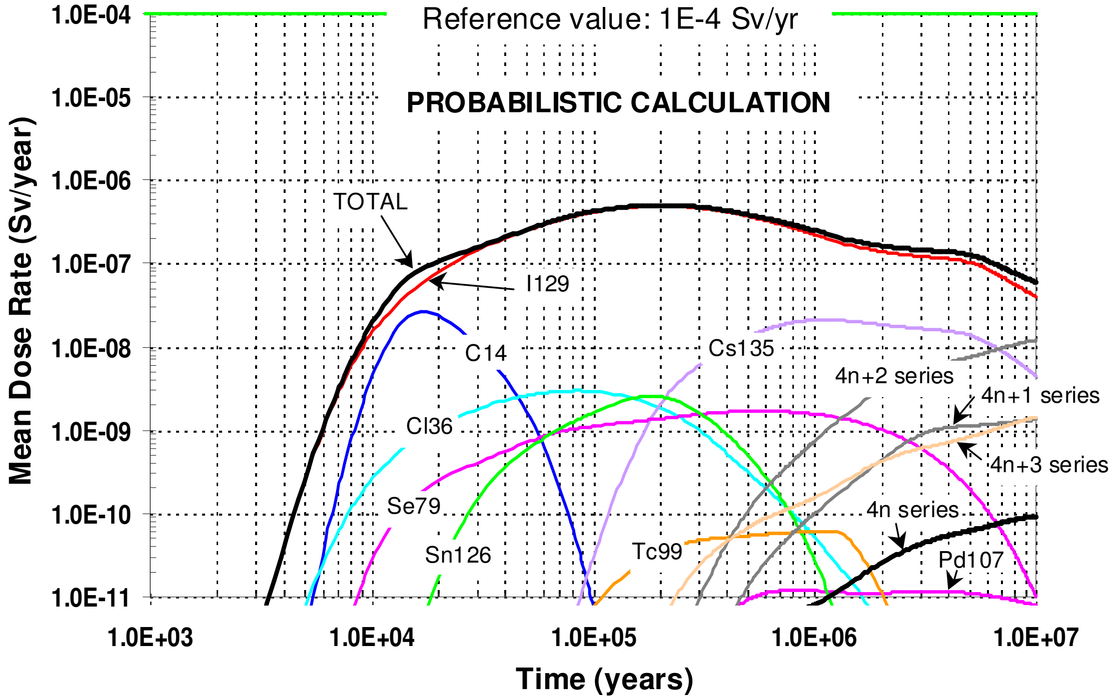


Figure - Mean Release Rates from the geosphere in the probabilistic calculations (reference scenario). Fission and activation products.Fig 6-19 de Enresa2000.

The figure shows that, in the reference scenario, C-14 is the second most influential radionuclide in terms of long term doses. Peak doses due to C-14 happen relatively early after 15,000 year approximately, but are several orders of magnitude below the reference value of 10^{-4} Sv/year.



Transport calculations for a repository in clay in the reference scenario

Only C-14 transport as a solute is considered. No calculations for C-14 transport as a gas have been performed.

Carbon is assumed to transport through the bentonite buffer, the clay and the marls as anion. The distribution coefficient in bentonite, Kd(C), is zero in the deterministic calculations while in the probabilistic calculations a uniform distribution between 0 and 0.005 m^3/kg is used. Carbon distribution coefficient in clay is 30% the Kd(C) in the bentonite and Kd(C) in the marls is only 10% of the distribution coefficient in bentonite.

The small sorption on the different argillaceous materials in the repository leads to a relatively fast diffusive transport through the different barriers. But solutes must cross 100m of clay and 110m of marls to reach the upper aquifer and 280m to reach the lower aquifer. As a consequence, transport times are very high even for non-retarded or weakly sorbed radionuclides (such as C-14), and hence C-14 decays nearly completely during its transport through the clay and marls. The activity of C-14 reaching the upper/lower aquifers is extremely small and produces negligible doses.

5 Priorities & Safety Issues

There are uncertainties in the initial amount of nitrogen in the different parts of a FA. Estimates of real concentrations of nitrogen (nor only upper bounds) would be useful.

Consequences of the possible transport of C-14 as a gas have not been analyzed.

Possible fast release of a fraction of the C-14 inventory in Zircaloy is reported in the literature. The importance of this potential IRF probably is small, since the Safety Assessments already considers an IRF for the C-14 created in the UO_2, and degradation of the metallic components is assumed to be quite rapid.

