



POSIVA 2012-11

Safety Case for the Disposal of Spent Nuclear Fuel at Olkiluoto — Complementary Considerations 2012

Posiva Oy

December 2012

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ISBN 978-951-652-192-6
ISSN 1239-3096



Posiva-raportti – Posiva Report

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Raportin tunnus – Report code

POSIVA 2012-11

Julkaisuaika – Date

December 2012

Tekijä(t) – Author(s) <i>Posiva Oy</i>	Toimeksiantaja(t) – Commissioned by <i>Posiva Oy</i>
Nimeke – Title SAFETY CASE FOR THE DISPOSAL OF SPENT NUCLEAR FUEL AT OLKILUOTO – COMPLEMENTARY CONSIDERATIONS 2012	
Tiivistelmä – Abstract <i>Complementary Considerations</i> sits within Posiva Oy’s Safety Case “TURVA-2012” report portfolio and has the objective of enhancing confidence in the outcomes of the safety assessment for a spent nuclear fuel repository to be constructed at Olkiluoto, Finland. The main emphasis in this report is on the evidence and understanding that can be gained from observations at the site, including its regional geological environment, and from natural and anthropogenic analogues for the repository, its components and the processes that affect safety. In particular, the report addresses diverse and less quantifiable types of evidence and arguments that are enclosed to enhance confidence in the outcome of the safety assessment. These complementary considerations have been described as evaluations, evidence and qualitative supporting arguments that lie outside the scope of the other reports of the quantitative safety assessment. The experience with natural analogues for the long-term durability of the materials involved and the extent of processes provides high confidence in our understanding of the disposal system and its evolution. For each engineered barrier and key process, there is increasing analogue evidence to support the conceptual models and parameters. Regarding the suitability of the Olkiluoto site to host a spent fuel repository, a number of factors have been identified that indicate the suitability of crystalline host rock in general, and that of the Olkiluoto site in particular. The report also provides radiation background information for the use of complementary indicators, which aid in putting the results of the safety analysis presented in <i>Assessment of Radionuclide Release Scenarios for the Repository System</i> and <i>Biosphere Assessment</i> in a broader perspective to show that the radiation originating from a spent nuclear fuel repository remains in most cases much below natural background radiation or that caused by non-nuclear industries.	
Avainsanat - Keywords Spent nuclear fuel, repository, KBS-3 disposal method, Olkiluoto, complementary indicators, natural analogues, anthropogenic analogues, safety case.	
ISBN ISBN 978-951-652-192-6	ISSN ISSN 1239-3096
Sivumäärä – Number of pages 262	Kieli – Language English



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Raportin tunnus – Report code

POSIVA 2012-11

Julkaisuaika – Date

Joulukuu 2012

Tekijä(t) – Author(s) Posiva Oy	Toimeksiantaja(t) – Commissioned by Posiva Oy
Nimeke – Title TURVALLISUUSPERUSTELU KÄYTETYN YDINPOLTTOAINEEN LOPPU-SIJOITUKSELLE OLKILUODOSSA – TÄYDENTÄVÄT TARKASTELUT 2012	
Tiivistelmä – Abstract Tämä raportti on osa Posiva Oy:n TURVA-2012-turvallisuusperustelun raporttisalkkua ja sen tavoitteena on esittää täydentäviä tarkasteluja, jotka tukevat Olkiluotoon rakennettavan käytetyn ydinpolttoaineen loppusijoituslaitoksen turvallisuusarvion tuloksia. Raportin päätavoite on tuoda esiin paikan soveltuvuuteen liittyviä todisteita ja ymmärtämystä sen laajemmasta alueellisesta geologisesta ympäristöstä ja kehityshistoriasta, sekä tietämystä luonnon- ja antropogeenisistä analogioista liittyen loppusijoitussysteemiin, sen komponentteihin sekä näiden pitkäaikaiskäyttämiseen. Erityisesti tässä raportissa käsitellään sellaisia pitkäaikais-turvallisuusarviota tukevia todisteita ja argumentteja joita ei voida suoraan kvantitatiivisesti arvioida, mutta joilla on merkitystä ilmiöiden ymmärtämisessä ja erilaisten ratkaisujen perusteluissa. Tällaisia voivat olla sellaiset kvalitatiiviset täydentävät arviot, todisteet ja argumentit, joita ei ole esitetty muualla Posivan kvantitatiivisessa turvallisuusarviodokumentaatiossa. Luonnonanalogioista saatu kokemus ja tieto liittyen eri loppusijoitussysteemeissä käytettävien materiaalien ominaisuuksiin ja niihin liittyviin prosesseihin tuo lisävarmuutta loppusijoitusjärjestelmän ja sen tulevaisuuden kehityskulkujen tuntemukseen. Jokaista teknistä vapautumisestettä ja keskeistä prosessia kohden on olemassa kasvava määrä analogiatodisteita tukemaan malleja ja parametreja. Olkiluodon soveltuvuuteen käytetyn ydinpolttoaineen loppusijoituslaitoksen sijoituspaikaksi liittyen on tunnistettu lukuisia seikkoja, jotka osoittavat kiteisen kallion soveltuvuutta yleisesti ja Olkiluodon soveltuvuutta erityisesti käytetyn ydinpolttoaineen loppusijoitukseen. Raportissa annetaan myös säteilyyn liittyvää taustatietoa, jonka avulla täydentävien turvallisuusindikaattoreiden analyysituloksia raporteissa <i>Assessment of Radionuclide Release Scenarios for the Repository System</i> ja <i>Biosphere Assessment</i> voidaan laajemmin vertailla esim. luonnon taustasäteilyyn ja muun kuin ydinvoimateollisuuden päästöihin.	
Avainsanat - Keywords Käytetty ydinpolttoaine, loppusijoitustila, KBS-3-ratkaisu, Olkiluoto, täydentävät indikaattorit, luonnonanalogiat, antropogeeniset analogiat, turvallisuusperustelu.	
ISBN ISBN 978-951-652-192-6	ISSN ISSN 1239-3096
Sivumäärä – Number of pages 262	Kieli – Language Englanti

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GLOSSARY

ACAA	American Coal Ash Association.
ACZ	Alkaline cement zone.
AECL	Atomic Energy of Canada Limited.
AF4	Asymmetric flow field flow fractionation.
AFM	Atomic Force Microscopy.
Annual dose	Refers to the sum of the effective dose (see ‘dose’) arising from external radiation within the period of one year and the committed effective dose from the intake of radioactive substances within the same period of time (GD 736/2008).
AP	After Present.
ARAP	Alligator Rivers Natural Analogue Project.
BFZ	Brittle Fault Zone.
b.g.l.	Below ground level.
BIOPROTA	Project which was set up to address the key uncertainties in long term assessments of contaminant releases into the environment arising from radioactive waste disposal (www.bioprota.com).
BP	Before Present.
BWR	Boiling Water Reactors (Olkiluoto 1&2).
CARE	CAvern-REtrievable – concept for HLW/SF involving long-term storage in deep underground caverns before backfilling and closure of the caverns for final disposal.
CCS	Carbon Capture and Sequestration.
CER	Complementary Evaluations Report.
CFM	Colloid Formation and Migration (experiment in Grimsel URL).
CLAYTRAC	NEA’s project where natural tracer data were evaluated to assess potential impacts of disposal of radiological waste in geological repositories.
CRP	Coordinated Research Project.
CSH	Calcium Silicate Hydrate.
DFN	Discrete Fracture Network (an approach used in groundwater flow modelling).
DiP	(Government) Decision-in-Principle.
DOC	Dissolved Organic Carbon.
Dose	Effective dose, see also ‘Annual dose’.
EBS	Engineered Barrier System, which includes canister, buffer, backfill and closure.
ECOBA	European Coal Combustion Products Association.
EDX (EDAX)	Energy Dispersive X-ray analysis.

EDZ	Excavation Damaged Zone; section of the rock that is irreversibly damaged by the excavation of the tunnel.
Effective dose	The effective dose (E) to an individual is calculated from a weighted average of the whole-body (H) to different body tissues, with the weighting factors (W) designed to reflect the contribution of different tissue exposures to the risk to the whole organism: $E = \sum_i H_i W_i$.
EID	Earth Impact Database.
EIP	Environmental Integrity Project.
ENRESA	Company in charge of the management, storage and disposal of the radioactive wastes produced in Spain.
EP	Excavation Project.
EPA	U.S. Environmental Protection Agency.
EPR	European Pressurised water Reactor (trade name for the pressurised water reactor used at OL3).
ESCA	Electron Spectroscopy for Chemical Analysis.
FEBEX	Full-scale Engineered Barriers Experiment (experiment in Grimsel URL).
FEPs	Features, Events and Processes.
Fortum	Fortum Power and Heat Oy. Owner of the Loviisa nuclear power plants and co-owner of Posiva Oy.
GD	Government Decree.
GTS	Grimsel Test Site.
H12, H17	Japanese safety assessments.
HLW	High Level (radioactive) Waste.
HPF	Hyperalkaline Plume in Fractured Rock (experiment in Grimsel URL).
HZ	Hydrogeological Zone. A site scale hydrogeological zone is a planar or nearly planar formation that through elevated transmissivities and frequency of interconnected fractures allows a continuous groundwater flow to concentrate within it over distances of several hundreds of metres.
IAEA	International Atomic Energy Agency.
ICP-AES (ICP-OES)	Inductively Coupled Plasma-Atomic (optical) Emission Spectroscopy.
ICP-MS	Inductively Coupled Plasma-Mass Spectrometry.
IFA/UNEP/UNIDO	International Fertilizer industry Association, United Nations Environment Programme, United Nations Industrial Development Organization.
ILW	Intermediate Level (radioactive) Waste.

INTESC	INTernational Experiences in Safety Cases for geological repositories.
JAEA	Japan Atomic Energy Agency.
JNC	Japan Nuclear Cycle Development Institute. Japan Atomic Energy Research Institute (JAERI) and JNC unified and become JAEA in 2005.
JOGMEC	Japan Oil, Gas and Metals National Corporation.
KBS-3	An abbreviation of <i>kärnbränslesäkerhet</i> (nuclear fuel safety) version 3. The KBS-3 method for implementing the spent nuclear fuel disposal concept based on multiple barriers.
KBS-3H	(Kärnbränslesäkerhet 3-Horisontell). Design alternative of the KBS-3 method in which several spent nuclear fuel canisters are emplaced horizontally in each deposition drift.
KBS-3V	(Kärnbränslesäkerhet 3-Vertikal). The reference design alternative of the KBS-3 method, in which the spent nuclear fuel canisters are emplaced in individual vertical deposition holes.
LIBD	Laser-Induced Breakdown Detection.
L/ILW	Low and Intermediate Level (radioactive) Waste.
LLW	Low Level (radioactive) Waste.
LO1–2	Loviisa reactors 1 and 2.
LPG	Liquified Petroleum Gas.
m.a.s.l.	Metres above sea level.
MI	MIgration experiment.
M/ML	Magnitude / Magnitude Level.
MX-80	A high grade sodium bentonite from Wyoming, U.S., with a montmorillonite content of 75–90 % (properties as specified in this report and references herein).
NAS	U.S. National Academy of Sciences.
NASA	National Aeronautics and Space Administration.
NASCENT	Natural Analogues for the Storage of CO ₂ in the Geological Environment.
NAWG	Natural Analogue Working Group.
NEA	Nuclear Energy Agency.
NIED	National Research Institute for Earth Science and Disaster Prevention.
NORM	Naturally-Occurring Radioactive Material.
NRC	National Research Council of the U.S. NAS.
OL1–2	Olkiluoto 1 and 2 reactors.
OL3	Olkiluoto 3 reactor.

OL4	Fourth reactor to be constructed in Olkiluoto. Expected to be similar to OL3 in TURVA-2012 safety case.
ONKALO	Underground research facility constructed at Olkiluoto
OPA	Opalinus Clay – a candidate host rock for the Swiss repository for long-lived wastes. It is a Mesozoic (180 Ma old) sediment, described as an indurated claystone, occurring in the Zürcher Weinland.
Opal-CT	Opal formed of Cristobalite and Tridymite.
OPC	Ordinary Portland Cement.
Outcrop	The visible surface exposure of underlying geological units. Hard rocks such as granite and gneiss tend to form outcrops as they are more resistant to weathering processes, including glaciation, that erode softer rocks and form soil and surface sediments that cover potential outcrops (see also <i>roche moutonnée</i> , below).
PAH	Polysyclic Aromatic Hydrocarbon.
PAMINA	Performance Assessment Methodologies IN Application to guide the development of the safety case. European Union, Sixth framework project. Reports can be downloaded from: www.ip-pamina.eu .
PCS	Photon Correlation Spectroscopy.
POSE	Posiva's Olkiluoto Spalling Experiment.
P&T	Partitioning and Transmutation.
PWR	Pressurised Water Reactor.
PZC	Point of Zero Charge.
REE	Rare Earth Element.
Reference Area	This is the area approximately 100 x 200 km that encompasses the Olkiluoto site within it and within which reference mires and lakes were identified as the basis for the assessment of the future biosphere development of the Olkiluoto site (see Fig. 1-3 in Haapanen et al. 2010).
RMS	Risk Management Solutions (US commercial consultancy).
<i>Roche moutonnée</i>	Outcrop of hard rock that has been shaped by the action of a glacial movement and erosion. The roche typically has a low, smooth, rounded end pointing 'upstream', with reference to the direction of ice movement, and a higher, rougher, ice-plucked, downstream end. The surfaces may be marked by glacial striations.
RSC	Rock Suitability Classification. The aim of the RSC is to define suitable rock volumes for the repository, deposition tunnels and deposition holes.
RTD	Research, Technical development and Design (acronym RD&D is also used).

RTF	RadioToxicity Flux.
RTI	RadioToxicity Index (see definition in Appendix A).
RWMD	Radioactive Waste Management Directorate (of NDA).
SCC	Stress Corrosion Cracking.
SEM	Scanning Electron Microscopy.
SF	Spent (nuclear) Fuel.
SFR	Sparsely Fractured Rock.
SKB	Swedish Nuclear Fuel and Waste Management Co.
SKI	Swedish Nuclear Power Inspectorate, now SSM Swedish Radiation Safety Authority. The authority took over the responsibility and tasks from the Swedish Radiation Protection Institute and the Swedish Nuclear Power Inspectorate when these ceased to exist on 30 June 2008.
SPC	Single Particle Counting.
SRB	Sulphate-Reducing Bacteria.
STEM	Scanning-Tunnelling Electron Microscopy.
STUK	Radiation and Nuclear Safety Authority in Finland.
TDB	Thermodynamic Database.
TDS	Total Dissolved Solids.
TEM	Ministry of Employment and the Economy, previously Ministry of Trade and Industry (KTM) in Finland.
TEM	Transmission Electron Microscopy.
TENORM	Technologically-Enhanced Naturally-Occurring Radioactive Material.
TESM	Terrain and EcoSystems development Modelling.
TGG	Tonalitic-Granodioritic-Granitic gneiss.
THMC	Thermal-Hydrological-Mechanical-Chemical.
TILA-99	Name of Posiva's safety assessment 1999.
TOC	Total Organic Carbon.
TRUE	Tracer Retention Understanding Experiments.
TURVA-2012	Posiva's safety case supporting the construction licence application submitted in 2012 for the Olkiluoto spent nuclear fuel disposal facility.
TVO	Teollisuuden Voima Oyj. Owner of the Olkiluoto power plants and co-owner of Posiva Oy.
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation.
URL	Underground Research Laboratory.
VVER-440	Pressurised water reactor type at Loviisa

WFD	Water Framework Directive.
WP-Cave	Concept for long-term storage of spent nuclear fuel in an isolated rock cavern.
XRD	X-Ray Diffraction.
YVL	STUK's (see STUK) regulatory guide series for nuclear facilities.

FOREWORD

This report is a result of joint effort of several persons. Fiona Neall (Neall Consulting Ltd., UK) was the editor and also contributed to the report. Other contributors were Russell Alexander (Bedrock Geosciences, Switzerland), Heini Laine, Nuria Marcos and Thomas Hjerpe (all Saanio & Riekkola Oy, Finland), Graham Smith (GMS Abingdon Ltd., UK) and Marja Vuorio (Posiva Oy, Finland).

The progress of the report was supervised by the SAFCA project group consisting of Ari Ikonen and Marja Vuorio (Posiva Oy), Pirjo Hellä, Thomas Hjerpe, Heini Laine, Nuria Marcos, Barbara Pastina and Margit Snellman (all Saanio & Riekkola Oy), and Paul Smith (SAM Switzerland GmbH). They also reviewed the report (or parts of it). Paul Smith's input in particular, was essential to the final structure of the report. Annika Hagros (Saanio & Riekkola Oy, Finland) also reviewed the draft report at various stages.

The final report review was carried out by the following individuals: Mike Thorne (Mike Thorne and Associates Limited, UK), Lawrence Johnson (Nagra, Switzerland), Ivars Neretnieks (Chemima AB, Sweden) and Christer Svemar (SKB International AB, Sweden), their comments on the report are appreciated.

1 INTRODUCTION

The purpose of a repository for disposal of spent nuclear fuel is that individuals, society and the environment are protected from harmful effects of ionising radiation, now and in the future, in such a way that the needs and aspirations of the present generation are met without compromising the ability of future generations to meet their needs and aspirations (IAEA 1997). The consensus of the international opinion, based on examination of the wide range of potential waste management and disposal options available (see Chapter 4), is that only deep geological disposal offers the long-term passive safety required (NAS 1957; NEA 1999a,b; NRC 2003). The safety of geological disposal relies on natural and engineered (man-made) barriers, the functions of which are to isolate the spent nuclear fuel, to limit or prevent releases of radionuclides from the repository and to retard radionuclide transport, such that releases to the biosphere do not occur at unacceptable levels. The natural barrier consists of a host rock formation that must be of sufficient thickness to ensure isolation of the spent nuclear fuel at depth for as long as required. The host rock and the wider geological environment (the geosphere) must provide a stable and suitable physical and chemical environment to protect the engineered barriers.

In 1999, Finland selected geological disposal as the preferred long-term spent nuclear fuel management option (Government Decision 478/1999). The site at Olkiluoto in south-western Finland was chosen in 2000 after investigations at four candidate sites (Romuvaara, Kivetty, Hästholmen, Olkiluoto). The present report is part of the safety case in support of a license application to begin the construction of a deep geological repository at Olkiluoto (Figure 1-1).



Figure 1-1. Olkiluoto Island is situated on the coast of the Baltic Sea in south-western Finland. Photograph by Helifoto Oy.

1.1 Context of this report in the safety case documentation

Long-term safety of a spent nuclear fuel repository in Finland has been assessed and analysed since the planning of disposal began (e.g. Peltonen et al. 1985, Vieno & Nordman 1999). The first plan for the production of a safety case was presented in 2005 (Vieno & Ikonen 2005). That plan was based on the idea of a portfolio of reports that together would provide the argumentation for the long-term safety of a spent nuclear fuel repository at Olkiluoto. The plan was revised in 2008 (Posiva 2008). Particular attention is now given to the quality management of the safety case work, the management of uncertainties and the methodology for formulating and analysing scenarios. In the new plan, quality management is based on ISO 9001:2000 standard process thinking and enhanced with special features arising from the YVL Guides of the Radiation and Nuclear Safety Authority (STUK).

Since the plan presented in 2008, the portfolio has been expanded to cover aspects in the forthcoming updates of the YVL Guides as well as the feedback received from the authorities. The updated TURVA-2012 Safety Case portfolio is presented in Figure 1-2. The present report, *Complementary Considerations*, sits within the Safety Case portfolio and has the objective of enhancing confidence in the outcomes of the safety assessment. Reports included in the Safety Case portfolio (Figure 1-2) are referred to by name in the text (in italics). Full references for these reports are given at the beginning of the reference list.

1.2 Scope of the report

General scope of the report

Complementary Considerations addresses diverse and less quantifiable types of evidence and arguments that can be made to enhance confidence in the outcome of the safety assessment. In particular, demonstration of repository safety at times greater than a few thousands of years is not possible using conventional measures such as dose to an exposed population, so other arguments are required that rely on evidence gained from natural systems that have operated over geological timescales. A brief discussion of the difficulties and possibilities of demonstrating safety at long times is given below, as the scope of this report arises to a great extent from the need to take these considerations into account. Complementary considerations have been described (NEA 2004a, 2009c) as evaluations, evidence and qualitative supporting arguments that lie outside the scope of the other reports of the quantitative safety assessment. These arguments include, for example:

1. Support from natural systems for both key process understanding and total system performance;
2. Comparison of the methodology and results of safety cases made for other repository projects to ensure comprehensiveness, consistency and reasonableness of the present assessment;
3. Simplified bounding analyses of extreme, unrealistic cases for scenarios not considered in the quantitative safety assessment;

TURVA-2012		
Synthesis		
Description of the overall methodology of analysis, bringing together all the lines of arguments for safety, and the statement of confidence and the evaluation of compliance with long-term safety constraints		
Site Description	Biosphere Description	
Understanding of the present state and past evolution of the host rock	Understanding of the present state and evolution of the surface environment	
Design Basis		
Performance targets and target properties for the repository system		
Production Lines		
Design, production and initial state of the EBS and the underground openings		
Description of the Disposal System		
Summary of the initial state of the repository system and present state of the surface environment		
Features, Events and Processes		
General description of features, events and processes affecting the disposal system		
Performance Assessment		
Analysis of the performance of the repository system and evaluation of the fulfillment of performance targets and target properties		
Formulation of Radionuclide Release Scenarios		
Description of climate evolution and definition of release scenarios		
Models and Data for the Repository System	Biosphere Data Basis	
Models and data used in the performance assessment and in the analysis of the radionuclide release scenarios	Data used in the biosphere assessment and summary of models	
Biosphere Assessment: Modelling reports		
Description of the models and detailed modelling of surface environment		
Assessment of Radionuclide Release Scenarios for the Repository System	Biosphere Assessment	
Analysis of releases and calculation of doses and activity fluxes.		
Complementary Considerations		
Supporting evidence incl. natural and anthropogenic analogues		
	Main reports	
	Main supporting documents	

Figure 1-2. The TURVA-2012 safety case portfolio. The portfolio consists of safety case reports (green boxes) and supporting reports (blue boxes); brief descriptions of the contents are given (white boxes). Disposal system = repository system + surface environment.

4. Use of safety indicators other than dose to avoid having to take account of uncertainties in future human lifestyles (e.g. food production and consumption);
5. Use of complementary indicators that avoid having to account for biosphere evolution and geological processes on very long timescales;
6. Consideration of the calculation results from a wider perspective to consider significance of their assessed impact on human health and the environment compared with other risks.

The main emphasis in this report is on the evidence and understanding that can be gained from observations at the site and natural and anthropogenic analogues for the repository, its components and the processes that affect safety, corresponding to the first point, above. An introduction to the concept of natural analogues is included below as their central importance to the aims of this report means that a clear understanding is essential from the outset.

As described in Chapter 2, the relevant Finnish regulations (Guide YVL D.5, paragraph A09, Draft version 4, 17.3.2011, in Finnish) also require that complementary considerations include other methods of supporting the safety assessment by simplified bounding analyses (point 3 above) and considerations of the geological history of the disposal site. These subjects are however, included within more appropriate reports: *Performance Assessment* includes simplified, scoping calculations and *Site Description* includes consideration of the geological history of the site and its suitability for geological disposal of spent nuclear fuel: in the present report, the emphasis is on geological stability.

The use of complementary indicators for safety of the repository (point 4) is also included within *Assessment of Radionuclide Release Scenarios for the Repository System* and *Biosphere Assessment*, rather than the present report (a few examples only are used for spent nuclear fuel in Chapter 3). However, fairly comprehensive background information on some aspects of the use of complementary indicators is included in Chapter 11. The reason for including this material here is to put radioactive waste management in a wider perspective, and to show that there are sources of radionuclide fluxes arising from other industrial activities, and that these can be comparable with or larger than those arising from geological disposal of spent nuclear fuel. The comparisons (see point 5, above) are actually reported in *Assessment of Radionuclide Release Scenarios for the Repository System* and in *Biosphere Assessment*, but all the necessary background material is included within the present report.

It should be noted that the on-going work preparing for disposal is primarily being conducted with the KBS-3 vertical design variant (KBS-3V) in mind. However, in parallel with the preparation for the KBS-3V mode of disposal, an alternative horizontal disposal variant (KBS-3H) is being developed in cooperation with Sweden's Swedish Nuclear Fuel and Waste Management Co. (SKB). This report however is not so specific that differences in the design of 3V and 3H variants would greatly affect the conclusions. After all, the site, fuel and canister and most of the materials used are the same in both variants.

Assessing repository safety at long times

A major challenge in the development of a safety case for a deep geological repository for spent nuclear fuel is dealing with the long period of time over which the spent nuclear fuel remains hazardous (see Sections 2.2 and 3.3). Over such a period, a wide range of events and processes acts on a repository and its geological and surface environment. These events and processes, taking place over different time windows and at local to regional scales, result in increasing uncertainty in the future evolution of the repository and its environment. This means that arguments must be developed to show that this uncertainty can be addressed in a manner that is not only acceptable to regulators, who may in any case set out the types of arguments they want to see (see Chapter 2 for the regulatory requirements in Finland), but also convincing to less technical audiences who need to trust in the safety of the repository. Thus complementary lines of argument are required to compensate for increasing uncertainties affecting calculated releases at distant times. However, complementary arguments can also be made to address other aspects of safety, especially continuing isolation, even at times beyond when quantitative safety assessments can be supported. The Nuclear Energy Agency (NEA 2009a) suggests that “*complementary arguments might be based, for example, on the absence of resources that could attract inadvertent human intrusion and on the geological stability of the site, with low rates of uplift and erosion*”.

Another challenge with the long period addressed by the safety case is that although some experiments can be made in the laboratory, in underground research facilities (thus in the actual or similar host rock and geological environment) or in the field, these cover short timescales compared with long-term repository evolution. To try and address this specific problem, complementary arguments are made using analogous anthropogenic and/or geological examples of the materials and processes of interest (see the discussion of natural analogues, below) to show that understanding is good enough to extrapolate short-term experimental results to long-term performance.

A further challenge arising from the long time periods of interest in the safety case relates to how safety is quantified over these very long times. The most common indicators of safety are individual dose and risk (NEA 2002) and, of these, dose is much easier to communicate to a wider audience as it can be compared, for example, with the natural background radiation or medical radiation exposures (comparisons which are themselves complementary arguments).

Within the safety case, quantitative safety assessment using models and data tends to focus on potential radionuclide releases from a repository to the biosphere or surface environment. The uncertainties affecting the models can generally be quantified or bounded and dealt with in the safety assessment by using cautiously chosen parameter values, conservative model assumptions or evaluating multiple cases covering the ranges of uncertainty. However, where the consequences of calculated releases are to be expressed in terms of dose, the biosphere must also be modelled. The models of the way in which humans are exposed (e.g. ingestion via consumption of food or drinking of water) are closely related to human habits that can be predicted with confidence only in the very short term, basically in the order of decades. Quantitative criteria for radiation doses are applied only over time frames of a few thousands of years in accordance with

Finnish regulatory requirements. However, quantitative dose assessments may be useful beyond this time to show that radiological situation does not get dramatically worse after 3000 years.

To complement the quantitative estimates of doses, especially in the period beyond a few thousand years additional complementary safety indicators have been proposed by the International Atomic Energy Agency (IAEA 2003a) using fluxes and concentrations of naturally-occurring radionuclides in the undisturbed biosphere or geosphere for comparison with the calculated radionuclide releases from the repository. IAEA (2003a) also found that alternative indicators such as “crossover times” could be useful in illustrating safety. A crossover time is the point in time in the future at which either the activity or radiotoxicity (or radiotoxicity index – see Appendix A) of the radionuclides remaining in the engineered barriers or released to the geosphere decrease due to radioactive decay below the corresponding values for relevant natural materials such as the original uranium ore or the excavated host rock. The use of complementary indicators of safety is discussed further in Chapter 11.

The concept of natural and archaeological analogues

Argumentation by use of analogy is well established in many fields including philosophy, biology, linguistics and law (e.g. Petit 1992). For the specific case of radioactive waste management, the term “natural analogue” has developed a particular meaning associated with supporting arguments for the long-term safety of repositories (see Chapman et al. 1984, Miller et al. 2000). Key factors here are the heterogeneity and complexity of natural systems and, in particular, the very long timescales over which safety must be assured. The potential evolution of repositories, designed for specific types of waste and disposal site, can be simulated by the use of mathematical models, but the extent to which such models can be validated by conventional approaches is inherently limited. Here natural analogues – systems which have similar properties to components of repositories – have a unique role to play. It is not only natural systems that are of interest as archaeological materials can also provide analogues. The extent to which natural and archaeological analogue system evolution in the past can be understood and modelled with existing tools and data gives an indication of the ability to determine future development(s) of the repository.

The advantage of natural and archaeological analogues over short-term laboratory experiments is that they enable study of repository-like components and processes that have evolved over the timescales of relevance to a spent nuclear fuel repository safety assessment (rather than the days to months, or at most years, usual in laboratory tests). However, by their very nature, natural analogues often have ill-defined boundary conditions. By contrast, boundary conditions are generally well known in the well constrained (if less relevant) conditions of a laboratory. Thus, natural analogues and short-term laboratory experiments should generally be seen as complementary, with each having its own merits and limitations. Furthermore, well designed, realistic in situ field experiments can bridge the gap between the laboratory and natural analogues by offering repository-relevant conditions and intermediate timescales, with boundary conditions that are better defined than in most natural analogues (though perhaps less well defined than those of the laboratory experiments). In short, combining information from the three sources (i.e. long-term and realistic, if poorly defined, natural analogues;

medium-term, better constrained, in situ field experiments; short-term, less realistic but well-defined laboratory studies) can provide greater confidence that all relevant processes have been identified, as well as confidence in our understanding of the long-term impact of these processes. Such combined understanding also provides a basis for extrapolating laboratory-derived data to repository relevant timescales and conditions.

This is an important point as it is commonly not understood that it is not possible, as a general rule, to simply extrapolate the results from short-term experiments to a repository. For example, it is not possible to just say that if metal corrosion depth after 1 year was X mm, the corrosion depth after 1000 years will be 1000 times greater. It is now known that corrosion rates vary with time (due to the changes in environmental conditions, e.g. from aerobic to anaerobic conditions and due to build-up of corrosion products acting to protect the metal) and such extrapolation could lead to gross errors.

The initial use of natural analogues focused on improving understanding of key processes and model/database testing (e.g. McKinley 1989) and, indeed, this is still a major justification for some analogue projects. More recently, however, additional roles in public communication (e.g. West et al. 2002) and staff training (e.g. Alexander et al. 2008a) have received greater emphasis. In particular, using natural analogues to provide general support for the safety case (by studying the evolution of relevant systems over geological timescales) and to increase confidence in extrapolating results from laboratory and field experiments to the repository context have been a recent focus.

Improving system understanding can range from examining global concepts (e.g. fundamental feasibility of preserving geochemical anomalies for millions of years – see discussions in Miller et al. 2000 on Oklo and Cigar Lake, natural analogue sites in Gabon and Canada, respectively) to direct quantification of specific processes (e.g. matrix diffusion depths – see Smellie et al. 1985). Similarly, model testing can range from rather weak qualitative comparison of expectations with observations (e.g. relative retention of elements within Oklo reactor cores) to quantitative assessment of the relevance of laboratory databases (e.g. for material corrosion) to more formal assessment of the predictive capability of specific model and databases (e.g. blind predictive modelling of solubility limits; Pate et al. 1994). The challenge is to maximise the value of these tests, by assuring that materials and boundary conditions are as similar as possible to those in a repository. Nevertheless, it must be emphasised that relevant sites are no more than partial analogues of a repository, not a copy, and hence certain differences are inevitable.

It is beyond the scope of this report to provide an overview of the vast range of natural analogues that have been studied; for this the reader is invited to examine publications focused on just that, such as Miller et al. (2000, 2006), CSN (2004), Degnan et al. (2005) and Brasser et al. (2008). Rather, a few examples of relevant natural and archaeological analogues are presented for some of the more important areas of the safety case.

1.3 Feedback from KBS-3H Complementary Evaluations Report

A Complementary Evaluations Report (CER) for the KBS-3H design (Neall et al. 2007) was compiled as a part of the safety assessment of a KBS-3H spent nuclear fuel repository at Olkiluoto. STUK provided the feedback on the KBS-3H safety assessment including the CER as one of the main documents in their review (H221/4, H221/14, H221/15, STUK 2009). The feedback has been addressed in the on-going reporting, both in the present document and in other safety case portfolio reports as appropriate.

In their review of the report, STUK noted that the CER formally fulfils the requirements of STUK (Guide YVL E.5 now D.5), and emphasised that the role of complementary considerations will increase in resolving open issues identified by STUK. They also noted that the use of complementary safety indicators best addresses the requirements in Guide YVL D.5.

Specific questions were raised by STUK related to, for example, the way in which geothermal energy as a natural resource is exploited by a technology (borehole drilling) that may become an issue in the future. This is addressed in the Human Intrusion case in a BIOPROTA study (Smith et al. 2012).

Additional raised issues included ensuring the better use of monitoring data from the underground characterisation facility ONKALO at Olkiluoto to understand the THMC (Thermal-Hydrological-Mechanical-Chemical) evolution of the repository system. The impact of ONKALO construction and correspondence of the predictions with the observed outcome and thus testing of the site model are given in the Olkiluoto Site Description reports (Posiva 2005, 2009, *Site Description*). Further characterisation and modelling to study the EDZ (excavation damaged zone), rock spalling (POSE experiment), groundwater flow and salinity is on-going. *Performance Assessment* discusses the THMC evolution of the repository system.

Bounding analyses were also requested and further consideration of the geological history of the disposal site. These subjects are, as mentioned in Section 1.2, included within more appropriate reports: *Performance Assessment* include simplified, scoping calculations. *Site Description* includes consideration of the geological history of the site and its suitability for geological disposal of spent nuclear fuel. In the present report emphasis is on geological stability.

The consideration of analogues for processes arising from periods of glaciation, especially with respect to permafrost, was recommended and is addressed in the present report, as well as the recommended use of natural trace elements in Olkiluoto groundwaters as proxies for radionuclides released from the repository.

Cases to analyse, e.g. the effects of partial or complete loss of the safety functions of one component at a time as recommended by STUK will be addressed in *Assessment of Radionuclide Release Scenarios for the Repository System*.

1.4 Structure of the document

The structure of the report is as follows:

Chapter 1 discusses the context and scope of the report and provides an introduction to the use of complementary indicators for safety at long times and to the concept and use of natural analogues.

Chapter 2 discusses Finnish legal and regulatory requirements pertaining to the matters discussed in this report, as well as relevant international recommendations.

Chapter 3 describes the radiological hazards presented by spent nuclear fuel and gives the basic background information on the spent nuclear fuel properties and radionuclide inventory. Background information and data to support the arguments in this chapter are given in Appendix B at the end of the report.

Chapter 4 discusses the concept of geological disposal as a waste management option and the strengths and weaknesses of geological disposal compared to other options.

Chapter 5 discussed the KBS-3 method, the safety concept and the safety functions of the various components of the repository system.

Chapter 6 briefly describes the specific Finnish disposal system for spent nuclear fuel that is assessed in the present Safety Case, including a brief description of the Olkiluoto site.

Chapter 7 gives an overview of the evidence supporting the stability of deep host rock conditions with particular reference to the site suitability of Olkiluoto, including the geological and hydrogeological aspects as well as the resilience to external events.

Chapter 8 examines the natural and archaeological analogue evidence supporting the long-term stability of the repository system components and materials, with particular emphasis on the properties that are required for them to fulfil their assigned safety functions.

Chapter 9 looks at the natural analogues used to support the understanding of the key processes contributing to radionuclide retardation and slow migration.

Chapter 10 looks at the natural analogues used for future developments in the surface environment of the Olkiluoto site.

Chapter 11 introduces background information on the use of complementary indicators, such as concentrations and fluxes of naturally-occurring radionuclides in natural processes as well as in processes relating to major industrial activities.

Chapter 12 discusses the evolution of the repository system at very long times, over 1 Ma in the future.

Finally, Chapter 13 brings together the key points of evidence and arguments from the preceding chapters that support the robustness of the KBS-3 method, and the suitability

of Olkiluoto site, which together comprise the chosen system for disposal of spent nuclear fuel in Finland.

2 LEGAL AND REGULATORY REQUIREMENTS

2.1 Legal requirements concerning spent nuclear fuel disposal

The management of nuclear waste in Finland is regulated by the Nuclear Energy Act (990/1987) and the Nuclear Energy Decree (161/1988). These define, among other things, the liabilities of nuclear energy producers, the implementation of nuclear waste management, permit procedures and supervision rights. The Nuclear Energy Act was amended in 1994 so that all spent fuel and other nuclear waste created in Finland must be disposed of in Finland. Although the Nuclear Energy Act also prohibits the import and export of spent fuel and other nuclear waste to and from Finland, an amendment of the Act 10 (342/2008, issued 23 May 2008) allows the export of nuclear waste containing minor quantities of radioactive materials to other countries for appropriate processing.

According to the law, the Ministry of Employment and the Economy (TEM; previously the Ministry of Trade and Industry, KTM) decides on the principles to be followed in waste management of spent fuel and other nuclear waste.

The schedule for the disposal of spent nuclear fuel was established in the Ministry of Trade and Industry's decision 9/815/2003. According to the decision, the parties under the nuclear waste management obligation shall, either separately, together or through Posiva Oy, prepare to present all reports and plans required to obtain a construction license for a disposal facility for spent nuclear fuel as stated in the Nuclear Energy Decree by the end of 2012. The disposal facility is expected to become operational by the end of 2020.

The legislation concerning nuclear energy was updated in 2008. As part of the legislative reform, a number of the relevant Government Decisions were replaced with Government Decrees (GD). The decrees entered into force on 1st December 2008. The Government Decision (478/1999) regarding the safety of disposal of spent nuclear fuel, which particularly applied to the disposal facility, was replaced with Government Decree 736/2008, issued 27 November 2008.

Currently, the valid YVL Guides pertaining to nuclear waste management are Guides 8.1–8.5; additionally, a number of other YVL Guides may be applied in part to nuclear waste management. The Radiation and Nuclear Safety Authority (STUK) is in the process of updating its YVL Guides to comply with the new legislation. With the publication of the new YVL Guides, the guidelines will be updated and the number of separate Guides will be reduced. According to the current drafts, the Guides pertaining to the disposal of spent nuclear fuel will belong to the YVL-D series consisting of a total of five Guides. Guide YVL D.1 will deal with nuclear non-proliferation control, D.2 with the transport of nuclear material and nuclear waste, D.3 with the processing, storage and encapsulation of spent fuel, D.4 with nuclear waste management and decommissioning activities and D.5 with the disposal of nuclear waste. The latest draft of the Guide YVL D.5 (Draft 4, 17.3.2011 in Finnish only) was consulted for the preparation of this report.

2.2 Regulatory requirements for long-term safety

According to the Finnish Government Decree GD 736/2008, —*Compliance with the long-term radiation protection requirements as well as suitability of the disposal method and site shall be demonstrated by means of a safety case, which covers both expected evolution scenarios and unlikely events impairing long-term safety.* Furthermore, it is stated that —*The safety case comprises a computational analysis based on experimental studies and complementary considerations insofar as quantitative analyses are not feasible or involve considerable uncertainties.*

The regulatory requirements for the long-term safety of disposal are given in Guide YVL D.5 and its appendix. The guide lays out the principles according to which the disposal systems must be designed, built and operated. The appendix of Guide YVL D.5 gives the framework for the safety case for disposal that must be provided by the implementer.

The third Chapter defines the radiation safety requirements based on Nuclear Energy Act 990/1987 Section 7h:

Disposal of nuclear waste shall be planned so that as a consequence of expected evolution scenarios

- *the annual dose¹ to the most exposed people shall remain below the value of 0.1 mSv; and*
- *the average annual doses to other people shall remain insignificantly low.*

These constraints are applied in a time window during which the radiation exposure of humans can be estimated with sufficient reliability, and which extends over at least several millennia (Guide YVL D.5, paragraph 311). The dose assessment shall take into account changes in the environment that result from variations in sea level and from land uplift. The climate type, human habits and needs can be presumed to stay unchanged. The analysis shall at least take into account the use of contaminated water and use of contaminated agricultural and natural products.

For timescales exceeding several millennia, constraints on annual radioactive releases to the environment are provided by STUK, as follows:

Disposal of nuclear waste shall be planned so that, as a consequence of expected evolution scenarios, the average quantities of radioactive substances over long time periods, released into the environment from disposed waste, shall remain below the constraints specified separately for each nuclide by the Radiation and Nuclear Safety Authority.

¹ Annual dose refers to the sum of the effective dose arising from external radiation within the period of one year and the committed effective dose from the intake of radioactive substances within the same period of time (GD 736/2008). In this report “dose” refers to effective dose, and “annual dose” refers to the annual effective dose, unless otherwise explicitly stated.

1) at a maximum, the radiation impacts arising from disposal can be equivalent to those arising from natural radioactive substances in earth's crust

2) on a large scale, the radiation impacts remain insignificantly low (GD 736/2008).

These constraints are applied for limiting the radiation exposures arising beyond the assessment period during which the radiation exposure of humans can be assessed with sufficient reliability, and which shall extend at a minimum over several millennia (Guide YVL D.5, paragraph 311).

The nuclide specific constraints for the radioactive releases to the environment (average release of radioactive substances per annum) referred to above are as follows:

- 0.03 GBq/a for the long-lived, alpha-emitting radium, thorium, protactinium, plutonium, americium and curium isotopes
- 0.1 GBq/a for the nuclides Se-79, Nb-94, I-129 and Np-237
- 0.3 GBq/a for the nuclides C-14, Cl-36 and Cs-135 and for the long-lived uranium isotopes
- 1 GBq/a for the radionuclide Sn-126
- 3 GBq/a for the nuclide Tc-99 and Mo-93²
- 10 GBq/a for the nuclide Zr-93
- 30 GBq/a for the nuclide Ni-59
- 100 GBq/a for the nuclide Pd-107.

Guide YVL D.5 states that the unlikely events that need to be analysed in the safety case include at least: rock movements that may damage the canisters, drilling of a medium-deep water well on the disposal site, and core drilling or boring hitting a canister.

It is also stated that the radioactivity arising from disposal shall not detrimentally affect non-human biota. The radiation exposure caused to the non-human biota shall stay well below the levels which could cause a decline in biodiversity or cause recognisable harm to any living population.

Finally, both Decisions-in-Principle (DiPs) by the Parliament (2000, 2002) state that structural solutions shall be designed to prevent an uncontrollable chain reaction fed by neutrons, i.e. the spent nuclear fuel packages shall be designed to prevent the spent nuclear fuel from reaching criticality in repository conditions. This is translated into regulatory requirements for the processing, storage and encapsulation of spent fuel given in Guide YVL D.3.

² Mo-93 is not mentioned in Guide YVL D.5. However, based on a preliminary evaluation by STUK (personal communication May 9, 2012) that takes into account the inventory and the dose conversion factors for ingestion, it has been recommended that the same geo-bio flux constraints as for Tc-99 are used.

2.3 Regulatory requirements specific to this report

Guide YVL D.5, paragraph A09 includes more detailed requirements for the safety case and complementary considerations: *“The importance to safety of such scenarios that cannot reasonably be assessed by means of quantitative safety analyses, shall be examined by means of complementary considerations. They may include e.g. analyses by simplified methods, comparisons of the geological history of the disposal site. The significance of such considerations grows as the assessment period increases, and safety evaluations extending beyond time horizon of one million years can mainly be based on the complementary considerations. Complementary considerations shall also be applied parallel to the actual safety assessment in order to enhance the confidence in results of the analysis or certain part of it.”*

These requirements are consistent with the international consensus about the use of multiple, complementary lines of reasoning. One of the outcomes of the NEA’s INTESC (International Experiences in Safety Cases for Geological Repositories) Project is: *“Confidence in the plausibility of the [safety assessment] results can also be enhanced by providing multiple lines of reasoning for safety in addition to the calculated numerical values of e.g. dose or risk. Such complementary evidence and lines of argument may include observations of natural systems to support that safety over geological timescales is achievable at the site in question or studies to demonstrate the feasibility of engineering designs”* (NEA 2009c).

In addition to the main evidence provided by the extensive programmes of site characterisation and research and development on the engineered barrier system, the complementary lines of evidence include (NEA 2009c):

- corroborative analyses, i.e. alternative analyses to study the importance of assumptions and to confirm the fundamental understanding of the processes³;
- the strength of geological disposal vs. other concepts;
- support from natural and man-made analogues;
- qualitative analyses based on reasoned arguments that are scientifically sound (e.g. the analysis to ensure that all FEPs (features, events and processes) have been taken into account in the safety assessment)⁴.

Considering the handling of the very long time frames involved in the safety case, the INTESC project outcome report states: *“In considering safety (and the safety case) beyond the time frame covered by quantitative safety assessment, other qualitative arguments may be used. There can be powerful arguments – with a very credible basis – related, for example, to the expected stability of the selected geologic setting. These types of arguments have been made in past safety cases, typically with good external reception”* (NEA 2009c).

³ As noted previously, these analyses are documented within other reports, particularly the *Performance Assessment* report.

⁴ As with the corroborative analyses, these qualitative analyses are documented in other reports, mainly the *Assessment of Radionuclide Release Scenarios for the Repository System* report.

STUK's requirements and the international guidelines on complementary considerations are specifically taken into account in this report. The complementary indicators of safety, which can be applied parallel to the safety assessment based on the main line of reasoning, are discussed in Chapter 11. The results of the safety analysis are however, presented in *Assessment of Radionuclide Release Scenarios for the Repository System*.

3 THE RADIOLOGICAL HAZARD PRESENTED BY SPENT NUCLEAR FUEL

The radiological hazard presented by spent nuclear fuel was discussed in some detail in the earlier Complementary Evaluations Report (CER) (Neall et al. 2007), following the method of Hedin (1997). As the purpose of the disposal is to mitigate that hazard, it is appropriate to discuss the magnitude of the hazard in the context of the Finnish spent nuclear fuel inventory and site in this report.

Since the CER, the total inventory of spent nuclear fuel destined for the Olkiluoto repository has increased and the relative amounts of different reactor fuel types has also changed, so this chapter also includes an examination of the impacts of those changes.

3.1 Types of hazard

Following the terminology of IAEA (2007), the hazard associated with spent fuel can be expressed as the product of the probability of radiation exposure and the probability of a specified health effect occurring in a person or group as a result of that exposure. The type of exposure and health effect(s) in question must be stated. The hazard of spent nuclear fuel is mainly related to its radiotoxicity, which is a function of the time-dependent activity of the spent nuclear fuel and the type of radioactive emissions involved, but there is also a contribution from its chemotoxicity, i.e. when taken into or present in contact with the human body, spent nuclear fuel may cause harmful effects by way of chemical reactions. The hazard due to chemotoxicity is not addressed further here⁵, but it is of limited significance compared with the radiotoxicity except at very long times after disposal when the residual radioactivity in the repository is primarily due to U-238 and its progeny.

The probability of exposure is a measure of the likelihood of a person, or group of persons, being exposed to radiation in different situations, i.e. during transport, during interim storage or after deep disposal of spent nuclear fuel. The potential health effect due to the radiation exposure in question must also be stated, e.g. risk of deterministic effects (hematopoietic, gastrointestinal and neurovascular syndromes) or stochastic effects (risk of induced cancer or serious hereditary effects). The discussion here is limited to stochastic health effects, caused by internal or external exposure to radiation.

When the source of radiation is outside the human body, the main hazard from spent nuclear fuel is related to exposure to external γ - (and neutron-) radiation.

Other types of radiation (α - and β -radiation) are hazardous mainly if the radioactive substances enter the human body, primarily via ingestion of food and water and inhalation.

⁵ Neall et al. (2007) provided a brief summary of the assessments of chemotoxicity of radionuclides released to the biosphere that had been carried out at that time. Neall et al. (2007) also listed some of the methodological difficulties in assessing chemotoxicity. A more recent overview of the methodology is given in Wilson et al. (2011) with some suggestions for how these difficulties might be overcome.

3.2 Spent nuclear fuel inventory and characteristics

Nuclear fuel consists of sintered uranium dioxide (UO_2) pellets enriched to roughly 3–5 % (i.e. increasing U-235 relative to U-238 from its natural abundance of 0.72 %). The pellets are stacked, according to design specifications of the reactor core, into fuel rods – sealed cladding tubes of corrosion-resistant metal alloy (zirconium alloy) – and the fuel rods grouped into assemblies that form the core. More information about dimensions and components of the fuel assemblies is given in Chapter 6 (Section 6.2).

The current inventory taken into account in TURVA-2012 safety case for the Olkiluoto repository is 9000 tU (see *Description of the Disposal System*). This is made up of several different types of spent nuclear fuel that originate from the various Finnish nuclear power plants (*Design Basis*):

- 2893 tU spent nuclear fuel from the Boiling Water Reactors (BWR) at Olkiluoto (OL1 and OL2); this is referred to as BWR fuel;
- 1107 tU spent nuclear fuel from the VVER reactors (Russian type pressurised water reactors) at Loviisa (LO1 and LO2); this is referred to as VVER fuel;
- an estimated 5000 tU spent nuclear fuel from the Pressurised Water Reactor (PWR) under construction at Olkiluoto (OL3) and from the planned fourth reactor at Olkiluoto (OL4) for which it is assumed that the spent nuclear fuel will be similar to that from OL3.

The different spent nuclear fuel types have different characteristics that are a function of the reactor design, enrichment of U-235 compared with natural uranium, the design of the fuel assemblies and the irradiation history (e.g. heat generation rate and ‘burn-up’, that is, the thermal power generated per kilogramme of uranium expressed in MWd/kgU).

The initial activity arising from the three spent nuclear fuel types, and its decay over time after discharge from the reactor (i.e. cooling time), is compared in Figure 3-1 (using data from Anttila 2005) for fuel with an initial enrichment of 4 %. Spent nuclear fuel with a burn-up of 40 MWd/kgU is compared in three of the curves so that the differences between the reactor fuel types can be seen. However, the key parameter characterising spent nuclear fuel especially that produced over a lengthy period when nuclear power plant operating conditions may change is the burn-up. Generally, higher burn-up fuel has a higher activity and decay heat on unloading from the reactor. This difference can be seen with the fourth curve on Figure 3-1, for OL3 fuel with a burn-up of 60 MWd/kgU. The maximum anticipated burn-up for LO1–2 is 55 MWd/kgU and for OL1–3 is 50 MWd/kgU, although the trend is to further increase the burn-up in the future (Saanio et al. 2009).