Organic/inorganic speciation of carbon in the repository is an important topic, due to the strong effect on C-14 transport through the barriers. Due to its relatively short half-life, inorganic speciation (and hence, significant sorption on the barrier materials) would lead to a great decrease in C-14 releases from the barriers.

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CArbon-14 Source Term

CAST

NAGRA contribution to D6.1

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List of Contents

1 Geological Disposal Concept

In Switzerland, the Nuclear Energy Law requires that all types of radioactive waste be disposed of in deep geological repositories. The Swiss Radioactive Waste Management Programme (NAGRA, 2008a) foresees two types of deep geological repositories: a high-level waste repository (HLW repository) for spent fuel (SF), vitrified high-level waste (HLW) and long-lived intermediate-level waste (ILW), and a repository for low- and intermediate-level waste (L/ILW repository). Note that ILW will be emplaced in separate tunnels within the HLW repository.

In the currently ongoing Swiss site selection process, Nagra proposed the Opalinus Clay as host rock for both repository types (NAGRA, 2014a). Opalinus Clay features a very low hydraulic conductivity, a fine, homogeneous pore structure and a self-sealing capacity, thus providing a strong barrier to radionuclide transport and a suitable environment for engineered barrier systems (NAGRA, 2002a).

2 ^{14}C Inventory

A summary of relevant waste types contributing to the ^{14}C inventory for the Swiss radioactive wastes can be found in Table 1.

Waste type	^{14}C-Inventory	N impurity Content (ppm)
Fuel assembly -PWR – BWR 35 to 65 GWd/tHM	55 – 105 GBq/tHM	Fuel: 25 Zircaloy: 70 – 80 Steel: 400-800
Fuel assembly -cladding (zircaloy)	0.05 – 0.08 GBq/kg	70 – 80
Fuel assembly -structural parts (steel)	0.2 – 0.4 GBq/kg	400 – 800
Reactor core internals	< 0.7 GBq/kg	90
Reactor pressure vessel	< 0.02 GBq/kg	50
Ion Exchange Resins	0.002 – 0.01 GBq/kg	
Vitrified HLW (CSD-V)	1 – 3 GBq/package	75
Compacted ILW metallic waste (CSD-C)	15 GBq/package	Zircaloy: 70 – 80 Steel: 400-800

Table 1. Inventory of ^{14}C in different Swiss radioactive waste types

A total of 2.7×10^{14} Bq of ^{14}C resulting from the inventories of SF, HLW and ILW is assumed to be disposed of in the HLW repository (NAGRA, 2014a,b). An additional 1.9×10^{14} Bq of ^{14}C resulting from ion exchange resins (discharged throughout the NPP operation lifetime), reactor core steel wastes, and steel components arising from the decommissioning of the NPPs (e.g. reactor internals, pressure vessel), are assumed to be disposed of in the L/ILW and partly in the ILW repository (NAGRA, 2014a,b).

The fuel assembly inventories are determined based on data from the waste producers (e.g. discharge burnups, initial enrichments, expected burnups for future discharges) and the use of state-of-the-art depletion/burnup calculations (ORIGEN-S/ARP). Additionally, international benchmarks are used for the validation of the nuclide inventory calculations for numerous measurable nuclides. Similarly, burnup calculations are used for the estimation of the ^{14}C content in the Zircaloy cladding and other structural parts of the assemblies. For already discharged fuel, detailed data are available and, therefore, small uncertainties of the corresponding inventories are expected. For future arising of spent fuel, calculations are based on expected discharge characteristics (e.g. fuel burnup). In general, bounding (maximum) inventory values are conservatively derived based on sensitivity analyses of characteristics such as burnup, initial enrichment and impurity content.

For reprocessing wastes (CSD-V and CSD-C), the inventories are based on guaranteed parameters and declarations from the waste producers. They are further supported by burnup calculations and conservative correlation factors.

The ^{14}C inventories for the ion exchange resins are based directly on measurements, whereas the inventories for activated steel components are based on neutron transport and activation calculations (MCNP/ORIGEN-S and other neutronics/activation codes developed by Nagra), assuming conservative impurity contents.

The speciation of ^{14}C in the waste is based upon the following assumptions (NAGRA, 2014c):

- In SF and HLW the ^{14}C is assumed to be organic (IRF 10 % for SF and 20 % for HLW, the rest is congruently released).