In previous studies (e.g. KBS-3H Safety Studies: Smith et al. 2007a,b, Neall et al. 2007), OL1–2 (BWR) spent nuclear fuel with an enrichment of 4 % and burn-up of 40 MWd/kgU was used as the reference fuel. As can be seen from Figure 3-1, this is slightly non-conservative in terms of the activity of the spent nuclear fuel over time. Here, the OL3 spent nuclear fuel with a burn-up of 60 MWd/kgU (and 4 % enrichment) is used as the reference fuel. This is the value for maximum assembly average burn-up

as used in the safety case (Pastina & Hellä 2010) and it is a cautious value as it overestimates the inventory.

The radionuclide inventories presented in Anttila (2005) are used in this chapter as these are considered for the purpose of demonstrating the scale of the hazard associated with spent nuclear fuel.

After discharge from a reactor, the spent nuclear fuel is stored under water until radiation intensity and decay heat decrease sufficiently to allow encapsulation. The decay heat for each canister is limited by design, depending on the type of fuel: 1830 W or 863 W/tU (OL3 fuel), 1700 W or 806 W/tU (OL1–2 fuel) and 1730 W or 950 W/tU (LO1–2 fuel) (Raiko 2005). The heat load limits are used for repository dimensioning purposes and to ensure that the temperature in the buffer surrounding the canister does not exceed 100 °C. The maximum design temperature for the surface of the canister was set to 90 °C in Raiko (2005). This gives rise to a necessary cooling time of between 30 and 50 years, depending on burn-up. Figure 3-2 shows the decay heat generated by the different fuel types – not surprisingly it mirrors the activity decrease shown in Figure 3-1. The cooling period of 30–50 years is marked on the figure along with the maximum decay heat load for an OL3 canister. It is clear that whereas 30–50 years is sufficient for the 40 MWd/kgU burn-up fuels to meet the heat load limits, the high burn-up OL3 fuel would require special measures or longer cooling times. In order to get a sense of the long-term heat output, it may be worth noting that after 1000 years, the decay heat per tonne of spent nuclear fuel is about the same as the heat generated by an adult human (~100 W per person).

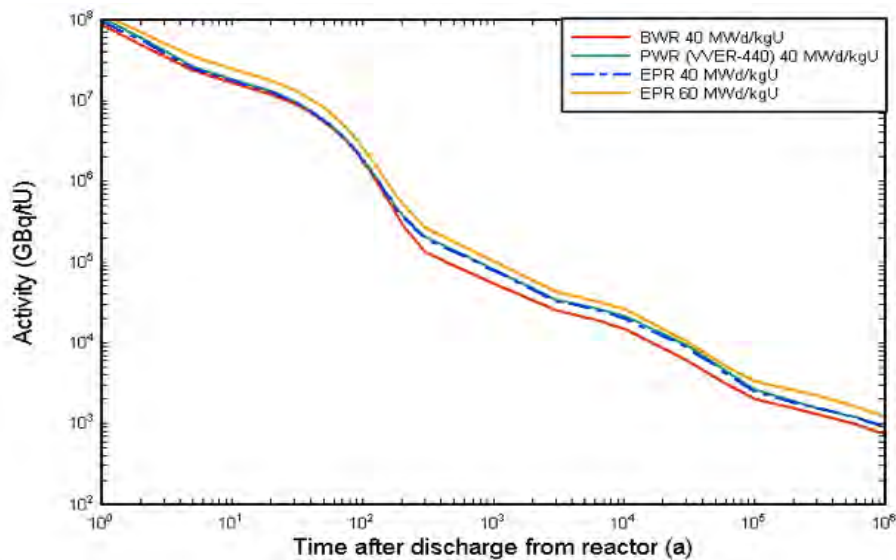


Figure 3-1. Spent nuclear fuel types from the three different reactors and their activity over time after unloading from the reactor. The enrichment for each fuel type is 4.0 %. The burn-up was 40 MWd/tU for each type apart from the fourth (yellow curve) EPR 60 MWd/kgU which shows the impact of the higher burn-up for the OL3 PWR (EPR = European Pressurised Reactor) fuel (data from Anttila 2005).

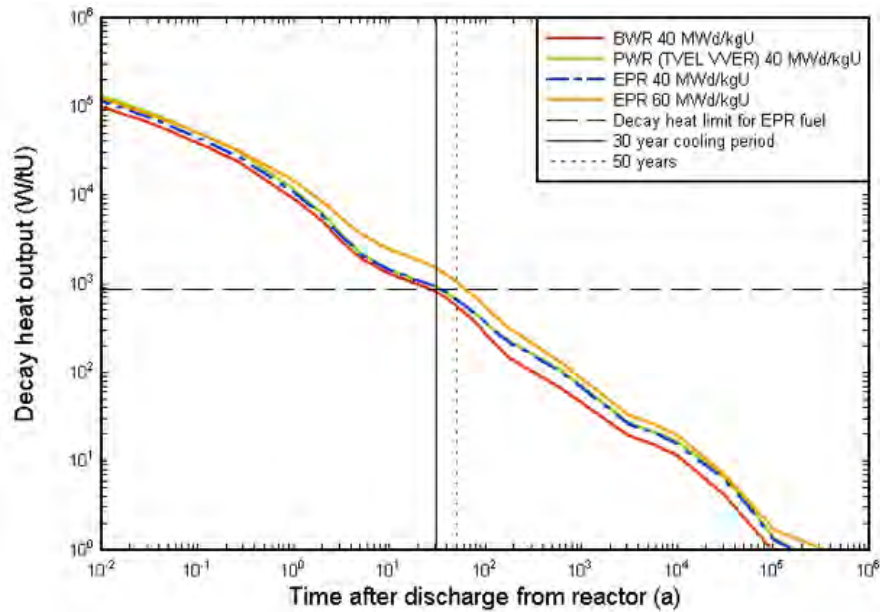


Figure 3-2. Spent nuclear fuel types from the three different reactors and their decay heat output over time after unloading from the reactor. The enrichment is 4.0 %. The burn-up was 40 MWd/tU for each type apart from the (yellow curve) EPR 60 MWd/kgU, again showing the impact of the higher burn-up for OL3 fuel (data from Anttila 2005).

3.3 Evolution of radiological hazard over time

The radiotoxicity index (RTI, defined in Appendix A) for 1 tonne and 9000 tonnes of Finnish spent nuclear fuel to be disposed of in the repository at Olkiluoto is shown in Figure 3-3 as a function of time after removal from the reactor (data from Appendix Table 4.2.3.4, Anttila 2005). This is compared with the natural radionuclides contained in 1.15 km³ ⁽⁶⁾ of tonalite host rock at Olkiluoto⁷ and with that of natural uranium ore corresponding to the volumes of the KBS-3V deposition drifts. The tonalitic-granodioritic-granitic-gneiss is also used as an example to estimate the RTI of the volume of Olkiluoto host rock removed during excavation of the deposition drifts; this RTI is also shown in Fig. 3-3.

For the natural uranium ore, different uranium concentrations (uranium ore grades) are considered: 8 %, which is the average concentration for the Cigar Lake uranium deposit in Canada (Cramer & Smellie 1994) and 0.16 % which is the average grade of the uranium mineralisation in the Finnish Palmottu U deposit (Ahonen et al. 2004).

⁶ 1.15 km³ is used as a reference volume since it is approximately the volume of rock between the repository at 420 m depth and the surface, using an approximate repository footprint of 2.7 km², which is slightly larger than the approximately 2 km² given in *Description of the Disposal System*.

⁷ See Chapter 6 for a description of the rock types at the Olkiluoto site. The proportion of tonalitic-granodioritic-granitic gneisses (TGG) is about 8 % in the drill cores studied so far at Olkiluoto (Andersson et al. 2007), and thus the examples presented in this report are only indicative.

Figure 3-3 shows that the radiotoxicity of the spent nuclear fuel is substantially reduced over the course of time. After 1 million years, the radiotoxicity of the total inventory of spent nuclear fuel has dropped to below that of a volume of natural uranium ore (Cigar Lake 8 % U) sufficient to fill the KBS-3V deposition tunnels and is close to that of the 1.15 km³ of Olkiluoto tonalite which overlies the repository footprint.

Another way of putting the potential hazard of spent nuclear fuel in perspective is to compare the activity contained in spent nuclear fuel with the activity in natural uranium that was mined and enriched to produce the fuel (Hedin 1997). Figure 3-4 shows the radiotoxicity index of 1 tonne of Finnish OL3 spent nuclear fuel and the main contributing radionuclides as a function of time. This is compared with the RTI of 8 tonnes of natural uranium that was used to produce the fuel (from Hedin 1997). After about 250,000 years, the radiotoxicity of spent nuclear fuel has dropped to that of the natural uranium from which it was produced (if it is assumed to be in equilibrium with its progeny). This point is often termed ‘the cross-over’ – the time at which the spent nuclear fuel might be said to represent a lesser hazard than the uranium ore from which it was produced.

Figure 3-5 shows the activity of 1 tonne of OL3 spent nuclear fuel with a burn-up of 60 MWd/kgU as a function of time. In this case, the data are normalised to the activity of the equivalent amount of natural uranium ore used to produce the spent nuclear fuel. This figure essentially shows the same data as Figure 3-4 in another form.

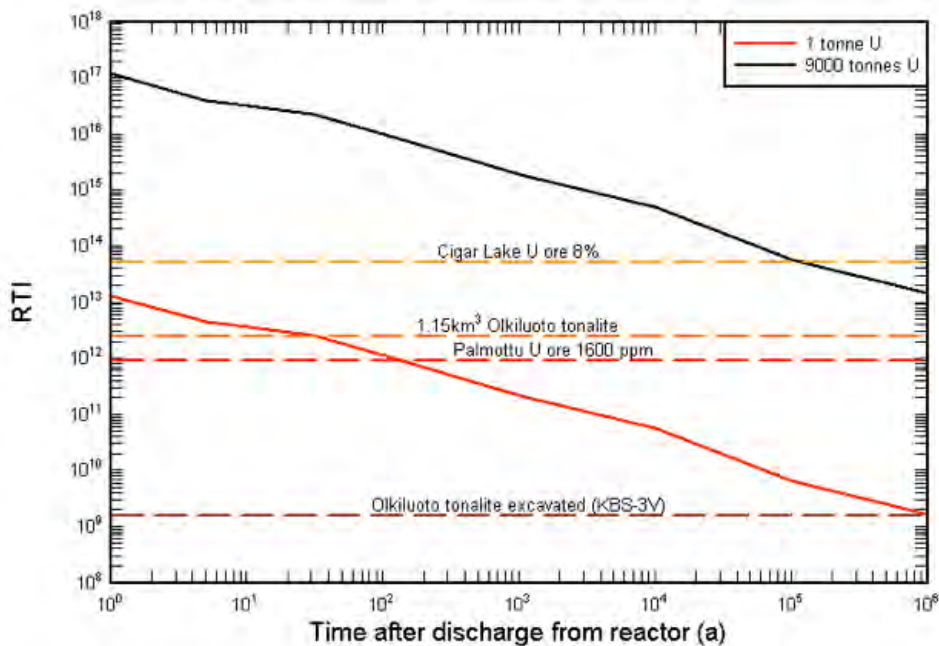


Figure 3-3. Radiotoxicity index (RTI, defined in Appendix A) of 1 tonne and 9000 tonnes of Finnish spent nuclear fuel. Also shown are the RTI values for the volume of different grades of U which would fill the deposition tunnels of the KBS-3V repository, as well as the RTI for the volume of Olkiluoto host rock that would be removed during excavation of the deposition tunnels for a KBS-3V repository (taking the Olkiluoto “tonalite” as representative of the TGG at the site, see Table B-6 in Neall et al. 2007). The calculations and data for this figure are given in Appendix B.

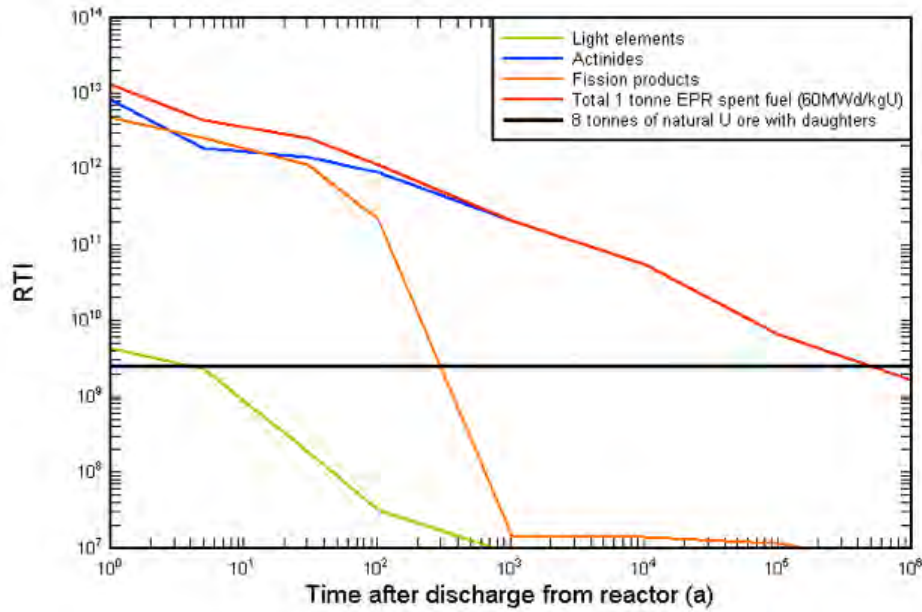


Figure 3-4. Radiotoxicity index (RTI) of 1 tonne of Finnish OL3 (EPR in figure) spent nuclear fuel with a burn-up of 60 MWd/kgU and the radionuclide groups contributing to the RTI (data from Anttila 2005). Also shown is the RTI of 8 tonnes of natural uranium from which the precursor fuel was derived.

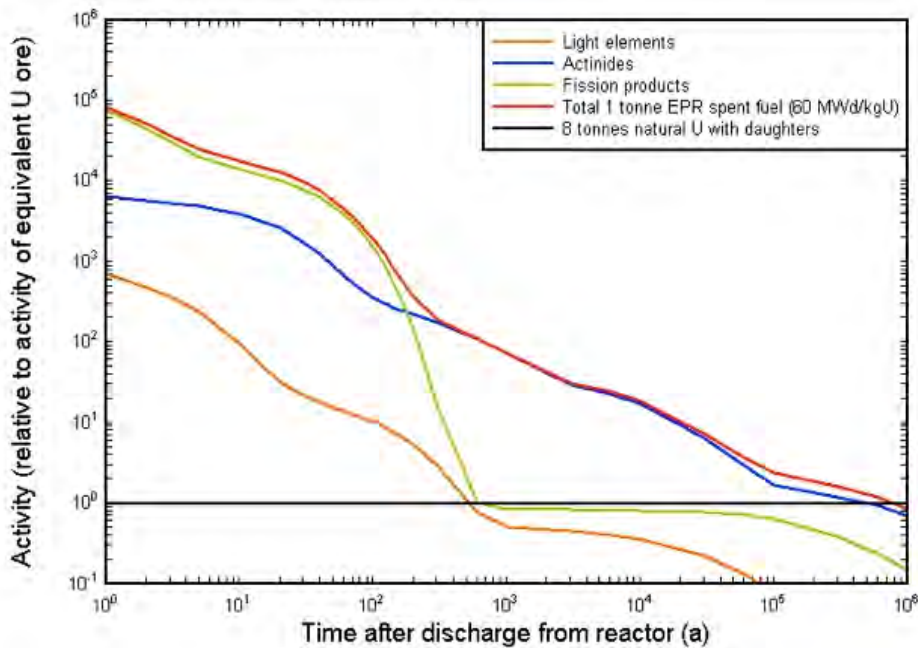


Figure 3-5. Relative activity of uranium ore and spent nuclear fuel as a function of time. The total activity of one tonne of Finnish OL3 (EPR in figure) spent nuclear fuel with a burn-up of 60 MWd/kgU is shown (relative to the natural U ore activity of approximately 1.4 TBq). The spent nuclear fuel activity is also shown separated by type of radionuclide. The light elements and fission products decay relatively quickly so that the activity of the spent nuclear fuel after 1000 years is mainly due to the actinides and their progeny.

As in Figure 3-4, the activity of spent nuclear fuel is substantially reduced over the first 1000 years after removal from the reactor as the activation and fission products decay. But what is interesting when activity rather than RTI is considered is that the cross-over occurs much later, at around 750,000 years. This indicates that it is the shorter-lived activation and fission products that contribute more strongly to the radiotoxicity of the waste and the continuing activity of the long-lived actinides and decay products present relatively less of a hazard. Of all the radionuclides initially contained in spent nuclear fuel, basically what will remain in the far future, are the long-lived uranium isotopes U-238 and U-235, with half-lives of $4.5 \cdot 10^9$ years and $7 \cdot 10^8$ years, respectively, together with the radionuclides that are continuously formed by radioactive decay. Thus, the nuclear fuel cycle gives rise to a significantly increased activity during a period of roughly 1 million years. Thereafter, the total activity of the fuel cycle material is roughly equal to the natural uranium minerals that were used to produce an equivalent amount of fuel.

As noted in NEA (2009a), such comparisons need to be used with caution. This is not only because the isotopic compositions of natural systems will differ from those of the initial spent nuclear fuel, the eventual repository releases and the remnants of the spent nuclear fuel in the repository at long times but also because the assumption should not be made that natural occurrences of uranium ore are necessarily harmless. Furthermore, the comparisons do not address the hazard associated with external radiation (although, as noted previously, this hazard is very considerably removed by the location of the spent nuclear fuel within a deep geological repository), nor the different “accessibility” of the radiotoxic material when it is in its natural form compared to the spent nuclear fuel surrounded by the man-made barriers. Figures 3-3, 3-4 and 3-5 nevertheless indicate that the timescale over which spent nuclear fuel presents a hazard deserving special attention is in the order of one million years. This, however, does not imply a requirement for complete containment⁸ of radionuclides by the repository for this period of time. The surrounding barriers (buffer and geosphere), being part of the isolation system, are expected to mitigate any potential radiological effects of an earlier than expected canister failure. Nor does it imply that the time period in excess of one million years can be ignored in the safety case. As also noted in NEA (2009a), in addition to U-238 with a half-life of $4.5 \cdot 10^9$ years, other radionuclides (especially some of those created artificially in nuclear reactors) that could be important in terms of the hazard from external radiation persist out to one million years or longer. Thus, even though the hazard potential of spent nuclear fuel decreases markedly over time, spent nuclear fuel can never be said to be intrinsically harmless.

⁸ the definition used here for the containment is also supported by IAEA definition (IAEA 2007).

4 GEOLOGICAL DISPOSAL AS A WASTE MANAGEMENT OPTION

Studies of different options for spent nuclear fuel management have been going on for 5 decades (e.g. NAS 1957). Presently, geological disposal is internationally the preferred strategy for disposal of spent nuclear fuel or long-lived high level radioactive waste (HLW) (IAEA 2003a, CORWM 2006, Blue Ribbon Commission 2012). The safety of geological disposal relies on natural and engineered (man-made) barriers, the function of which is to prevent or retard release of radionuclides from the repository to the biosphere at unacceptable levels. The natural barrier consists of a bedrock formation which must be of sufficient depth to ensure continued isolation. Engineered barriers are designed to support bedrock with specific characteristics and to isolate the radioactive substances for long periods of time.

This chapter gives a brief review of the development of geological disposal concepts. The review concentrates on methods suitable for HLW disposal, especially spent nuclear fuel, and the concepts discussed are those where the geosphere makes a significant contribution to the safety case. The historical evolution of the disposal options from a wider point of view (including LLW and ILW) can be found for example in McKinley et al. (2007).

A brief summary of the alternatives to geological disposal is also given below, along with the justification for not considering them further. The alternatives to geological disposal are here divided (according to McKinley et al. 2007) into politically blocked options, technically impractical options and non-options (e.g. indefinite storage).

In 1999, Finland selected geological disposal as the preferred long-term spent nuclear fuel management option (Government Decision 1999/478). A brief discussion of the disposal options discussed in the early phase of the Finnish nuclear waste management programme is included at the end of this chapter.

4.1 Alternatives to geological disposal

Sub-seabed and Antarctic icecap disposal are the politically blocked options discussed here. Technically, sub-seabed disposal is equivalent to disposal on land with some benefits (such as high dilution of releases, low porewater fluxes, very low risk of human intrusion). There are several studies of disposal variants; however most of the options are banned by international convention with the exception of sub-seabed formations which are accessed from land. The main problem with coastal facilities is that given the long timescales involved, the effect of sea-level changes needs to be considered very carefully.

Antarctic ice sheet disposal is suggested from time to time due to the isolation from population centres, however Article 5 of the Antarctic Treaty (1961) explicitly prohibits disposal of radioactive waste.

Technically impractical solutions are partitioning and transmutation (P&T), (non-Antarctic) ice sheet disposal, disposal into oceanic subduction zones and disposal in space. P&T is theoretically possible but more research is needed before the method can be implemented. The principle behind P&T is that long-lived isotopes are irradiated to

produce shorter-lived products. The drawbacks are that there would be a need for new reactors which have to be in operation for more than 100 years, and there would still be waste that would require disposal. There would also be an increased risk of exposure to workers and, based on present experience with reprocessing, the net result of P&T would be to replace a homogeneous, very long-lived waste with a waste product that is likely to be a chemically heterogeneous mixture of isotopes with a wider range of half-lives – making the task of disposal more complicated.

Space disposal relying on existing technology can be ruled out as a very high risk, high consequence strategy. Rocket launches are high cost and have a large environmental impact. Statistically the risk of catastrophic failure during a spacecraft launch is significant (in the range of 1–10 %, depending on the space programme, McKinley et al. 2007) and the consequences of spreading a load of nuclear waste across the earth are potentially catastrophic. More exotic alternatives for space transportation have been suggested such as rail-guns and space elevators. These studies only exist on paper and would need major advances in materials technology and are associated with high costs and risks.

Disposal of HLW and spent nuclear fuel into oceanic subduction zones is currently banned under the London Convention 1972 (IAEA 1972) which forbids the dumping of wastes at sea. However, even if this were not the case, as a concept for radioactive waste disposal it has major problems, particularly with implementation (e.g. waste containers would need to be drilled into the descending oceanic plate to ensure that they were not merely mangled along with the seafloor sediments during subduction) and quality assurance (e.g. there may be a very long time between disposal of waste and actual subduction as the subduction zones are not uniform conveyor belts, and how is integrity of the waste maintained over this period).

Disposal in ice sheets and permafrost have been studied for a long time, however the Antarctic Treaty excludes the use of polar caps leaving only sites in Greenland and certain Canadian and Russian islands as alternatives. As this disposal method relies on the permanence of the ice sheet over the timescale of interest, global warming concerns have now reduced the credibility of this option. Another downside is the remoteness of these areas, which is good from a safety viewpoint, but add to the handling and transportation costs.

The non-option mentioned earlier is long-term surface storage. Monitored storage is already part of the management of spent nuclear fuel for a limited period of time. One concern here is the longevity of institutions needed for the control or monitoring of long-term storage. This option would transfer the responsibility to future generations, which is not considered to be an ethical solution. The IAEA has also expressed the opinion that true institutional control is realistic only for wastes that contain predominantly short lived isotopes (IAEA 1992, 2002, 2003a).

Some reasons have been suggested for delaying final disposal and relying on long-term storage for the present, e.g. technology may be developed which provides a better management option, or the waste could be considered as a resource that may be utilised in the future. The former argument though is contrary to the ethical principle that the generations benefiting from nuclear power also should take responsibility for the

resulting waste. The latter argument has led to the idea of combined deep storage with disposal, as discussed below, as an alternative to conventional repositories.

4.2 Geological disposal options

Discussion of geological disposal is usually taken to refer to specially designed and constructed repositories. It is possible, however, to use existing underground cavities for disposal, e.g. disused mines, particularly as an option for lower activity or shorter-lived waste. Here only specially designed repositories for HLW and spent nuclear fuel are discussed.

Extended storage options, e.g. CAvern-REtrievable (CARE), combine monitored storage provided by long institutional control and a disposal system. With the CARE concept, the basic components of a typical intermediate level waste facility are implemented in a geological setting. HLW waste packages are contained in multipurpose casks, possibly after a period of surface storage, and transferred underground for a further long-term storage period. During this period the waste casks can be monitored and retrieved at any time and, when so decided, the storage caverns can be backfilled to form a permanent disposal facility. With this option, the ethical dilemma of passing the responsibility to future generations is at least partially avoided. The operational safety during this long institutional control period is still an issue, since perturbations such as earthquakes and civil disturbances can have an impact. It should also be noted that this concept, and similar extended storage options, have not been the subject of in-depth feasibility or safety studies. It could be that, in providing additional flexibility for waste management, they may be difficult to engineer to provide adequate levels of long-term safety, especially for spent nuclear fuel.

Rock melting is another variant of the geological disposal option. It is not seriously being considered by any waste disposal agency due to impracticality and the limitations of existing technology to develop a robust safety case. It is mentioned only for the sake of completeness. The radioactive waste would be emplaced at high enough concentrations, possibly in a super-dense waste package, so that radiogenic heat would be sufficient to cause partial or complete rock melting (Schneider & Platt 1974). After cooling of the rock, the wastes should be sealed or incorporated into the solid rock mass. Concerns with this variant, which has not been considered in any detail, include:

- the fate of important volatile radionuclides such as C-14 and I-129;
- the management of operational phase perturbations, such as possible contamination of interim depth geological formations;
- quality assurance of the emplaced waste;
- adequate sealing of the borehole;
- modelling of the hydrothermal consequences of high temperatures;
- requirements for site characterisation.

4.2.1 Geological disposal in crystalline rock

Different geological environments have been studied and are summarised for example by Bowden (2002). For crystalline rock, four different systems have been studied in Sweden: KBS-3, long tunnels, WP-Cave and deep boreholes.

Deep boreholes is a concept where spent nuclear fuel is deposited at a depth of several thousand metres. Here the barrier role of the geosphere is emphasised, since at depth it is claimed that groundwaters are completely stagnant and no engineered barriers, other than canisters (for waste handling and emplacement) and borehole seals, are needed. A benefit of the concept, compared with KBS-3, is that it offers better protection against human intrusion. Drawbacks include the operational risks involved in construction of the deep, large diameter boreholes and, particularly, deposition of the waste containers which cannot be carried out with the degree of control and assurance required with currently available technology.

In the long tunnels concept, spent nuclear fuel is deposited in a few parallel tunnels several kilometres long at a depth of 400–700 metres. This alternative has environmental advantages since the quantity of excavated rock is much smaller than in KBS-3 (although this partly arises from the use of fewer, larger disposal canisters). The drawback is that the safety challenges during operation are greater and also during retrieval of damaged canisters, should that be required.

In the WP-Cave concept, water flux in a deposition area is decreased by various engineering means, which create a hydraulic cage. The WP-Cave concept is technically complicated and it is difficult to show that long-term safety can be guaranteed. It would need more research and development to improve the technology and design, and to assess the safety.

In the KBS-3V design, the spent nuclear fuel is deposited in deposition holes in a system of short tunnels and, in the variant KBS-3H design, the spent nuclear fuel is deposited in 100–300 m long horizontal deposition drifts; both variants are at a depth of 400–700 metres. One of the main advantages of the KBS-3 repository is that it can be adapted to the bedrock, since the deposition tunnels and holes can be characterised in situ. It is currently believed that a KBS-3 repository can be built, operated and closed in a controlled manner. So far the evaluations and assessments carried out have shown that the KBS-3 method can satisfy the requirements of safety and radiation protection. The concept is discussed in more detailed in the next chapter.

4.2.2 Disposal options considered in Finland

In Finland the possibilities for spent nuclear fuel disposal were reviewed in the 1970s and '80s for example by Ryhänen et al. (1981), TVO (1982) and Raumolin (1982). The options considered were direct disposal of spent nuclear fuel and reprocessing the fuel followed by disposal of the reprocessing waste (Ryhänen et al. 1981). At that time, it was noted that the only available option was direct disposal, since there were no reprocessing plants on an industrial scale available to Finland. Nevertheless it was decided in 1982 to follow on-going international research and continue to evaluate other spent nuclear fuel management alternatives.

Raumolin (1979) mentioned that geological disposal was chosen and the Swedish KBS design was to be further considered in Finland. The same year, a safety analysis for the conceptual repository was carried out for both reprocessing waste and spent nuclear fuel (Puttonen et al. 1979). Those preliminary investigations indicated that, in both cases, the exposure rates would be expected to be lower than the natural background radiation

level. By 1982 the reprocessing option was dropped as direct disposal had been shown to be a feasible option, the safety analysis concentrated only on spent nuclear fuel and also utilised some Olkiluoto site-specific data (Anttila et al. 1982).

The basic principles for the waste management were stated by Raumolin (1982), these were; integrity, known technology, safety, correct timing and flexibility. Indeed, geological disposal was already then widely accepted as the best option for final disposal since the Swedish KBS study in 1978 (Raumolin 1982). The design would also remain reversible for decades (in practice, as long as the facility is in operation), which allows considerable flexibility.

5 THE KBS-3V DESIGN AND THE SAFETY CONCEPT

5.1 KBS-3 disposal method

The 2001 DiP states that disposal of spent fuel shall take place in a geological repository at the Olkiluoto site, developed according to the KBS-3 method. In the KBS-3 method, spent nuclear fuel encapsulated in water-tight and gas-tight sealed copper canisters with a mechanical-load-bearing insert is emplaced deep underground in a geological repository constructed in the bedrock. According to the DiP, the repository shall be located at minimum depth of 400 m. In Posiva's current repository design, the repository is constructed on a single level and the floor of the deposition tunnels is at a depth of between 400–450 m in the Olkiluoto bedrock.

Posiva's reference design in the construction license application is based on vertical emplacement of the spent nuclear fuel canisters (KBS-3V; Figure 5-1). Currently, an alternative horizontal emplacement concept (KBS-3H) is being jointly developed by SKB and Posiva (Figure 5-1).

5.2 Posiva's safety concept

Long-term safety in the KBS-3 method is based on the multiple barriers principle. Radioactive substances are *contained* in several barriers that support each other, yet are as independent from each other as possible so that the failure of one barrier will not endanger the overall functioning of the *isolation* of the radioactive substances from humans and their environment.

The safety concept, as depicted in Figure 5-2 is a conceptual description of how *safe disposal* of spent nuclear fuel is achieved using the KBS-3 method, taking account of the characteristics of the Olkiluoto site.

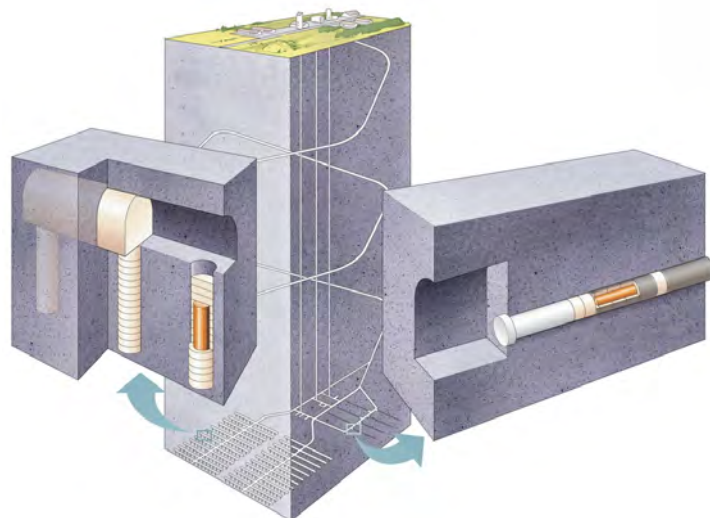


Figure 5-1. The KBS-3V (left) and KBS-3H (right) are alternative designs of the KBS-3 spent nuclear fuel disposal method. KBS-3V is the reference design, which is the main focus in this report.

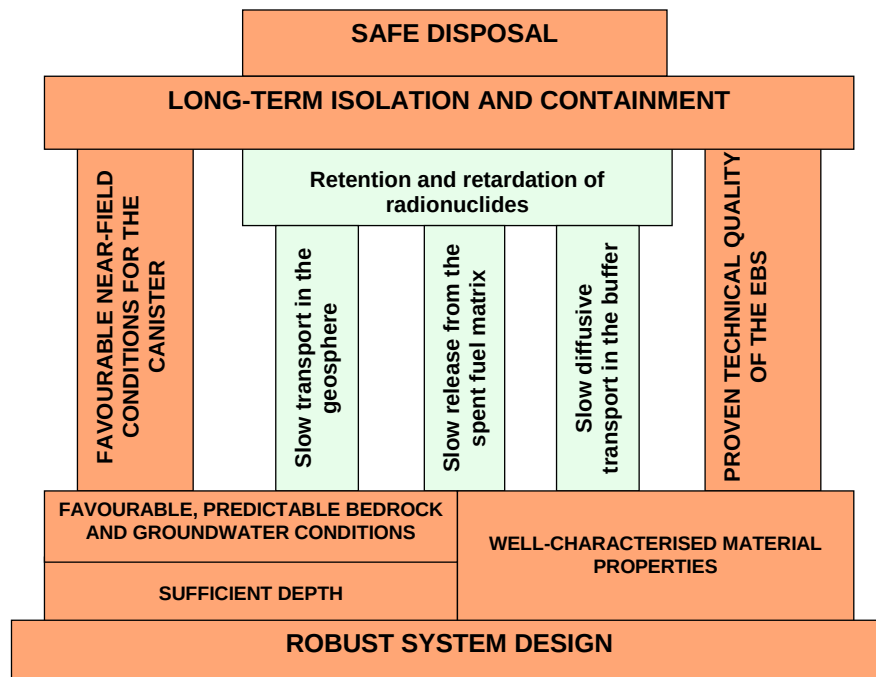


Figure 5-2. Outline of the safety concept for a KBS-3 type repository for spent nuclear fuel in a crystalline bedrock (adapted from Posiva 2003). Orange pillars and blocks indicate the primary safety features and properties of the disposal system. Green pillars and blocks indicate the secondary safety features that may become important in the event of a radionuclide release from a canister.

The features that favour long-term isolation and containment are shown as red pillars and blocks in Figure 5-2, which illustrates the safety concept for a KBS-3 type repository. The figure also shows, as green pillars and blocks, secondary features that ensure safety is maintained in the event that some releases of radionuclides from the canisters occur over time. These primary and secondary features of the safety concept are described in the following text (and indicated in *italics*).

A key element of the safety concept is the existence of multiple barriers so that it is unlikely that any single detrimental phenomenon or uncertainty could undermine the safety of the whole system, as required by Government Decree GD 736/2008:

“The long-term safety of disposal shall be based on redundant barriers so that deficiency in one of the barriers or a predictable geological change does not jeopardise long-term safety.”

The primary features and processes associated with the green columns in Figure 5-2 are, at least partly, independent from each other, consistent with the multibarrier concept. Safety functions assigned to the barriers in the KBS-3V design– the canister, the buffer, the backfill, the closure and the host rock – are described in Section 5.3 of this report.

In the KBS-3V design, isolation of the radionuclides associated with the spent nuclear fuel is provided by encapsulating the fuel in sealed gas and water-tight copper-iron

canisters. The likelihood of any radionuclide releases occurring from the canisters is kept small by an environment around the canisters that favours their longevity (*favourable near-field conditions for the canisters*) and by the *proven technical quality of the EBS (engineered barrier system)*. The EBS includes the canisters themselves and a surrounding clay buffer that protects the canisters from rock movements and from potential detrimental substances in the groundwater, the backfill (and plug) of the deposition tunnel that supports the buffer and the rock, and the closure components (backfill materials, plugs) in other parts of the disposal facility. The latter are designed to be compatible with, and support the safety functions of, the canister, the buffer, the backfill of the deposition tunnels and the host rock. For example, backfilling and sealing of the repository cavities (including tunnels, shafts and boreholes) support the safety functions of the host rock by giving mechanical support to the rock and preventing the formation of transport pathways (flow paths). They also contribute to discouraging inadvertent human intrusion into the repository. The surface environment does not have any barrier role and is thus not assigned any safety functions or target properties.

Other elements of a robust system design include *sufficient depth* for the repository, *favourable and predictable bedrock and groundwater conditions* and *well characterised material properties* of both the bedrock and the EBS. The characterisation of the Olkiluoto site and the strategy for repository design are focused on a volume of bedrock situated at depth of 400 – 450 metres. At such depths, the likelihood of inadvertent human intrusion is low and favourable and predictable bedrock and groundwater conditions, such as reducing conditions and slow movement of groundwater, are found.

Should any initially defective canisters be present or subsequent breaches in the canisters occur, the consequences of radionuclide releases for humans and other biota inhabiting the surface environment will be mitigated by the *slow release from the spent fuel matrix, slow diffusive transport in the buffer and backfill, and slow radionuclide transport in the geosphere*. Together, the engineered barriers and the rock provide for retention and retardation of radionuclides. Radioactive decay also continues during transport. These are depicted in Figure 5-2 as secondary features of the safety concept (green blocks and pillars) since they become important only in the event of canister failure.

5.3 The barriers and their safety functions

The safety concept indicates the roles that the barriers play in the disposal system achieving safe disposal. These roles are called safety functions. More detailed description of the safety functions and performance targets of the repository system can be found in the *Design Basis*.

The long-term safety barriers in the KBS-3V design are:

- the canister
- the buffer
- the backfill and plug in the deposition tunnel,
- the closure components, and
- the host rock.

The spent nuclear fuel matrix, while not a barrier specifically engineered for its performance in a repository, is nonetheless a very effective impediment to the release of many radionuclides. UO_2 was chosen as a reactor fuel because it is stable under intense irradiation, chemically durable compared to many other types of fuel and because many radionuclides remain in the fuel matrix. Furthermore, zirconium alloy cladding was selected because it is highly corrosion resistant. Although no safety functions are assigned to the spent nuclear fuel itself, its properties, such as low dissolution rate of the UO_2 fuel matrix, are taken into account in the assessment of the radionuclide release and transport calculations.

The safety functions of the EBS components and host rock are summarised in Table 5-1.

Table 5-1. *Safety functions assigned to the barriers (EBS components and host rock) in Posiva's KBS-3V repository design.*

Barrier	Safety functions
Canister	Ensure a prolonged period of containment of the spent nuclear fuel. This safety function rests first and foremost on the mechanical strength of the canister's cast iron insert and the corrosion resistance of the copper surrounding it.
Buffer	Contribute to mechanical, geochemical and hydrogeological conditions that are predictable and favourable to the canister, Protect canisters from external processes that could compromise the safety function of complete containment of the spent nuclear fuel and associated radionuclides, Limit and retard radionuclide releases in the event of canister failure.
Deposition tunnel backfill	Contribute to favourable and predictable mechanical, geochemical and hydrogeological conditions for the buffer and canisters, Limit and retard radionuclide releases in the possible event of canister failure, Contribute to the mechanical stability of the rock adjacent to the deposition tunnels.
Host rock	Isolate the spent nuclear fuel repository from the surface environment and normal habitats for humans, plants and animals and limit the possibility of human intrusion, and isolate the repository from changing conditions at the ground surface, Provide favourable and predictable mechanical, geochemical and hydrogeological conditions for the engineered barriers, Limit the transport and retard the migration of harmful substances that could be released from the repository.
Closure	Prevent the underground openings from compromising the long-term isolation of the repository from the surface environment and normal habitats for humans, plants and animals. Contribute to favourable and predictable geochemical and hydrogeological conditions for the other engineered barriers by preventing the formation of significant water conductive flow paths through the openings. Limit and retard inflow to and release of harmful substances from the repository.

6 THE DISPOSAL SYSTEM FOR SPENT NUCLEAR FUEL FROM OLKILUOTO AND LOVIISA NUCLEAR POWER PLANTS

The disposal system consists of the repository system components (host rock, spent nuclear fuel, canisters, buffer, deposition tunnel backfill and plugs, and closure components) and the surface environment (biosphere). These are described briefly in the following sections. The initial state of the whole disposal system is presented in detail in the *Description of the Disposal System* and references therein. The design basis and details for repository components are given in *Design Basis* and *Production Line* reports (*Canister-, Buffer-, Backfill-, Closure- and Underground Openings Production Line*).

6.1 The Olkiluoto site

The Olkiluoto Island (about 10 km²), which has been selected as the site for the spent nuclear fuel repository, is located on the Baltic Sea coast in the south-western Finland (Figure 1-1). The Olkiluoto site, as seen today, is the outcome of events and processes that have taken place over billions of years, from those reflected in the geological properties of the rocks forming the geosphere, to the much shorter-term changes related to climate-driven processes (mainly groundwater flow and groundwater composition) and the geomechanical effects of crustal isostatic rebound. A detailed description of the Olkiluoto Site is given in *Site Description* concentrating on the host rock and in *Biosphere Description* for the surface environment.

6.1.1 Geological setting and host rock

The crystalline bedrock of Finland is part of the Precambrian Fennoscandian Shield, which, in south-western Finland, consists mainly of Early Palaeoproterozoic metamorphic and igneous rocks belonging to the Svecofennian Domain. This domain developed between 1930 Ma and 1800 Ma ago, either during one long Svecofennian orogeny, or during several, separate orogenies. The rocks of Olkiluoto are divided into two major classes (Aaltonen et al. 2010a, see also Figure 6-1). The first are supracrustal high-grade metamorphic rocks including various migmatitic gneisses, tonalitic-granodioritic-granitic gneisses, mica gneisses, quartz gneisses, and mafic gneisses, and the second are igneous rocks including pegmatitic granites and diabase dykes. The metamorphic supracrustal rocks have been subjected to polyphase ductile deformation producing thrust-related folding, shearing, strong migmatization and pervasive foliation. The most important rock-forming minerals in the Olkiluoto rocks are quartz, potassium feldspar, plagioclase, biotite ($\pm$ other micas) and hornblende ($\pm$ other amphiboles).

The bedrock at Olkiluoto has been subjected to extensive hydrothermal alteration processes, which are estimated to have taken place at temperatures from slightly over 300 °C down to 50 °C (Gehör et al. 2002). Based on the grade of alteration, two different types of hydrothermal alteration can be distinguished: a fracture-controlled type and a pervasive (or disseminated) type. The fracture-controlled alteration indicates that hydrothermal fluids have passed through the rock along planar features, with the alteration being restricted to incipient fractures or narrow zones adjacent to them. The pervasive alteration shows the strongest changes – its effects occur as spots or finely disseminated throughout the rock and in the fracture fillings. The main hydrothermal alteration minerals in the Olkiluoto rocks are illite, kaolinite, sulphides and calcite. The

overall trend of alteration at Olkiluoto is the replacement of framework silicates by sheet silicates. Typically the episodic alteration has increased the concentrations of CO₂ and Ca in the whole rock composition; the altered rocks at Olkiluoto have also undergone K-metasomatism with K gain and Na+Ca loss.

The fault zones at Olkiluoto are mainly SE-dipping thrust faults formed during contraction in the latest stages of the Svecofennian orogeny, approximately 1800 Ma ago, and these were reactivated in several deformation phases. In addition, NE-SW striking strike-slip faults are also common (see Figure 6-1). The occurrence of fracturing varies between different rock domains, but the following three fracture sets are typical for the site (i) east-west striking fractures with generally subvertical dips to both the north and south, (ii) north-south striking fractures with generally subvertical dips to both the east and the west and (iii) moderately-dipping to gently-dipping fractures with strikes that are generally sub-parallel to the aggregate foliation directions in a particular fracture domain.

The strength and deformation properties of the intact rock, as well as its thermal properties, depend essentially on the mineral composition and structure, i.e. rock type. The rocks with higher quartz content have higher thermal conductivity and diffusivity than the ones consisting of mafic minerals. The heterogeneity of the rock properties at Olkiluoto is, therefore, reflected in the variation of the thermal and rock mechanics properties and seen e.g. in the anisotropic thermal properties due to foliation and gneissic banding. Currently the temperature at the repository level is 11–12 °C. The thermal conductivity of the rock at 25 °C varies from 2.49 to 3.20 W/(m·K), the average being 2.91 W/(m·K), depending on the rock type. The specific heat capacity at 25 °C is on average 712 J/(kg·K).

Olkiluoto is situated away from the active plate margins. In Fennoscandia, the orientation of the major principal stress is attributed to an E-W compression from the mid-Atlantic ridge push and a N-S compression from the Alpine margin, resulting in a roughly NW-SE orientation of major principal stress (Heidbach et al. 2008). This is also supported by the regional in situ data from Olkiluoto and other Finnish sites studied during the site selection programme. Changes in isostatic load due to glaciations and related isostatic adjustment and the existence of the brittle deformation zones change the stress regime at the site. Currently, a thrust faulting stress regime is present, i.e. the horizontal stresses are larger than the vertical stress, $\sigma_H > \sigma_h > \sigma_v$ and the principal stresses are approximately oriented horizontally and vertically, respectively. The orientation of σ_H at the site is found to vary slightly with depth and at the repository depth is in the range NW-SE to E-W. The vertical stress is predominantly well represented by the weight of the overlying rock mass.

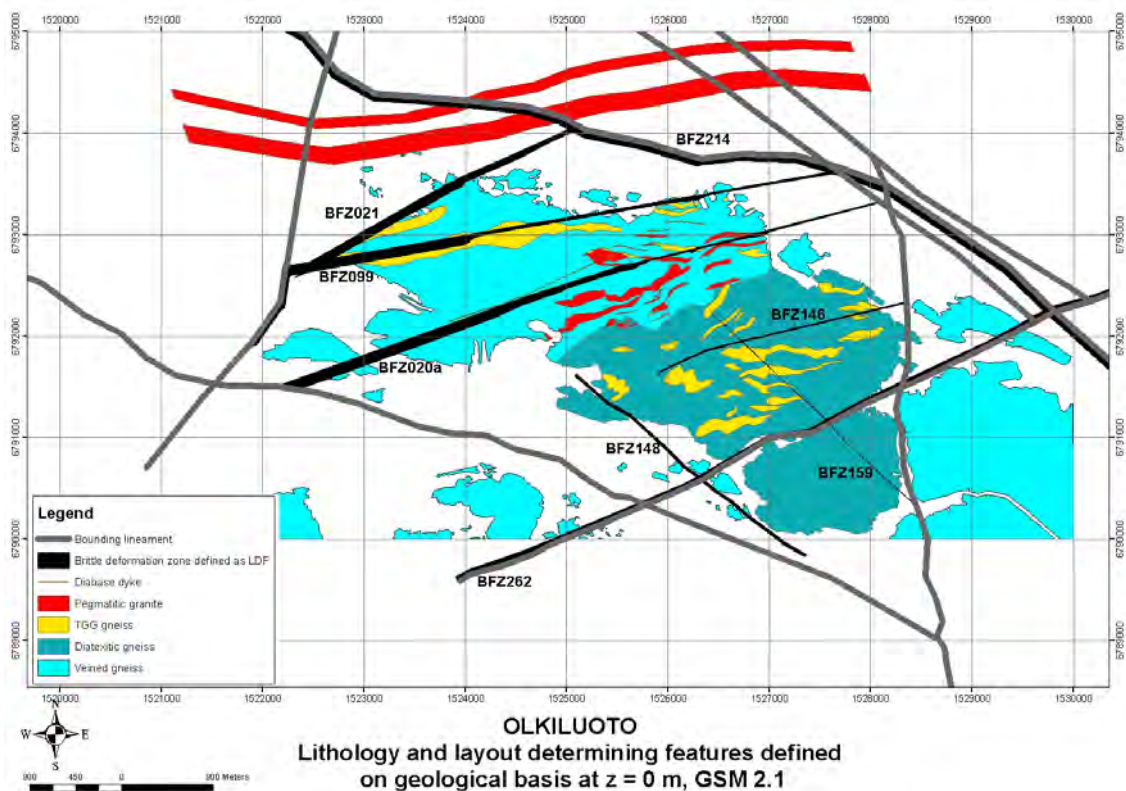


Figure 6-1. A surface geological map of Olkiluoto Island showing lithology and brittle deformation zones defined as layout determining features.

6.1.2 Hydrogeology and hydrogeochemistry

In the crystalline bedrock at Olkiluoto, groundwater flow takes place in hydraulically active deformation zones (hydrogeological zones) and fractures. The larger scale hydrogeological zones, which are related to brittle deformation zones, carry most of the volumetric water flow in the deep bedrock. There is a general decrease of transmissivity of both fractures and the hydrogeological zones with depth. Under natural conditions, groundwater flow at Olkiluoto occurs mainly as a response to freshwater infiltration dependent on the topography, although salinity (density) variation driven flow also takes place to a lesser extent. The porewater within the rock matrix is stagnant but exchanges solutes by diffusion with the flowing groundwater in the fractures.

The complexity of the fracture groundwater and matrix porewater chemistry reflects the history of the Olkiluoto area and the effects of periods of glaciation and associated infiltration of fresh meltwater, submersion below sea level and the influence of marine water, and the slow interaction between the groundwater, porewater and the minerals of the rocks over millions of years. Currently, the groundwater chemistry over the depth range 0–1000 m at Olkiluoto is characterised by a significant range in salinity and forms a relatively layered system (see Table 6-1). Fresh waters (salinity <1 g/L) rich in dissolved carbonate are found at shallow depths, in the uppermost tens of metres. Brackish groundwater, with salinity up to 10 g/L dominates at depths between 30 m and about 400 m. Brackish sulphate-rich waters are common in the depth layer 100–300 m,

whereas brackish chloride water, poor in sulphate dominates at depths of 300–400 m. Saline groundwaters (salinity >10 g/L) dominate at still greater depths.

The matrix porewaters are rather different as they are exchanged more slowly: in the upper part of the bedrock (0–150 m), the groundwater and matrix porewater seem to be in equilibrium, suggesting a similar origin and strong interaction between these two groundwaters. However, at deeper levels (150–500 m), the matrix porewater is less saline and increasingly enriched in $\delta^{18}\text{O}$ which has been interpreted to represent fresh water conditions during a warm climate, probably the preglacial Tertiary period (Posiva 2009).

The brackish and saline groundwaters below 300 m depth are assumed to be very old and water compositions are missing distinct features from the Quaternary glacial cycles that have occurred over the last approximately two million years. Shallower groundwaters, in the range down to 300 m depth have been affected by infiltrating waters of glacial, marine and meteoric origin during the alternating periods of glaciations and interglacials. However, only features from the latest Weichselian glaciation and postglacial period are observed, indicating the dynamic hydrogeological characteristics of this upper bedrock compared with the deeper bedrock. The current distribution of the groundwater types is the result of progressive mixing and reactions between various initial water types, which represent some of the major events at the site during its geological history. The initial water types have been recognised to be, from oldest to youngest, (i) brine, (ii) subglacial water, (iii) glacial meltwater, (iv) Littorina seawater, and (v) meteoric water (Site Description, Section 11.4.1). Present-day Baltic seawater, which is basically diluted Littorina seawater, is also recognised from shallow depths.