- ILW and L/ILW waste:
  - Homogeneous metallic waste from NPP and nuclear research facilities: congruent release (CR) in organic form (exception Al: instantaneous release (IR))
  - Homogeneous concrete waste from NPP and from the research facility CERN: organic IR
  - Homogeneous concrete waste from the interim storage facility Zwilag and surface facilities of a future geologic repository: inorganic IR
  - Concrete waste from nuclear research facilities (mixture of steel and concrete): 50 % organic IR, 50 % organic CR
  - Operational waste of the research facility PSI: organic IR

- Mixed (operational and decommissioning) waste from PWR 70 % inorganic and 30 % organic (both IR)
- Mixed (operational and decommissioning) waste from BWR 90 % inorganic and 10 % organic (both IR).

Further details can be found in NAGRA (2014c).

3 The safety concept

The safety and barrier concepts are described in several reports (NAGRA 2002a, 2008b, 2014a). The barrier concept describes the design of the different engineered and geologic barriers in the repository. The barrier systems of the HLW and the L/ILW repository are illustrated in Figures 1 and 2 and consist of:

- Waste matrices: SF pellets, HLW glass matrix, solidified/embedded matrix for ILW and L/ILW
- Container: steel container for SF and HLW, concrete container for ILW and L/ILW
- Backfilling of the emplacement tunnels: bentonite backfill for SF and HLW; cementitious mortar backfill for ILW and L/ILW
- Backfilling and sealing of the access tunnels and other structures
- Host rock and confining rocks
- Geological situation

The safety concept foresees that safety of the overall system is ensured by both engineered and geological barriers with multiple redundant safety functions. In general, Nagra aims at a safety concept where both engineered and geological barriers contribute significantly to the safety of the overall system.

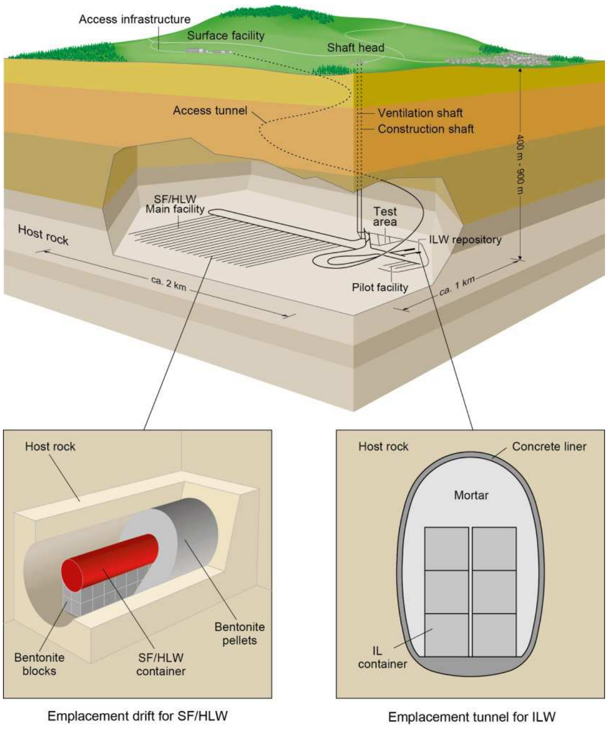

Figure 1. Conceptual outline of a deep geological repository for SF/HLW and ILW (NAGRA 2008b, modified)

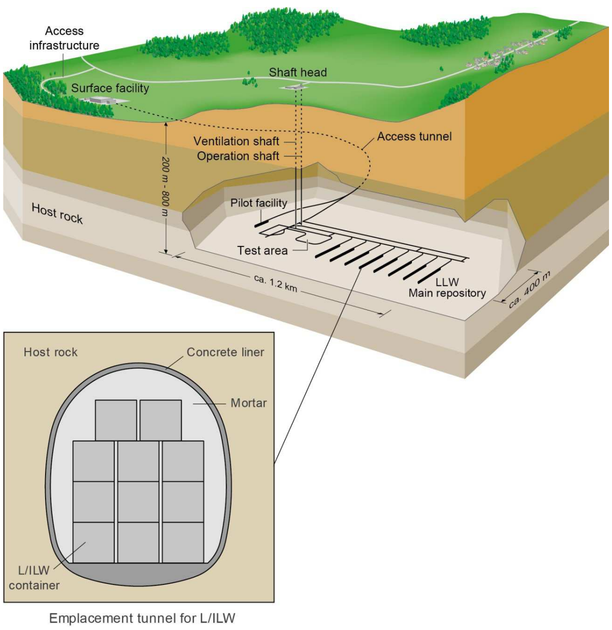

Figure 2. Conceptual outline of a deep geological repository for L/ ILW (NAGRA 2008b, modified)