Present-day Baltic seawater, which is basically diluted Littorina seawater, is also recognised from shallow depths. Water-rock interactions, such as carbon and sulphur cycling and silicate reactions, buffer the pH and redox conditions and stabilise the groundwater chemistry (Table 6-1). In addition, weathering processes during infiltration play a major role in determining the shallow groundwater composition.

Redox conditions at Olkiluoto are anoxic except in shallow infiltrating groundwater at a few locations. Pyrite and other iron sulphides are common in water-conducting fractures throughout the investigated depth zone indicating a strong lithological buffer against oxic waters over geological time scales. Two natural metastable redox interfaces are present (Figure 6-2): the upper is located mainly in the overburden where the conditions change from oxic to anoxic. The other is located at a depth of approximately 250–350 m. In this zone, sulphate-rich groundwater is mixed with the methane-rich brackish-Cl – saline groundwater system to give rise, at least locally, to high levels of dissolved sulphide as a microbially-mediated reaction product.

Table 6-1. Groundwater types and their origin at Olkiluoto (in the water-conductive fracture system, baseline conditions), observed depth range, total dissolved solids (TDS), redox conditions and pH (according to Site Description, Chapter 7).

Groundwater types; dominant origin and mixing evidence in groundwater samples	Depth range (z), masl	TDS g/L	Redox conditions	pH
Fresh HCO ₃ ; Meteoric infiltration	+10 – -40	<1	Oxic to Anoxic	5.2–8.1
Brackish HCO ₃ ; Mixing of meteoric infiltration with Littorina ⁹ Sea derived SO ₄ -rich brackish groundwater	0 – -130	1–3	Sulphidic	7.6–8.1
Brackish SO ₄ ; Littorina seawater and glacial meltwater mixed with ancient meteoric water-saline groundwater mixture	-60 – -300	4–9	Sulphidic	7.1–8.1
Brackish Cl; Ancient meteoric water - saline groundwater mixture with minor Littorina seawater component	-100 – -400	2–10	Sulphidic to Methanic	7.3–8.8
Saline Na-Ca-Cl; Ancient meteoric water - saline groundwater mixture	-320 – -480	10–18	Methanic	7.3–8.2
Saline Ca-Na-Cl; Ancient saline groundwater - meteoric water mixture	-410 – -570	18–30	Methanic	7.6–8.4
Highly saline Ca-Na-Cl; Ancient brine ¹⁰ - meteoric water mixture	below -570	>30	Methanic	7.0–8.1

⁹ During the end of the Weichselian glaciation the Baltic Sea developped to ist current stage via alternating lacustrine and marine stages that were Baltic Ice Lake (until 11 590 BP), Yoldia Sea (11 590 – 10 800 BP), Ancylus Ice Lake (10 800 – 9000 BP), Mastogloia Sea (9000 – 8000 BP), Littorina Sea (8000 – 3000 BP) and Baltic Sea (3000 BP– present). Olkiluoto was free of ice cover around 11,000 BP and emerged from Baltic sea around 3000 BP. (Eronen et al. 1995)

¹⁰ TDS in brine water is more than 100 g/L.

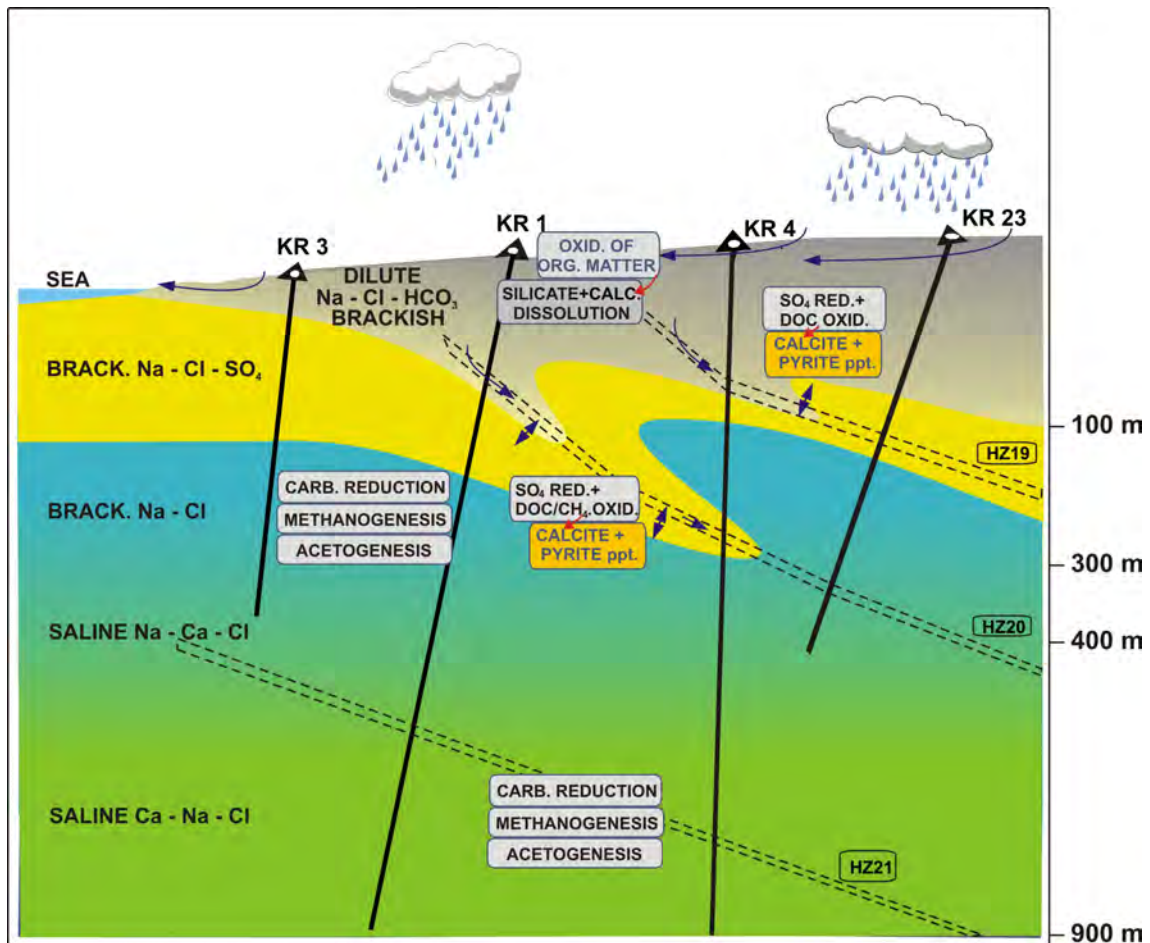


Figure 6-2. Illustrative hydrogeochemical site model of baseline groundwater conditions with the main water-rock interactions at Olkiluoto. Changes in colour indicate alterations in water type. The hydrogeologically most significant zones are represented. Blue arrows represent flow directions. Rounded rectangles contain the main sources and sinks affecting pH and redox conditions. Enhanced chemical reactions dominate the infiltration zone at shallow depths, and at the interface between Na-Cl-SO₄ and Na-Cl groundwater types. Note that the illustration depicts hydrogeochemical conditions in the water-conductive fracture system, not in the diffusion-dominated rock matrix (Site Description).

6.1.3 Surface environment

Topography in the Olkiluoto area, and in general in South-Western Finland, is flat and soil erosion rates are very low. Glacial erosion features such as glacially smoothed bedrock outcrops and *roches moutonnées* are common. As a result of the last glaciation, the bedrock depressions are filled with a thicker layer of overburden, mainly sandy till and fine-grained till (Huhta 2005, 2007, 2008, 2009, 2010, Lintinen et al. 2003, Lintinen & Kahelin 2003, Lahdenperä et al. 2005, Lahdenperä 2009).

The waters around Olkiluoto Island are shallow, except for a few areas where sea depths reach about 15 m. The seabed west of Olkiluoto is mainly till (about 35–45 %), bedrock (about 30–40 %) and various clay and mud deposits (about 15–25 %). The sea area

surroundings of Olkiluoto have an extensive area of gaseous sediments (Rantataro & Kaskela 2010). This is of particular interest because of the continued land uplift of the Olkiluoto area which will expose areas of current seabed sediments in the next few thousand years – the time frame for which the spent nuclear fuel repository releases must be quantified in terms of doses to a human population and other biota. In addition, the effects of land uplift are accentuated by eutrophication in the coastal areas, especially in shallow bays (Haapanen & Lahdenperä 2011). The ecosystem succession during uplift, the redistribution of sediments and groundwater flow will influence the areas of potential deep groundwater recharge and discharge from the repository (Haapanen et al. 2007, 2009). The changes in the landscape are, however, affected not only by uplift but also by changes in sea level that are related to changes in climate, both regionally and globally. The effect of uplift could be, at least to some extent, enhanced or reversed by changes in sea level. This is taken into account in the climate scenarios in *Biosphere Assessment*.

At present, freshwater (limnic) ecosystems are few in the Olkiluoto area and there are no natural lakes on the island. The nearby lake basins were isolated during the various stages of development of the Baltic Sea as a result of isostatic uplift and tilting of the land. The closest rivers are Eurajoki and Lapijoki (Lapinjoki), which discharge to the sea north and east of Olkiluoto, increasing the concentration of nutrients and solids, especially at the river mouths (Haapanen et al. 2009). The use of lakes and mires – currently absent from the island – in the surrounding region as analogues for future biosphere conditions at Olkiluoto is discussed by Haapanen et al. (2010). Figure 6-3 shows land uplift and an example of surface environment development through to 10,000 years from the present. The analogues for future biosphere evolutions at Olkiluoto are further discussed in Chapter 10.

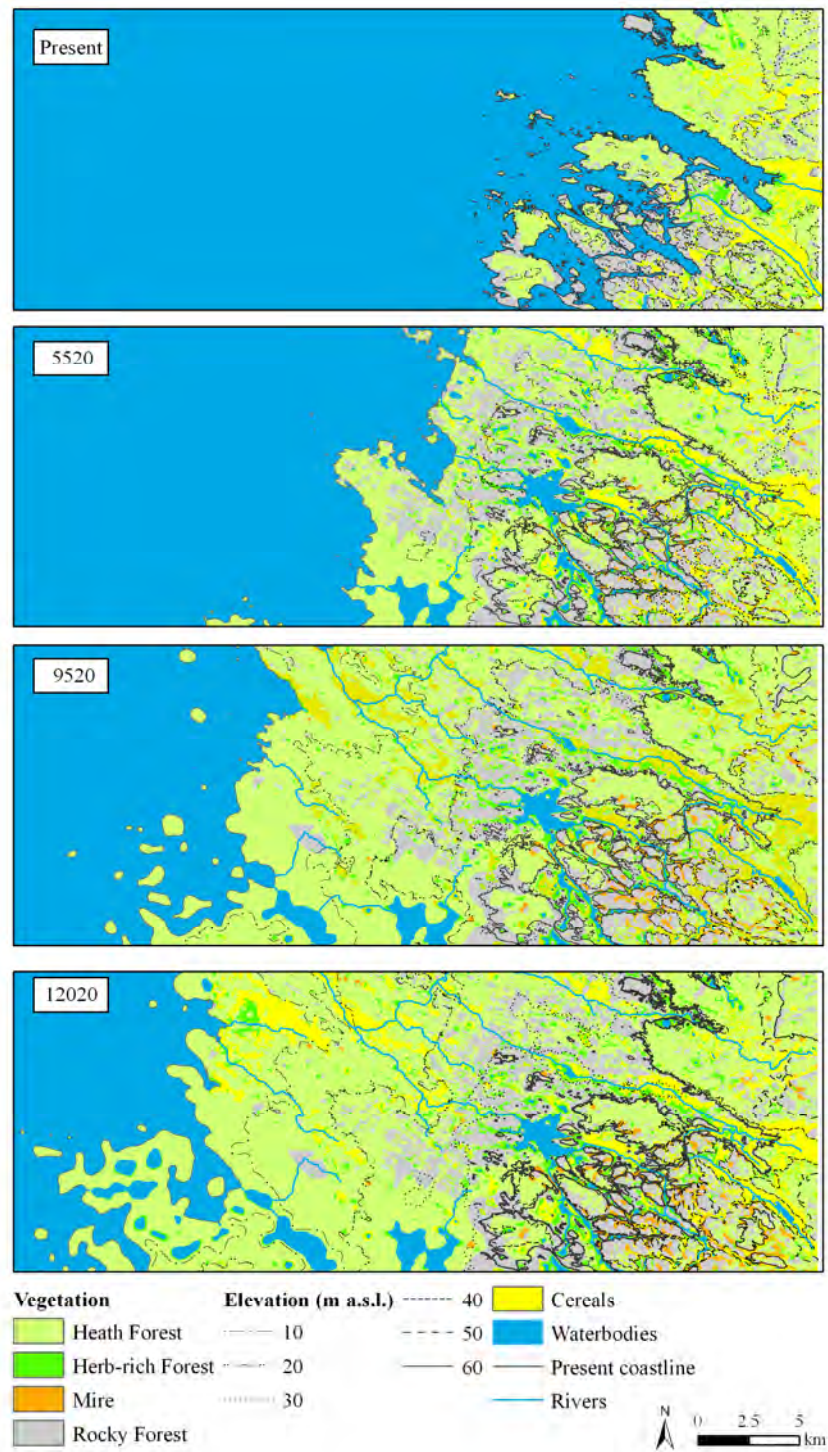


Figure 6-3. Land uplift and an example of the biosphere development through to 10,000 a after the present. Map data: Topographic database by the National Land Survey of Finland (permission 41/MYY/11) and Posiva Oy. Map layout by Jani Helin/Posiva Oy. Note: dates are given as AD, i.e. 12020 is 10,000 years after the reference date of 2020 AD.

6.2 Spent nuclear fuel

The geometry of the spent nuclear fuel elements depends mainly on the reactor type. Small differences may also arise depending on the manufacturer of the fuel element. The main features of the fuel elements for the LO1-2, OL1-2 and OL3 reactors are shown in Figure 6-4. The OL1-2 (BWR), LO1-2 (VVER) and OL3 spent nuclear fuel pellets are, from a chemical point of view, made of the same sintered material, UO_2 . However, the UO_2 pellet geometry, U-235 content and burnable poison (absorber) proportion, and cladding material, as well as other components of the assembly, are different depending on the reactor type. The average U-235 enrichment of a fuel assembly may vary roughly between 3–4 %, but within a single fuel rod/pellet could be nearly 5 %. In the future, the assembly average enrichment rate could be over 4 %, enabling higher burn-ups to be achieved. Fuel cladding is usually made of various types of zirconium alloys because the zirconium cross-section for thermal neutrons is very small and because zirconium alloys typically have great mechanical strength and good corrosion resistance. The other structural elements of the fuel assemblies (i.e. upper and lower tie-plates, end plug, spacer grid, and channel and nose piece) are fabricated from stainless steel, zirconium alloy or nickel-based alloys. For more details see *Description of the Disposal System* and references therein.



Figure 6-4. Representative illustrations (from left) VVER-440 (LO1-2), BWR (OL1-2) and OL3 type fuel assemblies. BWR and VVER-440 fuel elements are partly cut open to show the internal structures. The pictures are not to scale.

6.3 Canister

The canister structure consists of a massive cast iron insert covered by a 5 cm-thick copper overpack. Copper has been chosen as the overpack material because of its well-known characteristics, including its good thermal and mechanical properties, and its resistance to corrosion in reducing environments. Cast iron has been chosen for the insert to provide mechanical strength, radiation shielding and to maintain the fuel assemblies in the required configuration. The design and dimensioning analyses of the copper-iron canister for spent nuclear fuel are presented in Raiko (2012) as well as in the *Canister Production Line report*. There are three versions of the canister, one for each spent nuclear fuel type (Figure 6-5). The spent nuclear fuel is sealed in the canisters as whole fuel assemblies including the flow channel outside the VVER-440 and BWR fuel assemblies.

The canisters are designed to maintain their integrity for at least hundreds of thousands of years taking into account the processes and events that are considered likely to take place in the repository over this period. It is likely that canister integrity will be retained significantly beyond the design basis period, although extreme conditions may give rise to earlier canister failures. The performance of the canister over the design basis period is analysed in *Performance Assessment*. Early failure scenarios are considered in radionuclide release and transport calculations (*Assessment of Radionuclide Release Scenarios for the Repository System*).



Figure 6-5. Copper-iron canisters for the spent nuclear fuel from the LO1-2 , OL1-2 and OL3 reactors from left to right (Raiko 2005). All versions of the canister have the same outer diameter of 1.05 m. The height ranges from 3.6 to 5.2 m.

6.4 Buffer in the deposition hole

The canister will be surrounded by rings and blocks (on the top and bottom) of highly compacted bentonite, referred to as the “buffer” (Figure 6-6). The uppermost metres of the deposition hole will be backfilled with blocks of compacted bentonite and tunnel backfill (*Buffer- and Backfill Production Line*). The installation gap between the canister and buffer is approximately 10 mm and that between the buffer and the rock is approximately 50 mm. Initially, the inner gap will be filled with air and the outer gap with bentonite pellets (*Buffer Production Line*).

A high grade sodium bentonite from Wyoming, USA (commercial name MX-80) is the reference buffer material in the design (*Design Basis*). The initial state of the buffer is discussed in *Description of the Disposal System*.

6.5 Deposition tunnel backfill and plugs

The deposition tunnel leading to the deposition holes will be backfilled before closure to limit the hydraulic conductivity of the tunnels, prevent the loss of buffer density and provide mechanical stability to the tunnels. The current backfilling method is based on the block concept, involving filling most of the tunnel with pre-compacted backfill blocks and the remaining volume with bentonite pellets (Figure 6-7) (*Backfill Production Line*). The reference backfill block material is Friedland clay.

The deposition tunnels are closed by using a deposition tunnel plug, which is composed of concrete and natural filler materials (such as clay, sand and gravel) as presented in schematic Figure 6-7. The deposition tunnels will be plugged after the backfilling is completed to avoid significant water inflows. The plug also helps to keep the swelling backfill in place before the closure of the access tunnel. Low pH concrete (i.e. a concrete for which the leachate has a pH < 10 after an initial short period of pH > 11) will be used in the end plugs to minimise chemical interaction.

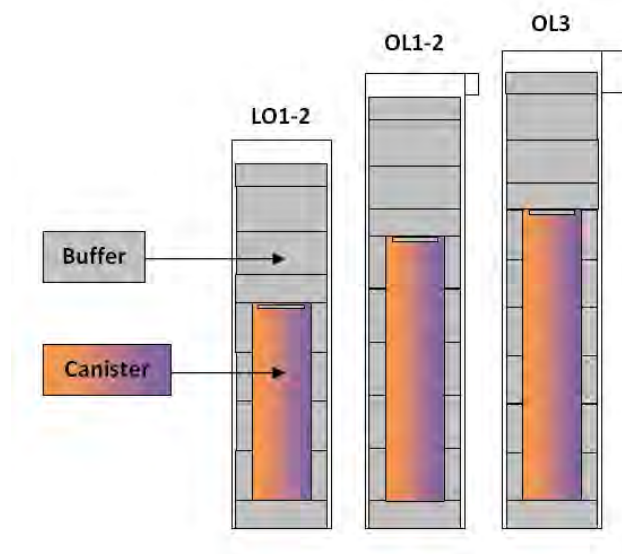


Figure 6-6. Schematic presentation of the buffer installation for the different canister types in deposition holes (modified from the Buffer Production Line report).

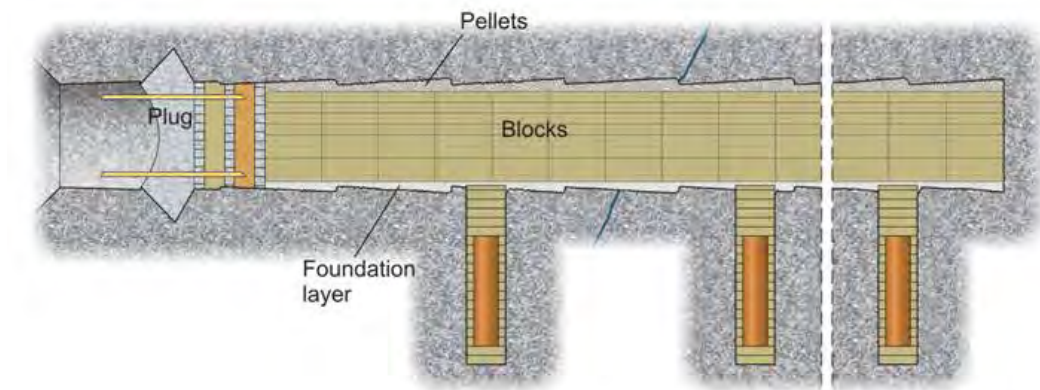


Figure 6-7. A schematic figure showing the main backfill components (*Backfill Production Line*).

6.6 Closure components

Closure will complete the isolation of the waste, restore and maintain favourable natural conditions in the bedrock, and prevent the formation of preferential flow paths and transport routes between ground surface and deposition tunnels and holes.

Closure of the repository covers all backfilling and plugs outside the deposition tunnels. The reference design for the closure of the repository is presented in Figure 6-8. The detailed description of the reference design, the backfill materials and methods, as well as the principles of the hydraulic, mechanical and intrusion-obstructing plugs are given in *Closure Production Line*.

The reference design for closure is such that there is a flexible tool-box approach available, providing alternative solutions throughout the closure process for the emplacement of backfill and plugs in a manner that meets the requirements set. Natural materials are utilised in backfill (such as clays, aggregates and mixtures of these). Hydrogeological zones (HZ) cut the repository volume (see Section 6.1.2 and Figure 6-2). In plugs, at least below structure HZ20 (a major hydrogeological zone lying above the repository which forms a hydraulic discontinuity – see Figure 6-2), low pH concrete will be used.

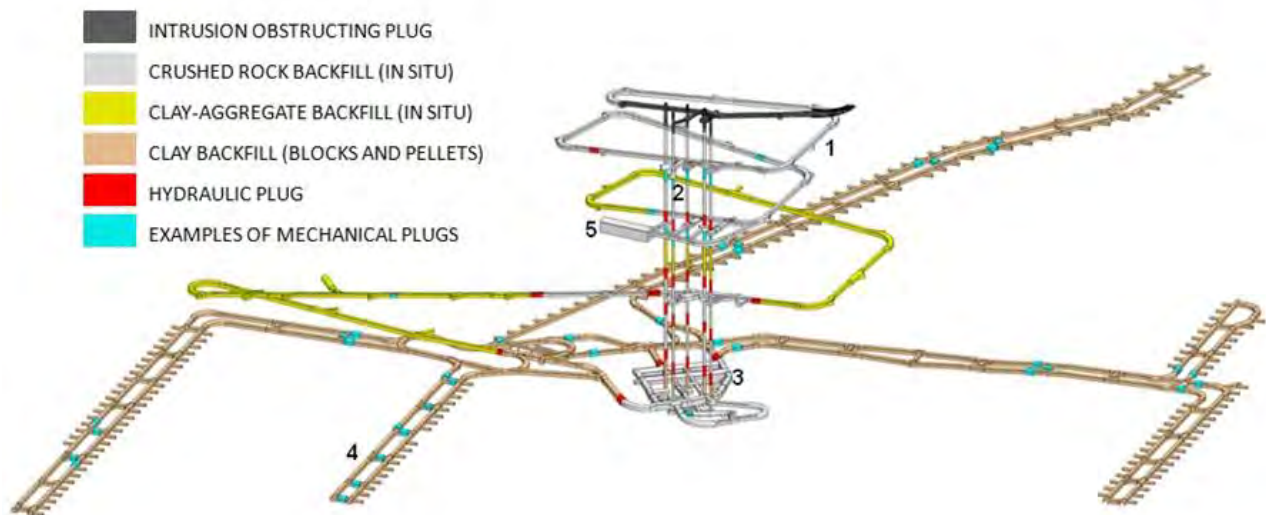


Figure 6-8. Reference design for closure showing the access tunnel (1) and shafts (2), technical rooms at the repository level (3) and the central tunnels (4) leading from the technical rooms to deposition tunnels, location of the L/ILW repository is also shown (5) (Closure Production Line).

6.7 Repository layout

The repository layout adaptation plans for 9000 tU (corresponding to 4500 canisters) at a depth of 420 m have been used in the TURVA-2012 Safety Case (Figure 6-9). This includes the spent nuclear fuel generated from the currently operating nuclear power plants as well as OL3, which is under construction, and OL4 (at the planning stage). The repository has been designed to host 20 % more canisters than the actual number, allowing a certain rejection rate for deposition holes (there are a total of 5400 deposition hole locations). In short, the available repository volume is primarily restricted by the layout-determining features: hydrogeological zones (HZ) and brittle fault zones (BFZ) and other lineaments identified in site investigations. No deposition tunnels or shafts are suggested that penetrate the layout-determining features, or the respect volumes set for them (Hellä et al. 2009). Respect volumes for a given feature are defined by the respect distances, which should be large enough to ensure a safe environment for disposal, taking into account the uncertainties in location and properties of the layout-determining features. The respect volumes will, however, be optimised so that good rock volumes are not unnecessarily lost. Deposition tunnels will be at least 25 m apart. More detailed discussions on the layout and its adaptation can be found in *Underground Openings Production Line, Description of the Disposal System* and in McEwen et al. (2012).

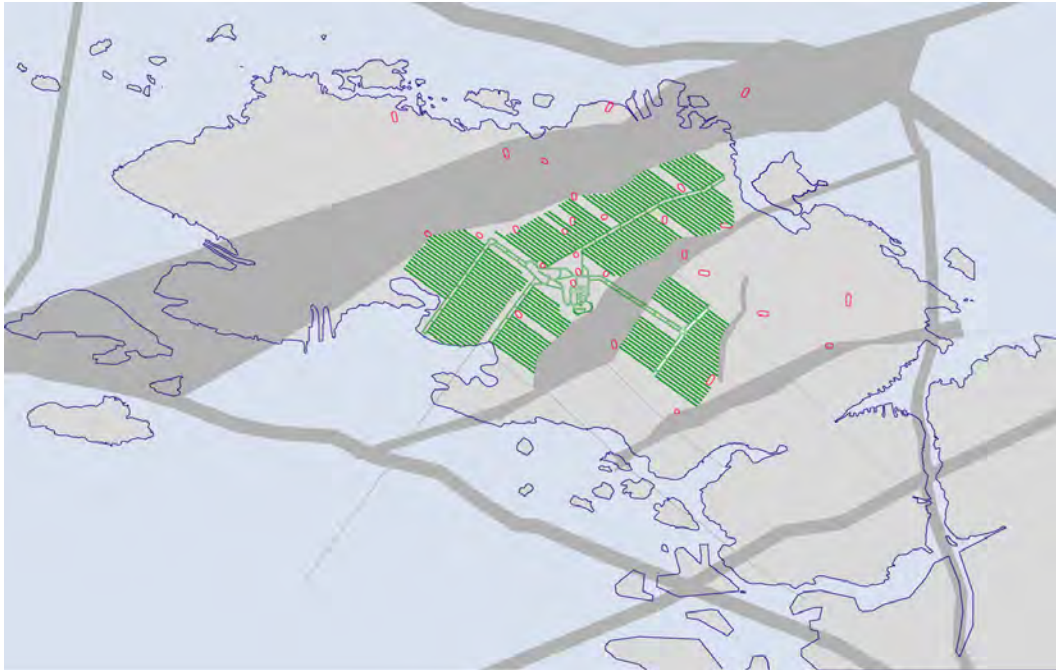


Figure 6-9. Layout adaption for a repository hosting 9000 tU of spent nuclear fuel used in the TURVA-2012 safety case, dark grey areas are not suitable for deposition tunnels according to the Rock Suitability Classification (RSC) as they are intersected by the layout determining features (LDFs) and their respect volumes. Red ovals denote respect distances to drillholes (Saanio et al. 2012).

7 EVIDENCE FOR THE STABILITY OF THE HOST ROCK CONDITIONS

As discussed in Section 7.1, the properties of the site are favourable and support the long-term safety of the repository. The most important source of confidence in site suitability is the overall site understanding and the predictability of the site behaviour in the future. This chapter discusses the topics of interest in reducing inherent uncertainties in the system performance during its long-term evolution by highlighting relevant processes that have occurred through the geological history of the site and by means of regional comparisons when applicable. Evidence for site suitability is presented, including also discussion of those properties that may not be favourable: by providing arguments as to their significance, it can be shown that they do not undermine site suitability. Further, complementary discussion of the stability and impact of the underground facility is presented as well as discussion of climate-driven processes, such as permafrost and introduction of dilute waters into deep rock formations, external events including earthquakes and meteorite impacts, and processes occurring over long timescales such as erosion.

7.1 On the suitability of the Olkiluoto site

The Olkiluoto site was selected based on identification of suitable geological units in the crust. The identification was founded on the concept of the “block model”, also called “mosaic structure”, introduced and discussed in many publications (see e.g. Tuominen et al. 1973; Vuorela & Hakkarainen 1982; Vuorela & Paulamäki 1988; Chapter 2 in Kuivamäki et al. 1998; see also Figure 7-1). The Finnish crystalline bedrock is a well-characterised system in which uplift-related tectonic movements take place in the existing fracture systems surrounding relatively intact bedrock blocks. This geological property forms a basis for site stability, as deposition hole positions can be chosen within given areas of bedrock surrounded by known geological structures along which possible tectonic movements will cause reactivation in cases of stress release (see McEwen et al. 2012).

During the detailed site selection process, four candidate sites were considered suitable and the final decision was made by taking into account other relevant factors such as spent nuclear fuel transportation, public acceptance etc. A summary of the site selection process is presented by McEwen & Äikäs (2000). Olkiluoto was selected as the site for the repository in 2000 (Decision-in-Principle by the Government 2000, ratified by the Parliament in 2001). Since then detailed studies at the Olkiluoto site, including construction of an underground research facility, have been going on. The results of the site characterisation activities are reported in the series of Olkiluoto Site Descriptions (latest one is *Site Description*). The continued studies have confirmed the overall suitability of the site. However, it has been recognised that long-term safety can be enhanced by locating the deposition tunnels and holes favourably with respect to Olkiluoto rock properties (see also Section 6.7). Therefore, the Rock Suitability Classification (RSC) system (Hellä et al. 2009; McEwen et al. 2012) has been established to provide practical tools for the host rock acceptance procedure during construction of the repository.

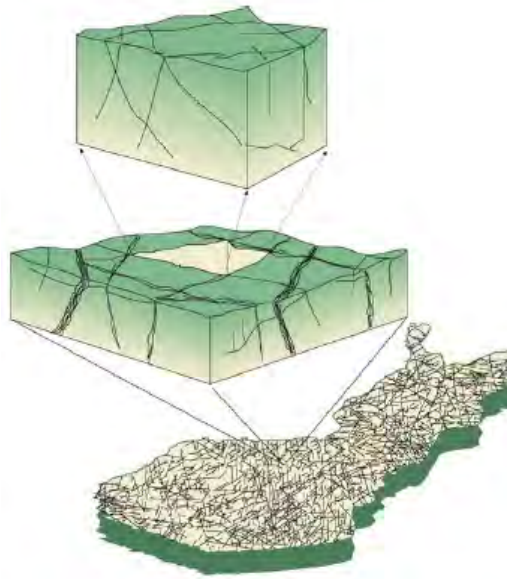


Figure 7-1. Block model concept (modified from Posiva Oy). In Finnish bedrock tectonic movements take place in the existing fracture systems surrounding relatively intact bedrock blocks. Major fracture zones bound large blocks within which the repository can be located – these are said to be the layout-determining features. Within these blocks, the bedrock may be subdivided by more minor fracture zones and individual fractures that can be avoided by placement of tunnels and deposition hole locations, depending on their size and hydraulic properties (see Section 6.7).

The Olkiluoto site is being affected by the construction and operations during the current study phase in ONKALO and will also be perturbed during the operational phase that is planned to continue until 2120, ending with the closure of the system. *Underground Openings Production Line* describes the effects of construction, which are evaluated in *Performance Assessment*. The initial state of the whole repository system, including the site, is given in *Description of the Disposal System*.

The suitability of the site has been discussed at length in *Site Description* and RSC reports (Hellä et al. 2009; McEwen et al. 2012). The following conclusions have been drawn:

- The Olkiluoto site is located in a geologically stable region with low tectonic activity, low relief and low erosion rates as well as a fracture system that provides plasticity to the bedrock;
- Even though there is a limited degree of EDZ formation and spalling, their impact on groundwater flow in the long term remains small;
- By using modern backfilling methods, the stability of the underground openings can be enhanced;
- There is a lack of valuable natural resources and future mining activities in the area are very unlikely;
- The groundwater flow in the repository volume is low and limited to a set of hydrogeological zones and to a sparse fracture network between these HZs thereby limiting the mobility of solutes or potentially released radionuclides;

- The hydrogeochemical system is reducing and has also other favourable chemical characteristics with respect to the performance of the engineered barrier system. The rock has substantial buffering capacity so that the favourable conditions will prevail also in future.

7.2 Mechanical stability of underground spaces

In *Features, Events and Processes*, Stress Redistribution (FEP 8.2.2) is described thus: *when an excavation is made in the rock, the stresses will be locally realigned to become parallel and perpendicular to the excavation surfaces, causing corresponding changes in the magnitude of the stresses* (Andersson et al. 2007, p. 96). Stress redistribution will occur almost immediately after excavation but could subsequently be affected by other tunnel excavations nearby. Small changes in the stress distributions are possible during the entire programme of repository excavation and operations (some decades).

Maintaining mechanical stability of underground spaces is a common issue faced in the mining and construction industries. The stability of underground excavations is affected by in situ stress, rock properties, opening spans and time since excavation (Potvin et al. 2005). The use of careful excavation techniques and rock support measures can minimise the effects on rock stability. Rock mass spalling and convergence can be further reduced by installing backfill that supports the rock.

In the mining industry, a method of “cut and fill” is widely used for increasing stability of underground openings. Long-term mining operations (decades in this context) have benefitted from backfilling techniques as it has been observed that backfilling provides regional stability for the bedrock (Landriault 2001; Williams et al. 2007). In Finland, backfilling of mines is a common procedure and has been conducted, for example, in the Pyhäsalmi and Vihanti mines. In the Pyhäsalmi mine, the excavation of the ore is carried out by blasting stopes in a pyramid shape leaving practically no pillars in the cavity. The method allows extraction of the ore almost completely from a large ovoid underground space at depths below 1000 m. Stress states are high and excavated areas are backfilled with consolidated rockfill. Backfilling allows the mine to operate under high rock stresses, which are redistributed after each stope has been excavated. Backfill has a high impact on the bedrock stability, limiting the occurrence of spalling and rock burst events. In the Olkiluoto repository, backfilling is somewhat different, especially with respect to development of swelling pressure by the backfill, a phenomenon absent in the mine backfills. During closure of some parts of the repository, other types of backfills may also be used that resemble more closely, in their mechanical properties, those used in mines. Swelling pressure provides an additional counterforce on the host rock, providing even better support for the host rock than is seen in the mines.

In the layout design, the stress state of the host rock is taken into account. The stress state at the Olkiluoto site has been determined for different depths using a wide variety of methods (see Posiva 2009, p. 534). According to the repository layout principles (e.g. Kirkkomäki 2009), the repository drifts will be aligned as far as possible with the direction of the maximal horizontal stress for reasons of mechanical stability.

Monitoring results are available from two shallow underground sites in Finland. Two decades of monitoring have been conducted at the two underground repositories for

operating waste from nuclear power plants in Finland (Öhberg et al. 2011). Teollisuuden Voima Oy (TVO) and Fortum Power and Heat company (Fortum) have constructed their facilities in tonalitic (Olkiluoto) and rapakivi granite (Hästholmen in Loviisa) formations, respectively. TVO's silos are located at a depth of 60–100 m below ground surface, whereas Fortum has the main underground openings at a depth of 100–120 m below ground surface. Rock stability at Olkiluoto has remained good and 20 years of monitoring has indicated only small deformations that are mostly due to operational temperatures (ventilation) of the rock caverns. In Hästholmen, the rock displacements have been of the order of 0.8 mm or less. For comparison, it can be noted that these repositories are in operation and not backfilled as the deep repository will be after closure and thus are providing information for the expected mechanical behaviour of the rock in the operational phase when parts of the repository are open.

7.3 Climate-change-driven processes

The Earth's climate system is a non-linear coupled dynamic system. Its long-term evolution is driven by external forcing factors but it also has a capacity for rich internal variation. The governing processes, and especially the role of the carbon cycle, are not yet fully understood. Nonetheless, climate will change in the future as it has done in the past based, at the very least, on external factors such as variations in the Earth's orbit and solar activity (see, for example, the summary in Pimenoff et al. 2011).

Whereas the continuation of a climate like today's, already affected by the emission of greenhouse gases, has no substantial effect on the safety of the repository (i.e. no climate-change-driven processes are expected), colder conditions in the very long term will give rise to new processes, hence the emphasis on climate change. The key issue for the post-closure safety of a repository is when the climate will shift from the boreal climate that currently prevails at Olkiluoto, and in Finland in general, to a much colder climate that may give rise to the development of permafrost and/or ice sheets. The presence of permafrost in the Olkiluoto region would have a significant impact not just on the surface environment but also on the hydrogeological and hydrogeochemical conditions in the subsurface. The presence of a thick ice sheet would have even more notable consequences in terms of also changing the topography after the ice sheet retreated – reversing the present post-glacial rebound by depressing Olkiluoto some hundreds of metres below the present-day altitude and thus possibly beneath future sea levels for some periods of time (see also Section 7.4).

7.3.1 Permafrost

The timing of the onset of permafrost and/or ice sheets has been estimated using models calibrated based on knowledge of past climate changes (e.g. Pimenoff et al. 2011). According to the most recent modelling exercises (see Table 3 in Pimenoff et al. 2011) the next cold period will probably start at around 50,000 years AP, although this is very sensitive to the level of atmospheric CO₂ assumed in the modelling. But it is also expected that there will be a succession of cold periods and glaciations over the assessment period of 1 Ma (Pimenoff et al. 2011). It is important to note that the changes between cold and more temperate periods mean that the Olkiluoto area could be subject to a number of different climatic environments. Pimenoff et al. (2011) note “...on a time-scale of 120,000 years, one must take into account climate periods lasting

several thousand years having the following features: an interglacial climate, a periglacial climate, a climate with an ice sheet margin near Olkiluoto, a glacial climate with an ice sheet covering Olkiluoto, and a climate with Olkiluoto being depressed below sea level after glaciation due to isostatic depression". Thus fluctuating conditions, as well as lengthy periods of cold climate, mean that the potential impacts associated with freeze-thaw processes and permafrost development must be assessed in order to ensure that the disposal system will function as designed.

The major issues to be addressed in relation to permafrost development are 1) if it will reach repository depth, and 2) if it does, what would be the consequences for the spent nuclear fuel, the EBS, and the host rock (including groundwater). Besides the permafrost impact, there is the additional question of whether repeated cycles of freezing could amplify any impacts caused by a single episode of freezing.

Permafrost and freeze-thaw processes have been assessed in detail in *Performance Assessment and Features, Events and Processes* and the conclusions are discussed briefly here. Also in connection with climate changes, groundwater flow in the geosphere may vary so as to drive dilute or saline waters into the disposal system and this is discussed in Section 7.3.2. Groundwater flow at Olkiluoto is specifically treated in *Site Description*; changes in salinity arising from climate evolution are dealt with in Löfman & Karvonen (2012).

Permafrost is defined as ground (soil or rock) that remains at or below 0 °C for at least two consecutive years (Williams & Smith 1989). This means that liquid water may be present within the permafrost as, depending on the depth (increasing hydrostatic pressure), adsorptive and capillary properties of the ground and, especially, groundwater composition, the porewater may not reach its freezing point (Hartikainen 2006, 2012). Furthermore, as noted by Hartikainen (2006, 2012), ground freezing is a gradual process over a temperature range in which water in the ground can exist in both solid and liquid phases. Permafrost hence consists of a partially frozen ground layer surrounded by an unfrozen ground layer.

The depth to which permafrost extends, and the temperatures within that zone are a function of the balance between the mean annual air temperature at the ground surface and the geothermal heat generation within the rock which creates a permanent heat flow from depth to the surface. Although there is evidence of the periglacial conditions prevailing during Weichselian (e.g. ice wedge casts, indicative of the active layer, see below), there are no physical indicators remaining of the maximum depth of the Weichselian permafrost in the Finnish bedrock, as stated previously in Pastina & Hellä (2006).

On top of a permafrost layer, there is an active layer which may be of centimetre to metre scale. In this layer, repeated freezing and thawing occur during the annual seasonal cycle. During the summer melting period, parts of an active layer will be oversaturated with liquid water, commonly giving rise to bogs and small ponds, while for the rest of the year the ground is frozen and mainly impervious to groundwater infiltration (Vidstrand 2003). It is this active layer which could be responsible for damage to surface sealing and closure structures as water percolates into cracks in the concrete components where the annual freezing and expansion would gradually degrade

the integrity of the plugs and seals.

The issue of the development of a saline front below permafrost, which could potentially influence repository performance and which is thought to arise by preferential exclusion of salinity as the groundwater freezes, has been recently discussed in Stotler et al. (2011). It has been convincingly shown that the saline waters in terrains affected by permafrost in the past (Canadian and Fennoscandian Shield) were not formed through a freezing process. Even if saline pockets could form during freezing, the amount of brine thus formed is small and not accumulated over repeated glacial cycles because of mixing processes in between the glacial cycles.

The present temperature of the bedrock at Olkiluoto at a depth of 100 to 200 m is 6–6.5 °C, at 400 m it is 10.5 °C, and at 500 m it is 12–13 °C (e.g. Anttila et al. 1999 and references therein). A permafrost layer reaching down to the repository at around -500 m (assuming no heating effect of the disposed canisters) would require a fall in the temperature of about 12 °C from the present day. Moreover, a period of thousands of years would be required to cool the sub-surface to that temperature. This would imply a mean annual air temperature of -6.2 °C, corresponding currently to the temperatures in central Alaska compared to the present day 5.4 °C at Olkiluoto (Cedercreutz 2004).

For the Olkiluoto area, there are no direct lines of evidence to show that permafrost did not reach repository depth in past glacial periods but there is some indirect evidence. For example, the findings of secondary uranium minerals, with measured ages between 70–100 ka and 28–36 ka BP (before present) at between 60 and 70 m depth in south west Finland at two sites adjacent to the Salpausselkä III ice marginal (Read et al. 2008), show that, at these periods, free water was available at this depth, which strongly suggests that permafrost could not have reached those depths. It is worth noting that these intervals, particularly the period 28–36 ka BP, are times at which permafrost development might have been at its most extensive, i.e. prior to the maximum ice cover of the Last Glaciation after a long period of oscillating, but generally cooling, temperatures.

Current analogues for permafrost development in similar conditions to those of Olkiluoto during the last glacial cycle are being investigated. The Greenland Analogue Project (Aaltonen et al. 2010b) has on-going investigations in Greenland which show that the permafrost depth at the forefront of the Russell Glacier (south west Greenland) reaches at most to 300 m depth.

It must be noted that the areas of permafrost occurrence in Canada and in Siberia are not good analogues for the Olkiluoto site as, in these locations, permafrost occurs inland (under continental climatic influences) and the geographical position of both Canada and Siberia differs from that of Olkiluoto, which affects seasonal temperature variations. Also, these regions were not subject to thick ice sheets. Thus, permafrost could continue to develop during glacial periods, which is not the case at Olkiluoto.

7.3.2 Dilute water intrusion

Many repository sites are situated in areas where there is potential for glacial impact on the host rock, as noted previously. A scenario being considered in SKB's safety

assessments (see SKB 2011) is the stage during glaciation when the ice-sheet margin is advancing or retreating over the repository and a large quantity of glacial meltwater is produced at the surface. In this scenario, dilute and oxygenated melt-water might penetrate down to repository depth and affect groundwater chemistry and redox conditions (e.g. Boulton et al. 1993, 2001), although the recent work of Jansson (2010) suggests that this is an over-simplification. Similar scenarios are discussed in *Performance Assessment* and *Formulation of Radionuclide Release Scenarios* even though it is considered unlikely that oxygen would actually reach repository depth.

Recent estimates for Fennoscandia and the Olkiluoto area show that insolation minima at 10–20 ka AP (after present), 50–60 ka, and 90–100 ka AP (e.g. Pimenoff et al. 2011) could induce glacial onset. The earliest minimum at 10–20 ka AP is considered unlikely due to high greenhouse-gas concentrations and low orbital eccentricity, but during the latter two periods, the onset of permafrost and/or glaciation seems likely.

In the case of a spent nuclear fuel repository at Olkiluoto, Schatz et al. (2013, in prep.) noted that transmissive fractures intersecting the canister deposition holes may provide pathways for dilute glacial groundwater to interact with the bentonite buffer. Localised free swelling of the buffer could occur with expansion into a fracture where it could lose density until it begins to flow in a gel state with the moving groundwater (i.e. is eroded). In addition, dilute and oxygenated glacial groundwater infiltration into the bedrock may trigger weathering processes of fracture minerals and fault gouges (cf. Banwart et al. 1994) and thus may have some impact on the sorption properties of the fracture minerals and if such groundwater reached repository depth, it could influence the redox conditions in the repository EBS.

Based on results in *Performance Assessment*, a few percent of canister positions may experience dilute conditions towards the end of the temperate period. These results are obtained by groundwater flow modelling assuming that the infiltrating groundwater has zero salinity. Dilute conditions may also be experienced during the glacial retreat, but the estimate of the number of canister positions experiencing these dilute conditions is strongly dependent on the possible duration of the meltwater intrusion and on the conceptual model for the interaction between the fracture water and the rock matrix. In reality, site evidence indicates that dilute glacial meltwater has never reached the repository depth and thus supports the contention that it is unlikely to do so in future.

In this section, selected information on the infiltration of surface and near-surface, dilute groundwaters at depth in crystalline host rock is examined in the light of site and natural analogue studies.

Natural analogue information of relevance

In his wide-ranging review of natural redox fronts from a broad range of geological environments, Hofmann (1999) noted the presence of numerous redox fronts in the sediments overlying the crystalline basement of northern Switzerland, a potential repository host rock. However, in the crystalline basement itself, it was noted that, in the otherwise reducing milieu the only indications of oxidised fluids were related to hydrothermal events in the open fractures (e.g. Alexander et al. 1990). There were no

indications of more recent, low temperature features associated with the presence of superficial waters, even though the area has been glaciated repeatedly.

In a more recent review of natural analogue studies of redox fronts, Sidborn et al. (2010) concluded that:

- The depth of oxidative leaching of uranium during recent time (< 1 Ma) due to infiltration of oxidising meteoric waters rarely exceeds 100 metres. Where the redox transition zone is found deeper, hydraulic conductivities of at least 10^{-6} m/s have been measured in the more conductive zones and the hydraulic gradient has been high;
- The extent of the alteration/oxidation is highly dependent on the porosity, including both matrix porosity and fracture porosity. A higher porosity implies a higher availability of reaction surfaces as well as pathways for transport of oxidants;
- The studies confirm that oxidising fluids are transported into the rock by matrix diffusion where they are buffered by mineral-water reactions.

A valuable analogue for the possible intrusion of dilute glacial meltwater at depth is offered at the Palmottu site (Figure 7-2) that, at the end of the last glaciation, was situated at the margin of the ice sheet being prone to the immediate influence of glacial waters. As at Olkiluoto, there are sub-horizontal fractures (Figure 7-3) that may have influenced in the deep penetration of dilute waters, limited to about one hundred metres depth. If in the future Olkiluoto is situated at the margin of an ice sheet, it is not expected that major changes would occur other than those observed at Palmottu.

Moreover, there is no evidence of significant oxidation or glacial water remnants at Olkiluoto at repository depth, if this had ever happened, would be expected to occur along the higher conductivity zones, something which is not observed (Pitkänen et al. 2009).

Conclusions and implications for Olkiluoto

The evidence examined here indicates that, although glacial meltwaters could have an impact on the repository by intruding dilute and oxidising surface waters at depth, most of these scenarios in which this occurs are based on highly conservative (and, arguably, over-conservative) assumptions. The current knowledge of the Olkiluoto site suggests that oxidising groundwaters never reached repository depth in the past and thus it is unlikely that this will happen in the future.

There is evidence from several sites in Europe that suggests that dilute and oxidising groundwaters can reach hundreds of metres into the bedrock (e.g., Sellafield; Milodowski et al. 2005), but generally only at sites that are relatively more permeable than the crystalline bedrock at Olkiluoto. The sub-horizontal fracture system at Olkiluoto could be responsible for limiting the intrusion of dilute glacial meltwaters at the site, with deeper intrusion being modified by mixing with older, more saline groundwaters at depth.

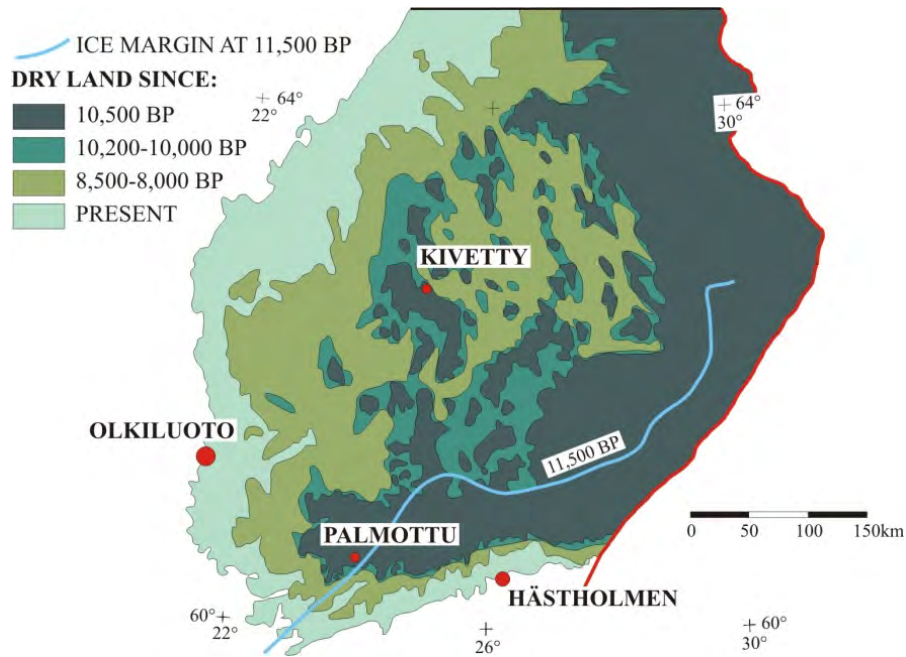


Figure 7-2. Situation of Palmottu with respect to the ice margin at the end of the last glaciation (Cedercreutz 2004; modified from Pitkänen et al. 2002).