The following mechanisms and processes contribute to the attenuated release of radionuclides from SF and HLW:

- the containment of radionuclides in the glass matrix of the HLW or in the spent fuel pellets
- the complete containment in the disposal containers for a minimum period of 10,000 years and radioactive decay
- low dissolution rates of the spent fuel and glass matrix
- low radionuclide solubility in the bentonite pore water
- sorption of radionuclides on the bentonite backfill
- low groundwater fluxes in the emplacement tunnels and host rock.

The following mechanisms and processes contribute to the attenuated release of radionuclides from ILW and L/ILW:

- low corrosion rates of metallic waste components
- low radionuclide solubility in the cementitious near field
- sorption of radionuclides in the cementitious near field
- low groundwater fluxes in the emplacement tunnels and host rock.

4 Treatment of ^{14}C in safety assessment

In a former safety case (in the framework of the Project 'Opalinus Clay'; Nagra, 2002a) and in Nagra's latest safety analysis (Nagra, 2014a) the expected evolution of the repositories in terms of radionuclide release is represented as 'Reference Case'. This involves a reference concept for radionuclide transport according to the geological model and reference parameter sets, both describing the most plausible situation and evolution. Further, as part of the so-called provisional safety analyses the regulator ENSI demands assessment cases with prescribed variations of parameters or consideration of different geologic situations ENSI (2010). In addition, 'Alternative Cases' and 'What if? Cases' are considered to explore relevant uncertainties and to evaluate the robustness of the system.

4.1 *Reference Case*

In the reference case, a containment period of 10,000 years is assumed for SF and HLW. Subsequent to this period, ^{14}C is assumed to be congruently released from the Zircaloy cladding within 30,000 years and from the SF matrix within 10 million years. However, instant release fractions of 10 % for the SF matrix and of 20 % for the cladding are additionally assumed. All ^{14}C released from SF is assumed to be in organic form (NAGRA, 2014a). In contrast, in a former safety case (NAGRA, 2002a), the ^{14}C from the cladding was assumed to be released in organic form, whereas the ^{14}C from the fuel matrix was considered to be inorganic.

The bentonite as backfill material for SF and HLW is fully water saturated at the time of canister breaching. The solubility of calcite limits the concentration of ^{14}C to 9×10^{-4} mol/kg$_{_H2O}$ (BERNER, 2014). Radionuclide transport is dominated by diffusion, whereas advective flow and gas transport are considered negligible.

For organic and inorganic ^{14}C in L/ILW and ILW, a containment period of 100 years is assumed. Subsequent to this period, ^{14}C is assumed to be congruently released from activated metals within 10,000 years or to be instantly released in case of other waste types (NAGRA, 2014a,c).

In the cementitious near field of ILW and L/ILW, linear sorption is assumed. The K_d-value of inorganic ^{14}C is 0.14 m^3/kg and that of organic ^{14}C (assuming small organic molecules) is 1×10^{-5} m^3/kg (WIELAND, 2014).

In the host rock Opalinus Clay, transport of radionuclides is dominated by diffusion. Transport-relevant parameters are considered to be homogeneous in space and time. No radionuclide transport is considered along tunnels, ramp or shaft or in the adjacent EDZ[1]. Transport of radionuclides in the host rock is retarded by linear sorption. The K_d of inorganic ^{14}C, describing ion exchange in the surface crystal layers of calcite, is 1×10^{-4} m^3/kg, whereas no sorption is considered for organic forms of ^{14}C (BAEYENS ET AL., 2014).

Transport and accumulation of radionuclides in the surface environment is analyzed by a dynamic compartment model. Doses are calculated for an adult individual assuming present day diet and entirely local food production.

A reference case calculation has been performed for each potential siting region in Nagra's latest safety assessment (NAGRA, 2014a). The results for a HLW repository in the siting region *Zurich Northeast* are reported here, in order to enable comparability with a former safety case (see below). A dose curve is presented in Figure 3, where ^{14}C is shown together with other dose contributing radionuclides. The maximum dose contribution of ^{14}C is 3.3×10^{-10} mSv/a, occurring after 6.3×10^4 years (NAGRA, 2014e).