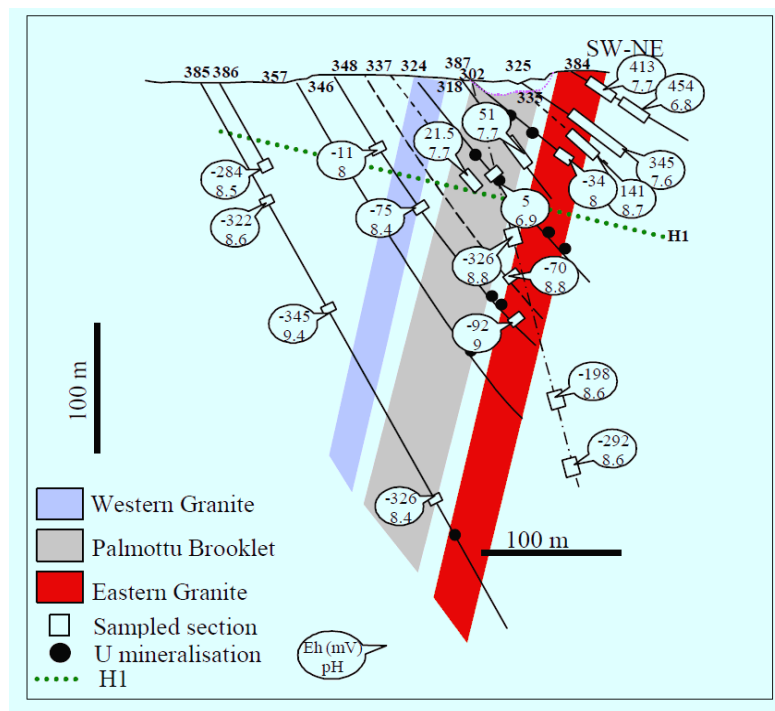


Figure 7-3. Vertical cross-section SW-NE through the Palmottu site indicating field Eh and pH values. Oxidising values are limited to above the sub-horizontal fracture H1 (Kaija et al. 2000).

7.4 Limited impact of earthquakes

Many national programmes have considered the impact of earthquakes on the repository near field (e.g. La Pointe & Hermanson 2002, Bäckblom & Munier 2002) and far field (e.g. JNC 2000a, Nagra 2002, Marcos et al. 2009) and they generally reach the conclusion that any potentially detrimental effects can be avoided by engineering solutions (e.g. careful emplacement of the engineered barriers – see Bäckblom et al. 2004; Pastina & Hellä 2006, for example) or adequate respect distances from existing fractures (e.g. JNC 2000a). Neall et al. (2007) provided a wide ranging summary of the current knowledge on earthquakes in Finland and noted that the repository location “...is advantageous with respect to the stability of the geosphere”. They further noted “...that the density and magnitude of earthquakes in Finland is lower than in other areas” (Figure 7-4) with earthquake magnitudes having never exceeded Richter 5 since records began in the 1880s (Marcos et al. 2009 and references therein). Saari (2008, 2012) noted that, within a radius of 100 km from Eurajoki, there have been only 9 recorded earthquakes, with the nearest event ($M = 3.1$) at Uusikaupunki, 35–40 km from Olkiluoto in 1926. Another event about 40 km from Olkiluoto occurred in Laitila in 2007 ($ML = 1.9$).

However, Neall et al. (2007) also pointed out that “...seismic activity in the future cannot be excluded” and future seismic activity in the Olkiluoto area can be expected due to earthquakes in the immediate aftermath of future glaciations (La Pointe & Hermanson 2002) as well as due to slow stress and isostatic readjustments following ice retreat and marine transgression.

With respect to post-glacial seismic activity, it should be noted that, although a survey in the Olkiluoto area discovered a small number of faults that may have post-glacial origin, none were found in the immediate vicinity of the actual site (Lindberg 2007). McEwen & Andersson (2009) noted that any postglacial movement on the largest scale deformation zones (which are at some distance from the repository) would result in displacements on smaller scale zones and individual fractures in the vicinity of the repository. Fälth et al. (2010) stated that “*The earthquake risk relevant to the safety of a KBS-3-type spent-fuel repository in Sweden is that of direct canister damage due to fracture shear displacements across deposition holes. Vibrations of the buffer-canister system will not damage the canister.*” For the SR-Site safety assessment (SKB 2010), a 0.05 m fracture displacement across a deposition hole counts as canister damage (regardless of the actual shear velocity and the actual intersection geometry).

In comparison, a recent analysis by Fälth & Hökmark (2011) suggests that any such postglacial displacements “...would be very modest at the Olkiluoto site...” with a maximum displacement of 0.005 m. The conclusions of both Neall et al. (2007) for Olkiluoto and, more recently, Fälth et al. (2010) for the Forsmark and Laxemar sites in Sweden are that any risk of canister damage can be minimised with an appropriate repository layout. For the spent nuclear fuel repository at Olkiluoto, the appropriate layout is thus constrained by the rock suitability classification criteria (RSC criteria, see McEwen et al. 2012). As such, this process will not be discussed further here.

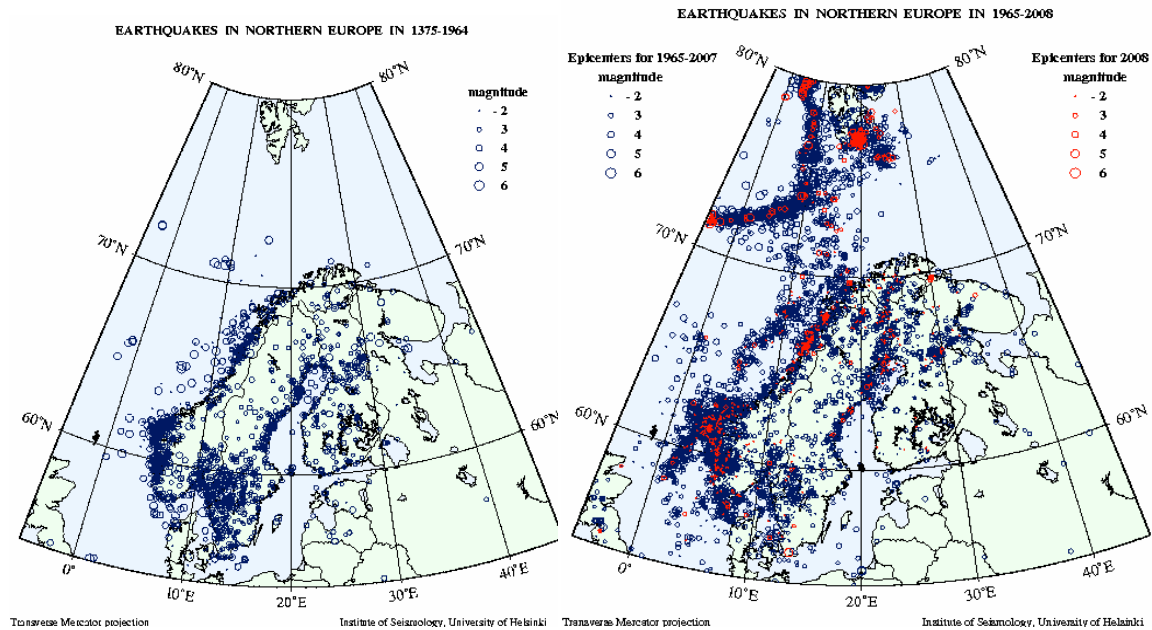


Figure 7-4. a) Earthquakes recorded in Northern Europe from 1375 to 1964 and b) Earthquakes monitored in Northern Europe 1965–2008 (University of Helsinki ¹¹).

However, as stakeholder concerns tend to cover wider aspects of potential earthquake impact on a repository, so a wider range of evidence is examined here to put potential earthquake impact in perspective.

7.4.1 Underground facilities

Until very recently, little novel work had been carried out on the likely impact of earthquakes on a repository since the study of the fractured granite at the Kamaishi URL (underground rock laboratory) from the 1990s. Here, 344 earthquakes were recorded from February 1990 to March 1998, with the largest earthquake having a magnitude M8.1. This clearly showed the ground acceleration from earthquakes rapidly dropping off with depth (Figure 7-5), implying that even powerful earthquakes should not impact a deep repository system to any great degree (JNC 2000a). Significantly, a similar pattern was observed for the Hosokura mine in sediments (JNC 2000b).

This has been backed by preliminary investigations of the 11 March 2011 moment magnitude (M_w) 9.0 Tohoku-Oki Earthquake which occurred on the Japan Trench plate boundary off the eastern shore of northern Honshu (Ide et al. 2011). This was a massive earthquake which induced stunning displacements across the entire country and is clearly of much greater magnitude than would be expected anywhere in the Fennoscandian Shield.

¹¹ Sources:
a) <http://www.seismo.helsinki.fi/bulletin/list/catalog/histomap.html>
b) <http://www.seismo.helsinki.fi/bulletin/list/catalog/instrumap.html>

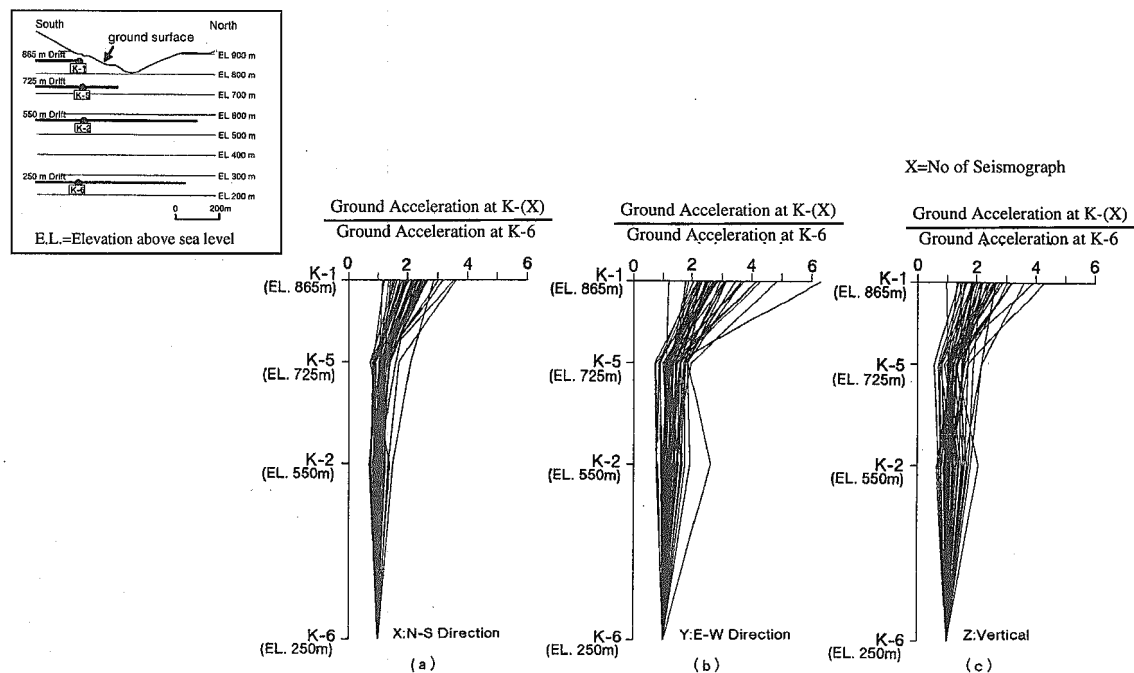


Figure 7-5. Ratio of ground acceleration following earthquakes measured in the Kamaishi URL (from Shimizu et al. 1996). (a) N-S direction, (b) E-W direction (c) vertical direction. This figure includes 41 earthquakes measured between January 1991 and December 1994.

In addition, many more earthquakes have shaken Japan in a few months than have impacted the Olkiluoto area in millennia. Nevertheless, the work newly published by NIED (National Research Institute for Earth Science and Disaster Prevention) shows that maximum ground acceleration measured on the surface (Figure 7-6a) was greater than that measured at a depth of 100–150 m (Figure 7-6b) in a series of boreholes placed across Japan. When the data are normalised (i.e. maximum ground acceleration at the surface/maximum ground acceleration at depth), it can be seen (Figure 7-7) that the impact at depth is generally much less than that on the surface. It should also be noted that these data are for boreholes which are much shallower than the Olkiluoto repository and so, by analogy with the Kamaishi study, it would be expected that maximum ground acceleration would be even less at greater depth.

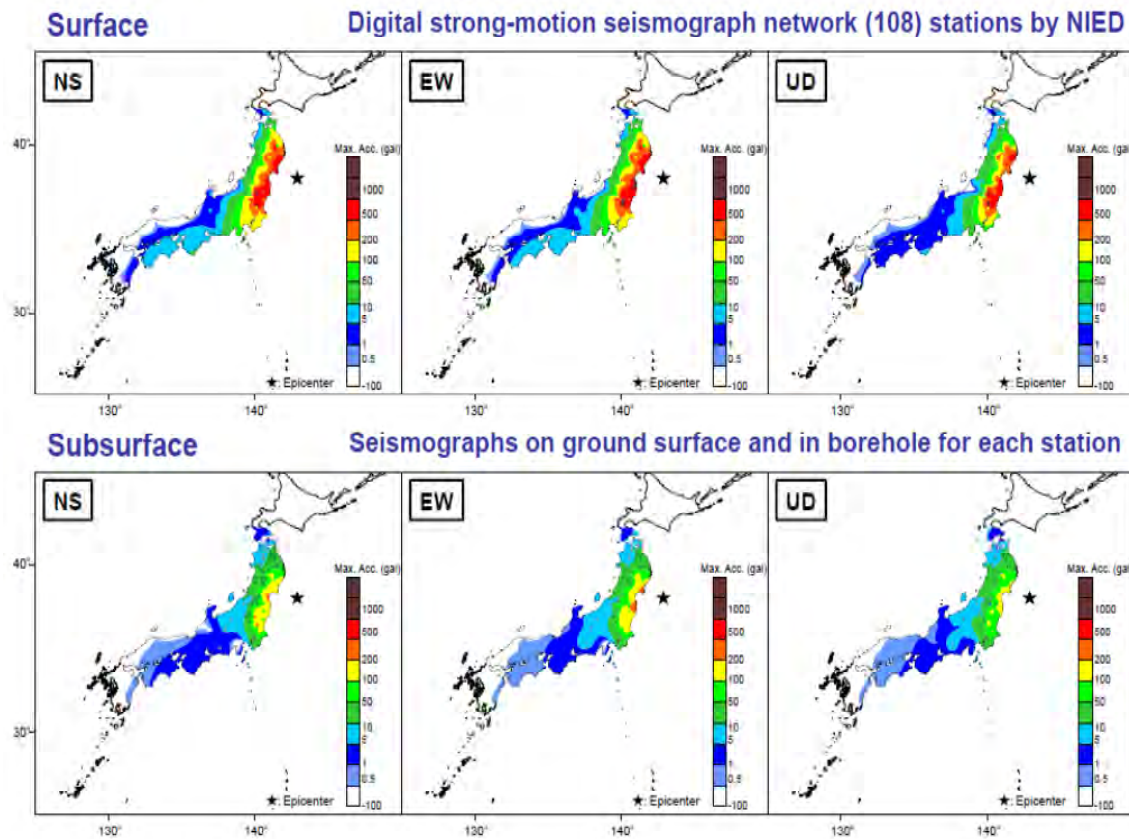


Figure 7-6. (a) Maximum accelerations measured on the surface across Japan; (b) measured at depth (100–150 m) in boreholes (NIED 2011). NS: north-south movement. EW: east-west movement. UD: vertical movement.

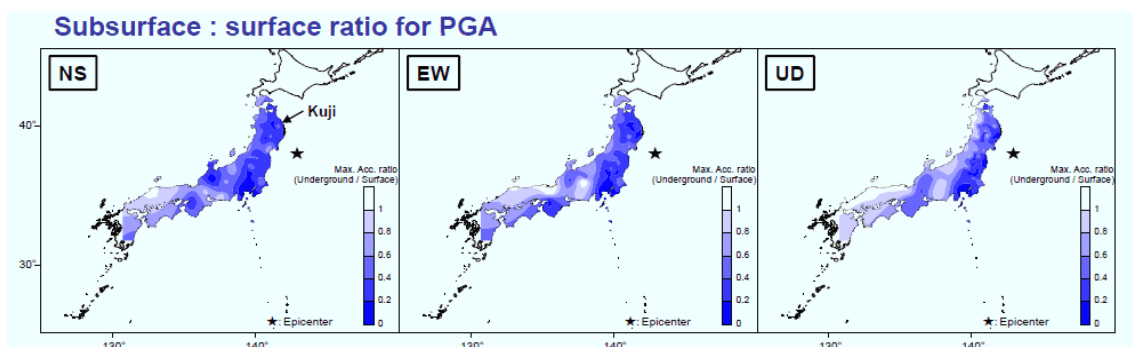


Figure 7-7. Same data as in Figure 7-6, normalised to show the ratio between surface and at depth (100–150 m) (NIED 2011). NS: north-south movement. EW: east-west movement. UD: vertical movement. PGA: peak ground acceleration.

The situation might not always be so clear, however, and Goto et al. (2009) analysed a different NIED dataset, using 69 sites with seismometers at depths greater than 300 m (Figure 7-8) and for 861 earthquakes where the maximum ground acceleration was >10 Gal. They showed that, although ground acceleration does generally drop off with depth (and distance from the earthquake hypocentre), this is not always the case (Figure 7-9) nor is there as simple a relationship as was shown in the early Kamaishi dataset.

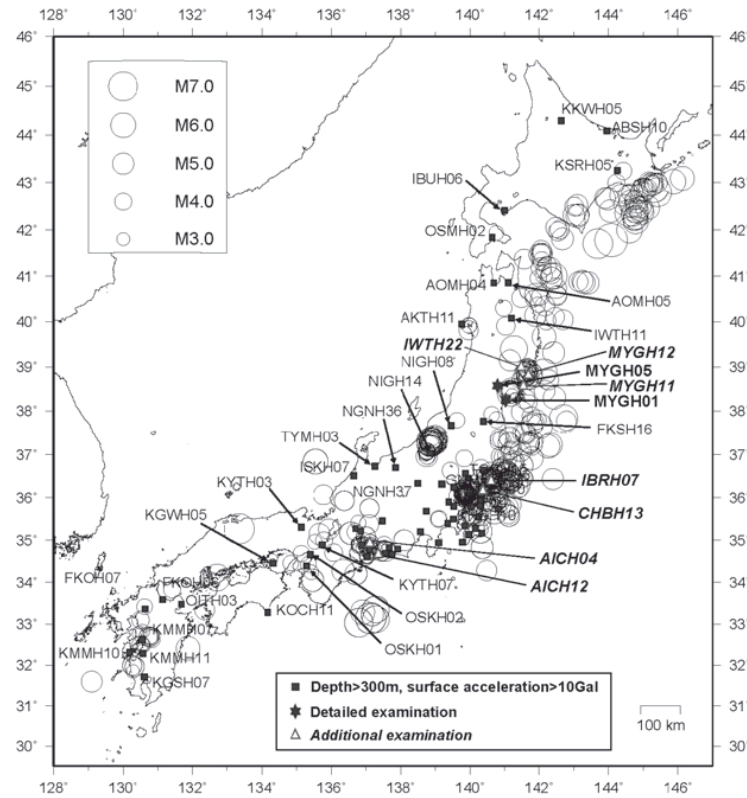


Figure 7-8. Location of earthquakes and monitoring sites analysed by Goto *et al.* (2009).

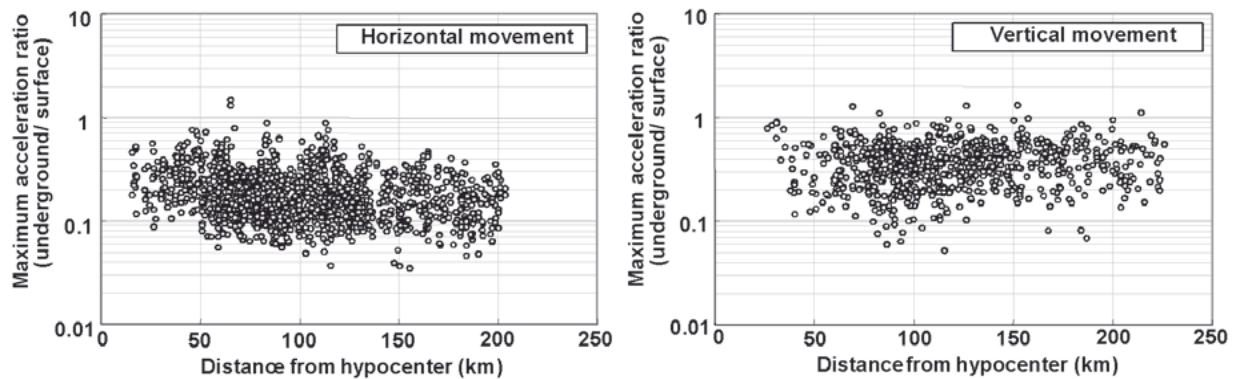


Figure 7-9. Maximum ground acceleration ratio relative to distance from the hypocentre (Goto *et al.* 2009).

In general, the data indicate that, where the repository host rock has a shear wave velocity distribution similar to or higher than the rock at the earthquake hypocentre, under certain conditions, the ground acceleration at depth can slightly exceed that at the surface. It is important to note that these conditions are the exception rather than the rule (note the very few points greater than 1 in Figure 7-9) and do not act to greatly magnify the ground acceleration at depth.

Many of the seismic data discussed above are from open tunnels or boreholes; as examples of the potential impact of seismic activity on a repository during the post-closure period (following tunnel backfilling) are more difficult to find as only another existing deep repository of a similar design to Olkiluoto would be a direct analogy. However, the behaviour of other, filled tunnels and caverns offers at least an insight into possible impacts. In Japan, JOGMEC (Japan Oil, Gas and Metals National Corporation) is responsible for storing a buffer supply of fuel and strategically important metals for the Japanese government. Currently, they store some 170 days national oil and 70 days national LPG (liquefied petroleum gas) usage in 13 surface and underground storage terminals across the country (see Figure 7-10). One of JOGMEC's storage terminals was very close to the epicentre of the Tohoku-Oki earthquake.

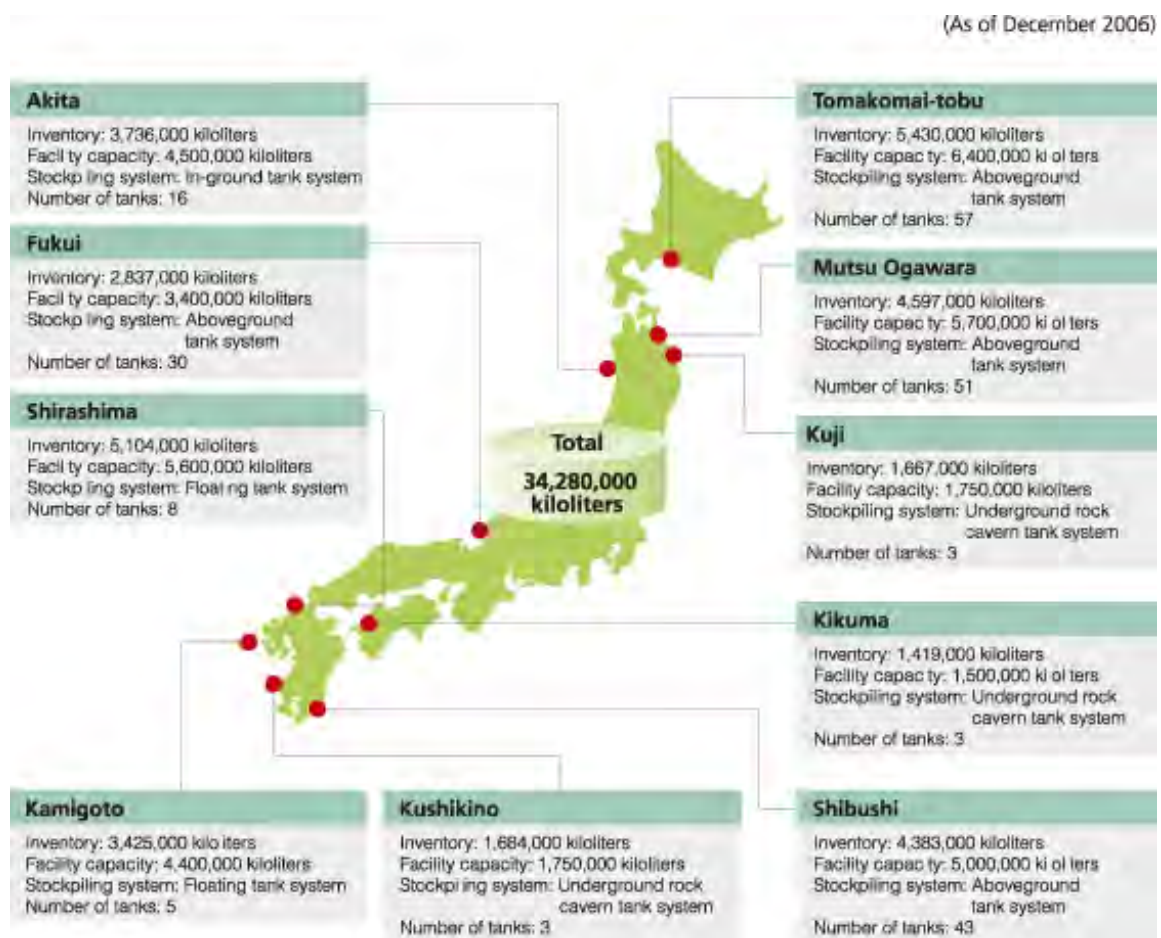


Figure 7-10. JOGMEC's fuel storage terminals in Japan (courtesy JOGMEC).