In the reference case in NAGRA (2002a), the maximum dose due to ^{14}C was about 10^{-6} mSv/a. The main reason for this difference is that in NAGRA (2014a) the confining rocks of Opalinus Clay were taken into account, which was not the case in NAGRA (2002a). In both cases, the contribution of ^{14}C is mainly borne by organic species released from SF.

[1] These processes are analyzed in a separate report (NAGRA, 2014e).

4.2 Alternative Cases and 'What-if' Cases

ENSI prescribed a parameter variation procedure to study the impact of different variants of radionuclide release from the repository and transport through the barrier system (ENSI, 2010). These variations comprise:

- increased water flux through the repository (upper bounding values)
- unfavourable diffusion coefficients (upper bounding values for diffusion in a homogeneous porous rock and lower bounding values for matrix diffusion in a fracture network)
- increased solubility limits for radionuclides (upper bounding values)
- decreased sorption coefficients in the near field and in the host rock (lower bounding values)
- consideration of alternative climate variants (variation of biosphere dose conversion factors)
- increased dissolution rate of spent fuel elements
- SF / HLW canister life time reduced to 1,000 years.

Basically, these variations describe scenarios of unfavourable performance of the barrier system. It is important to note that all these assessment cases are strictly based upon the reference case, i.e. for the parameter to be varied the pessimistic data set is used whilst all other parameters are kept at their reference values.

Again, only the results from a HLW repository in *Zurich Northeast* are discussed. The analysis of the results of this parameter variation shows that only the case with unfavourable diffusion coefficients results in an increased dose contribution of ^{14}C with a maximum of 2.2×10^{-6} mSv/a, occurring after 3.2×10^{4} years as shown in Figure 4. Again, this dose contribution is mainly due to organic species of ^{14}C and the contribution from the IRF dominates the ^{14}C contribution. All other alternative calculation cases do not differ remarkably from the reference case in their ^{14}C dose contribution (NAGRA, 2014e).

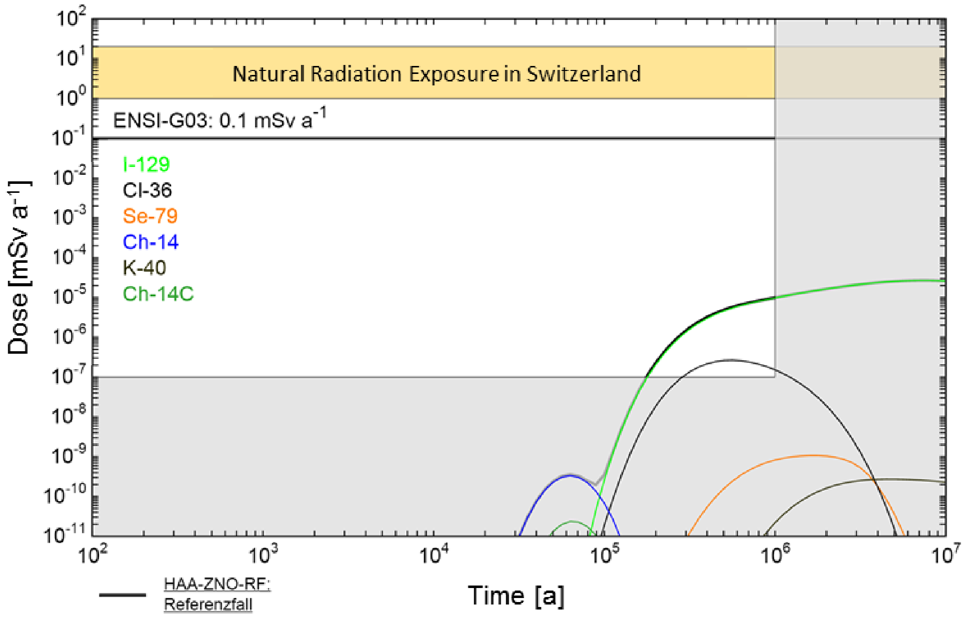

Figure 3. Dose curve calculated for an HLW repository in the siting region Zurich Northeast, Reference Case (NAGRA, 2014a,e). Ch-14 = organic ^{14}C IRF, Ch-14C = organic ^{14}C CR

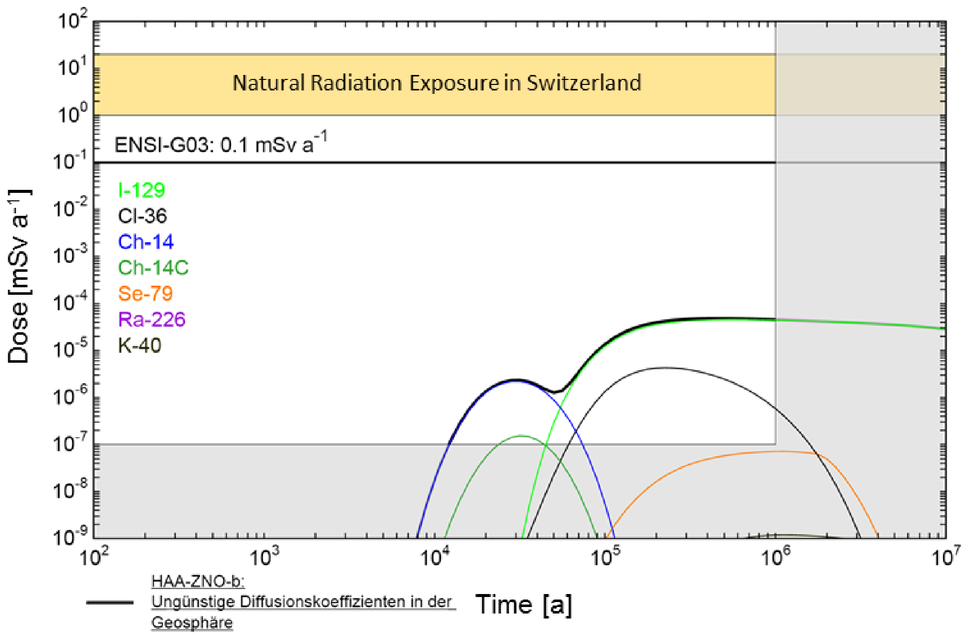

Figure 4. Dose curve calculated for an HLW repository in the siting region Zurich Northeast, Alternative Case 'Unfavourable Diffusion Coefficients' (NAGRA, 2014a,e). Ch-14 = organic ^{14}C IRF, Ch-14C = organic ^{14}C CR

Additionally, in a former safety case (Project Opalinus Clay; NAGRA, 2002a), the impact of a release of volatile radionuclide species along gas pathways was studied. $^{14}CH_4$ (methane gas as ^{14}C carrier) potentially released from structural parts of SF is assumed to be transported from the near field via gas pathways through the Opalinus Clay or access tunnels to an overlying argillaceous rock formation (Wedelsandstein). The calculation uses a semi-analytical approach (NAGRA, 2002b).

In one assessment case, gas pathways are assumed to exist in the Opalinus Clay but not in the access tunnels ('tight seals'). $^{14}CH_4$ migrates through these pathways to the Wedelsandstein formation where it accumulates and eventually diffuses as gas through the confining rock units.

Another conceptualization, which can be characterized as 'What-if' case, assumes gas pathways in the access tunnels ('leaky seals'). $^{14}CH_4$ is transported through the access tunnels into the Wedelsandstein formation.

The analyses in NAGRA (2002b) show that the doses for these cases are all below the regulatory dose guideline of 0.1 mSv/a. For the case 'tight seals', the resulting maximum dose due to ^{14}C in drinking water ranges between 6×10^{-7} and 3×10^{-6} mSv/a (SF) and between 1×10^{-7} and 4×10^{-7} mSv/a (ILW), whereas in the case of ‚leaky seals' the values are 4×10^{-5} mSv/a (SF) and 7×10^{-6} mSv/a (ILW).

5 Key issues and priorities

- In some assessment cases, namely with unfavourable diffusion coefficients or release of volatile ^{14}C species via gas pathways, ^{14}C can significantly contribute to the total dose rate from a geological repository.
- Open questions concern the remaining uncertainties in the inventory, the speciation of ^{14}C after release from the waste and the stability and potential retention of the different ^{14}C species in a cementitious or clayey environment. Further, our understanding and the modelling of volatile ^{14}C species through gas pathways will be reviewed and refined if necessary.
- It is expected that CAST can contribute to reduce the uncertainties in the speciation of ^{14}C.

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