The Kuji City (Iwate Prefecture) underground fuel storage terminal contains $1.75 \cdot 10^9$ litres of fuel in three caverns excavated from the rock (see Figure 7-11). The storage system had its above-ground facilities washed away by the tsunami which followed the earthquake but, as has been widely reported in the news media (e.g. Argus 2011), there have been no signs of any leak due to either the earthquake or the flooding.

The terminal at Kuji City is not directly comparable to a repository site insofar as a groundwater injection system is used to increase the pressure of the ground- and porewater around the caverns to above that of the cavern itself, minimising any possible leaks. However, although the compressibility and general behaviour of the fuel oil in the Kuji City caverns is clearly different from the bentonite, canister and spent nuclear fuel in the Olkiluoto repository tunnels, the fact that the cavern system retained its integrity despite being in close proximity to an earthquake of much greater magnitude than is likely to affect the Olkiluoto area increases confidence that deeply buried structures will be little impacted by earthquakes.

In an attempt to increase stability in some of the deep ore mines of South Africa, already mined out tunnels and stopes are backfilled (e.g. Kirsten & Stacey 1988), as discussed in Section 7.2, to increase stability of underground openings generally. Backfill (usually mining rubble from elsewhere in the mine) acts to dampen the impact of earthquakes by reducing the resonant frequencies of the unsupported tunnel walls. For example, Spottiswoode & Churcher (1988) reported that backfill reduced the spans of the detached strata (i.e. the tunnel walls) and thereby increased their resonant frequencies from 70 Hz to 140 Hz. The radiated seismic energy at 140 Hz is one quarter of that at 70 Hz, and therefore there would be far less “...excitement and shake-out...” of potentially loose material from the tunnel walls at the higher frequency.



Figure 7-11. Typical JOGMEC underground rock cavern used for storing fuel oil. JOGMEC specifically notes that the design “...features high resistance to earthquakes...” (JOGMEC 2011).

The reduced energy at the higher frequencies of vibration will also be more rapidly attenuated in the fractured ground. The backfill itself will act as a more effective damper through the dissipation of interparticle frictional energy than will conventional tunnel support. Squelch et al. (2001) noted that backfill caused a reduction in peak ground velocities and accelerations, confirming the observations of Adams & Gürtunca (1991), who showed that the backfill reduced the vibration time of the rock, following an earthquake, and that overall ground motion was damped by backfill, so minimising tunnel damage. However, the quality of the backfill emplacement was extremely important as poorly emplaced material (e.g. Figure 7-12) will fail to dampen the ground motion, leading to tunnel damage. Squelch et al. (2001) noted that backfill which has been packed carefully so that the stiffness is similar to the wall rock will behave best.

This work would appear to imply that the backfilling of tunnels at Olkiluoto immediately after canister emplacement may lead to safer working conditions during the operational phase and will certainly contribute to maintaining tunnel integrity during the post-closure period.



Figure 7-12. Example of variability of backfill placement at the Kopanang mine (South Africa) with a 1.5 m-wide gap left between backfill and the tunnel roof (from Squelch et al. 2001).

7.4.2 Conclusions

Several conclusions can be drawn from the preceding discussion and these include:

- Olkiluoto is considered a seismically quiet site;
- Generally, the maximum ground acceleration in tunnels is expected to decrease with depth (although recent work suggests it is not necessarily a simple relationship);
- Mining backfill experience suggests that filled tunnels will survive general seismic instability well – especially if backfill is emplaced in a quality assured manner;
- The mechanisms and processes relevant to this observation are relatively well understood and could be assessed in detail for Olkiluoto if necessary.

Thus, overall, there would appear to be no significant earthquake-related issues for the Olkiluoto site.

7.5 Erosion

Expected denudation rates in the Olkiluoto area are low and the estimated glacial erosion of the bedrock during a whole glaciation period is at maximum (very conservatively) 10 m, very likely in the order of just a few metres (see Table 7-1). Erosion related to meltwater discharge can produce more pronounced local erosional features than those expected from plucking and abrasion. Low-relief topography yields, in general, very low erosion rates. However, the deepest glacially-induced erosional features in Finland are meltwater discharge canyons and giant pot holes or “saivos” that are features commonly a few metres to a few tens of metres deep carved in the crystalline bedrock by melting waters and rock materials. Canyons are often related to already weaker zones in the bedrock, whereas potholes may form on seemingly intact crystalline rock. The deepest pothole found in Finland is “Paholaisen hiidenkirnu” with a depth of 15.4 m (northern Finland). Geomorphological pothole-like features in Lapland (saivos) can be deeper, at maximum 60 m deep, but in these cases the local topography affects the formation and it is likely that they have been polygenetically formed during several glaciation periods (Johansson 2005). On the other hand, some of the potholes (in Pihlajamäki, Helsinki) are older than the last deglaciation (Hirvas & Huhta 1997) but have a maximum depth of 8.7 m. In the Helsinki area there are also potholes formed during the last deglaciation having depths of a few metres at maximum illustrating the very low overall erosion in the area.

The overburden at Olkiluoto (and also mostly at the regional scale) is composed of Quaternary deposits that date from different stages related to the last glacial period. This implies that all the sediments formed prior to the last glaciation have been removed or resedimented during Weichselian glaciation and ice retreat. The bedrock is close to the surface and has more undulations in its topography than is seen in the overlying sediments. Thus, the average areal erosion rate does not necessarily tell the whole story and the erosional processes described above can also have occurred to some extent at Olkiluoto. During future glacial periods, the current Quaternary sediments are likely to be removed or redistributed (as has occurred in general in Finland, except in some locations where pre-Weichselian formations are found) and replaced by similar sedimentary cover.

Table 7-1. Regional average (bedrock) erosion rates for some locations in Sweden and Finland estimated by different authors.

Area/Location	Erosion	Method	Reference
Lapland (Parkajoki Sweden)	max. 1.6 m /Ma	Cosmogenic isotope analyses from weathering mantles and boulders	Stroeven et al. 2002
Lapland (Pyhä-Nattanen and Riestovaara, Finland)	Tors ⁽¹⁾ predating several glaciations	Cosmogenic isotope analyses from tors	Darmody et al. 2007
Sweden peneplain (Laxemar and Forsmark)	2–5 m during full glacial cycle	Literature review	Olvmo 2010
Low lands Sweden	0.2 to 4 m during full glacial period	Thickness of minerogenic Quaternary sediments	Påsse 2004
In Finland during the Weichselian	8.3 m	Same as above	Okko 1964

1: Tors are generally low, rocky hills or piles of rocks on hilltops, characteristic of weathered granitic terrains.

In the overall context of the repository, these erosional processes do not pose a risk for the intact rock as they are not sufficient to carve deeply enough to reach the repository level, even during multiple glaciations. Exposed topographic lows may be eroded more than the average value would imply, but this is still of limited extent compared to the repository depth. In case of closure structures for access tunnels, for example, erosion may affect these surface and near-surface structures.

7.6 Limited likelihood of damage due to meteorite impact

Meteorite impact is a phenomenon often used as a ‘what-if scenario’ by opponents of radioactive waste repositories, as in the question ‘What would happen if a meteorite struck the repository?’ Meteorite impact is usually excluded from consideration as a FEP in safety assessment studies on the basis that the probability that a meteorite would strike the repository site (say, $\sim 10 \text{ km}^2$) is extremely small, given that the total surface area of the earth is approximately $5 \cdot 10^8 \text{ km}^2$ – or about 5 million times larger than the repository area. In its strategy for design of nuclear facilities (specifically, power plants) for external hazards, the IAEA (2003b) excludes meteorite impact based on a conservative upper limit of probability of 10^{-7} per year (slightly variable according to latitude). This is for surface facilities; to have any effect on a deep geological repository, it is clear that the meteorite would need to be a great deal larger than that which could cause damage to a surface facility, thus raising the question of whether the meteorite impact or potential damage to the repository would be of greater concern to the local population.

The vast majority of meteorites entering the earth's atmosphere are small and burn up or disintegrate before hitting the Earth's surface. NASA (1994) states that most stony meteorites less than about 10 m in diameter are destroyed in the atmosphere by thermal explosions¹². A meteorite of sufficient size to excavate the radioactive waste from several hundreds of metres below the surface would probably need to be of the 100 m-class of objects. The infamous 1908 Tunguska event in Siberia was due to a meteorite some tens of metres in diameter¹³ which did not survive its passage through the atmosphere and exploded several kilometres above the surface and thus, despite the widespread damage to the forest, left no impact crater. This is the largest terrestrial impact event in recorded history. Although, as has been pointed out (RMS 2009), the fact that it was recorded, may be in part because of the interest of the scientific community investigating this event in a sparsely populated, remote area. Even in relatively recent times, before the widespread use of aerial and satellite photography, a similar event in, say, Antarctica or even Northern Canada or Australia, could have gone unnoticed or at least unrecorded, as would any fall over the ocean. Even so, it is estimated that meteorites of this magnitude enter the earth's atmosphere only once or twice in a thousand years (NASA 1994).

The composition of the meteorite is important if it is to produce an impact crater – iron meteorites are more likely to survive transit of the atmosphere intact. The well-known Barringer Crater (also known as Meteor Crater) in Winslow, Arizona (Figure 7-13) is the result of an iron meteorite about 50 m in diameter which struck about 50,000 a BP. The crater is 1.2 km in diameter and about 180 m deep – not deep enough to directly damage a repository at a depth of around 400 metres but geophysical surveying revealed that the fracturing below the actual crater extends to depths of around 1 km (Ackerman et al. 1975). In fact, it has been shown that there is a relationship between crater diameter and depth of fracturing as both relate to the impacting object's energy and velocity (Ahrens et al. 2002).

¹² This empirical observation, backed by understanding of the thermal properties of stony meteorites is currently under redefinition after a small stony meteorite was, unusually, observed to excavate a 15 m-wide, 3 metre-deep crater in Carancas, a remote but inhabited area of Peru. Scientists have hypothesised that its unusual behaviour was a function of its high speed and the angle of the trajectory (Schultz et al. 2008).

¹³ This is still subject to some debate; 40–60 m diameter is generally the estimated size, but size and composition – whether stony or more ice-rich and comet-like – are hard to disentangle from the available evidence. See, for example, Farinella et al. (2001).



Figure 7-13. The Barringer Crater (also known as Meteor Crater) Winslow, Arizona was the result of the impact of a 50 m diameter iron meteorite about 50 ka BP. The crater is 1.2 km in diameter and 180 m deep. (Photo: U.S. Geological Survey.)

Thus while it is possible in principle that a moderately-sized iron meteorite could affect the long-term performance of the repository system by altering geosphere properties, it should be noted that iron meteorites make up only about 5 % of meteorite falls and the majority of the rest may leave no significant crater but only a scattering of fragments (fragments of a size that nevertheless could potentially do some damage to a surface facility like a nuclear power station). Furthermore, the geosphere may be changed but the effect is localised to the vicinity of the impact, thus the probability of impact directly over the repository is still the key parameter in determining the hazard from such an event. Very crudely, the probability can be estimated as (frequency of 100 m-class objects striking the earth [per year]) $\times$ (percentage of iron meteorites) $\times$ (repository area [km^2] / earth surface area [km^2]) per year, or $0.002 \times 0.05 \times 10 / (5 \cdot 10^8) =$ approximately $2 \cdot 10^{-12}$ per year so that, even over a 1 Ma time span, the probability of a meteorite impact affecting the performance of the repository is still negligible.

An alternative way of considering the probability is to use the evidence of large meteorite impacts in the past. The Earth Impact Database (EID) comprises a list of confirmed impact structures from around the world. Many of these are very large structures (kilometres in diameter) as smaller features tend to be more quickly obscured by surface processes such as erosion, sedimentation, vegetation, agricultural practices etc. Thus this population does represent the large end of the spectrum of meteorite impacts. However, to date, there are 178 confirmed impact structures in the database, of

which 10 are located in Finland¹⁴ (Table 7-2). The oldest craters (Keurusselkä and Paaselkä) are less than 1800 Ma and the youngest is about 73 Ma (Lappajärvi) giving a rough estimate of one large meteorite impact every 170 Ma.

Table 7-2. Meteorite impact structures in Finland.

Crater name	Latitude/Longitude	Diameter (km)	Age (Ma)	Exposed	Drilled
Iso-Naakkima	N 62° 11' / E 27° 9'	3	> 1000	No	Yes
Karikkoselkä	N 62° 13' / E 25° 15'	1.5	~230	Yes	-
Keurusselkä	N 62° 8' / E 24° 36'	30	< 1800	Yes	No
Lappajärvi	N 63° 12' / E 23° 42'	23	73.3±5.3	Yes	Yes
Lumparn	N 60° 9' / E 20° 6'	9	~1000	No	Yes
Paaselkä	N 62° 2' / E 29° 5'	10	< 1800	Yes	Yes
Sääksjärvi	N 61° 24' / E 22° 24'	6	~560	Yes	Yes
Saarijärvi	N 65° 17' / E 28° 23'	1.5	> 600	-	Yes
Söderfjärden	N 63° 2' / E 21° 35'	6.6	~600	No	Yes
Suvasvesi N	N 62° 42' / E 28° 10'	4	< 1000	No	Yes

¹⁴ Terrains comprising old, stable rocks, especially with little relief, as in Finland, Canada, South Africa, Australia and Siberia tend to be over-represented in the database because structures are both more obvious and better preserved over long periods than in more active geological terrains.

8 EVIDENCES FOR THE SUITABILITY OF THE REPOSITORY DESIGN AND MATERIALS

8.1 Spent nuclear fuel stability

Many natural analogue studies have focused on the behaviour of spent nuclear fuel (see the review of Miller et al. 2000). Useful information can, for example, be obtained on its chemical stability in relevant environments and on the possibility of nuclear criticality. There are, however, limits to the analogy. For example, one gramme of natural uranium contains about 178 kBq of radioactivity. In comparison, after discharge from a reactor and 10 years of cooling and decay underwater, one gramme of spent nuclear fuel has an inventory of 750 MBq of radioactivity (Tait et al. 2000), and is more than 40,000 times more radioactive than is the original uranium ore. However, after approximately 100,000 years, due to the decay of most of the fission products, spent nuclear fuel has decayed to a similar level of radioactivity to that found in the natural uranium.

Further, the form and precise mineralogy of natural uranium dioxide differs from reactor spent nuclear fuel. Natural uraninite has formed at lower temperatures than commercial fuels, and also the presence of varying amounts of radiogenic lead in old uraninites (up to 20 % at Oklo) can affect its chemical stability compared with the effect of the small amounts of fission products in spent nuclear fuel. Nevertheless, natural analogues of spent nuclear fuel can be of use if these limitations are kept in mind.

8.1.1 Spent nuclear fuel dissolution

Short-term laboratory studies show that the dissolution rate of UO_2 depends on the redox state of the system. The more oxidising the conditions the faster is the UO_2 dissolution rate. The dissolution rate does not become significant until the fuel is oxidised to the U_3O_8 state because the crystal lattice structure of the material is significantly rearranged in this oxidation state.

Smellie (2011) also noted that detailed examination of the Cigar Lake site has:

- confirmed that the dissolution behaviour of UO_2 corresponds to the thermodynamic solubilities close to the hypothetical U_3O_7 phase;
- suggested that the predominant aqueous speciation of uranium is dominated by carbonate complexes;
- shown that no oxidative dissolution has been observed even where relatively high redox potentials (+0.1 to +0.2 V) were measured;
- indicated, based on mineralogical and geochemical information, that the threshold of oxidative dissolution occurs at redox potentials greater than +0.2 V.

Curtis (1996) also attempted to quantify the natural rate of UO_2 dissolution under reducing conditions at Cigar Lake by measuring the amount of fission products released from uraninite as an indication of the degree of ore dissolution. Tc-99 is produced continuously in measurable quantities by natural fission processes in the Cigar Lake orebody (Cramer & Smellie 1994) and so its release serves as a long-term indicator of ore dissolution under ambient reducing groundwater conditions. Uraninite corrosion rate calculations indicated values ($1.1 \cdot 10^{-6}$ to $1.5 \cdot 10^{-6}$ /a) in accordance with the then current

models for radionuclide release from spent nuclear fuel (e.g. SKB 1999, ENRESA 2000). But if I-129 is used as a tracer at Cigar Lake, the estimated release rate decreased to 2–4 orders of magnitude less than the Tc-99 rates.

However, as noted in Alexander et al. (2010), the technetium thermodynamic database (Tc TDB) at the time of these calculations was relatively weak and so it would be worthwhile repeating the calculations now using both an updated Tc TDB (e.g. Hummel et al. 2005) and updated radionuclide release models.

Overall, Miller et al. (2000) noted that both laboratory and natural analogue studies indicate that the kinetics of UO_2 dissolution, either as spent nuclear fuel or uraninite, is exceptionally slow under the reducing conditions expected in the near field of a spent nuclear fuel repository. Although dissolution rates cannot be quantified readily from natural analogue data (see comments above), the abundance of naturally-occurring uraninite, some over 1000 Ma old may be taken as a general indication of its stability in the geological environment. However, extrapolation of this apparent longevity to spent nuclear fuel in the repository environment is done cautiously, due to the uncertain long-term effects of the high levels of radiation dose experienced by spent nuclear fuel and the differences in the thermal history of the spent nuclear fuel and the ore.

8.1.2 Evidence for conditions favouring low dissolution rates

As stated above, the dissolution rate of UO_2 depends on the redox conditions in the vicinity of the spent nuclear fuel. These conditions can be affected by the radiolysis of the surrounding water or moisture. Miller et al. (2000) discussed natural analogues of radiolysis. Here, just one natural analogue example of radiolysis will be discussed. Although the first evidence of radiolysis in nature was found at Oklo, an arguably more relevant site is that at Cigar Lake in Canada as any radiolysis occurred under conditions more relevant to a deep radioactive waste repository. The Cigar Lake uranium deposit is a sandstone-hosted orebody located in the Athabasca Basin of northern Saskatchewan, Canada (Figure 8-1) and formed about 1300 Ma ago. The ore lies at a depth of 432 m below the surface and is located at the contact between the sandstone and altered basement rock and is separated from the sandstone host by zones of clay-rich altered sandstone (Figure 8-2; see Cramer 1994, for details). The deposit is hydrothermal in origin and consists of an irregularly-shaped lens about 2150 m long, 25 to 105 m wide and 1 to 20 m thick. The average grade of the ore is around 12 wt% uranium with maximum concentrations up to 55 wt%, the highest known for any uranium orebody. Alteration of the host sandstone resulted in clay (illite) formation around the ore zone which provides an analogue of the bentonite buffer around a waste package in a HLW or spent nuclear fuel repository.

It was decided to examine radiolysis at the site due to the presence of amorphous ferrihydrites and haematite (by oxidation of Fe^{2+}) at the ore/clay halo contact (see Figure 8-2). In the end, the iron-rich zone of the clay halo could be explained by late-stage, low-temperature hydrothermal fluids during the original ore formation (Smellie & Karlsson 1996), but laboratory studies of the ore itself provided important information on radiolysis effects.



Figure 8-1. Aerial view of Cigar Lake, Canada. The orebody lies over 400 m below the lake, but no radiological expression of the richest uranium orebody in the world is found on the surface. (Courtesy of producers of “Traces of the Future”.)

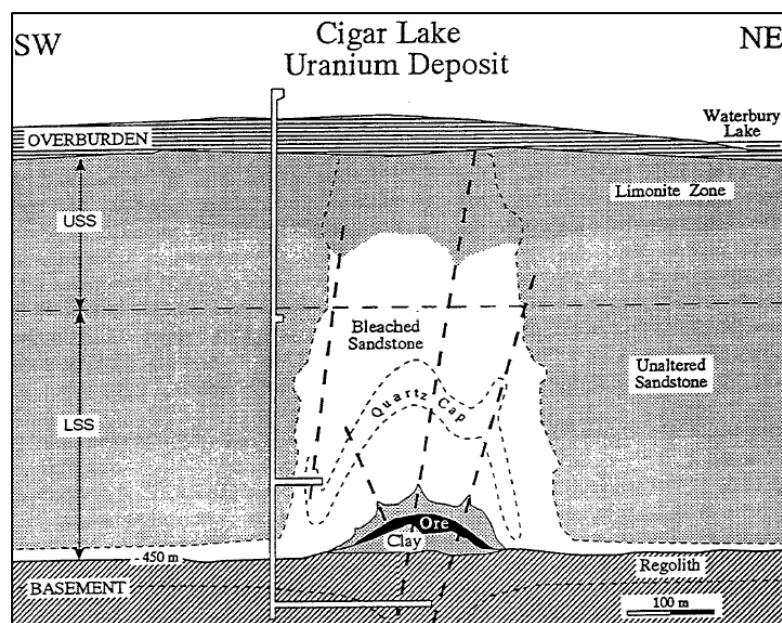


Figure 8-2. Schematic cross section through the Cigar Lake uranium deposit showing major lithological types, groundwater flow pathways, main fractures and some of the mine shafts and tunnels (from Karlsson et al. 1994).

By γ -irradiating natural UO_2 from Cigar Lake and analysing the solid phase, the amount of uranium that had reacted could be measured and a corrosion rate determined. A reasonable agreement was obtained between these experiments and the modelled results. However the modelling was found to overestimate (by more than an order of

magnitude) the effect of α -radiolysis, the dominant form at Cigar Lake. Calculation of radiolytic oxidation using SKB's then current models showed that the total Cigar Lake orebody should have been completely oxidised and dispersed within 17 to 170 Ma – something which clearly did not happen.

It was generally concluded at the end of the analogue study in 1994 that the effects of α - and γ -radiolysis at Cigar Lake were insufficient to cause significant oxidation of the UO_2 even when integrated over the lifespan of the orebody (Cramer & Smellie 1994). However, a subsequent re-analysis of the data by Smellie & Karlsson (1996) took a more realistic look at radiolysis, including the dependence on UO_2 grain-size, porosity, groundwater chemistry and the geometry of the radiation dose rate distributions from UO_2 grains. This established more accurately what fraction of radiation reached the pore- and groundwaters to initiate radiolysis.

Models (e.g. Liu & Neretnieks 1996) were tested against these data and the results indicated that lack of radiolysis in the orebody may be partly explained by large amounts of radiolytically-produced oxidants being neutralised by the reductive capacity of the UO_2 itself (Eriksen et al. 1995) or by reactions with impurities in the system. In the Cigar Lake context, therefore, based on laboratory experiments interfaced with direct observations and measurements, it was suggested that the observed oxidation of the uraninites from the orebody probably occurred shortly after the ore formation phase, and the reductive capacity, and hence stability of the orebody (i.e. analogous spent nuclear fuel), has essentially remained unchanged for the last 1200 to 1300 Ma (Smellie & Karlsson 1996). A similar conclusion regarding the ore's reductive capacity was reached for the Oklo reactors (Smellie 2005).

These studies rely on the assumption that the integrated radiolytic yield is simply proportional to the integrated dose. Experimental evidence related to the effects of hydrogen on dissolution suggests that the radiolytic mechanism is more complex. Carbol et al. (2009) have shown evidence for surface-catalysed reaction between hydrogen and radiolytic oxidants. As natural ores such as Cigar Lake produce both hydrogen and oxidants, this mechanism might also be expected there.

Finally, it is emphasised that not enough information on fluxes of oxidants and reductant at Cigar Lake is available to allow a direct comparison with the likely situation in and around a spent nuclear fuel canister.

8.1.3 Low likelihood of criticality

The Finnish regulations (Section 2.2) require that the spent nuclear fuel packages be designed to prevent the spent nuclear fuel from going critical in repository conditions. If criticality was possible, it could lead to a serious loss of containment of the canister and also affect other repository barriers.

Induced nuclear fission (criticality) has been studied in detail at the Oklo and Okélobondo deposits in Gabon, Africa, where at least 16 'natural reactors' have been documented, each having undergone criticality some 2 Ga ago (Figure 8-3). The data were reviewed by Smellie (2005) and the following main conclusions were reached:

- The reactor zones had the requisite physical and nuclear characteristics to form a self-sustaining chain reaction, given the abundance of U-235 present (about 4 %) nearly 2 Ga ago;
- The criticality events were self-limiting and there is evidence of a rapid cycling between critical and sub-critical states over periods of hours;
- The compact mass of the reactor zones would be conducive to minimising collapse and maximising thermal neutron utilisation in the uranium. Moreover, the surrounding sandstones, some exhibiting fracturing, would provide ample water to moderate and reflect neutrons;
- The inventory of fission products and the slight excess of U-235 in some zones support the conclusion that induced fission of U-235 and neutron capture by U-238 occurred in the area;
- The reactor zones during criticality, operating at 150–160 bars and 300–400 °C, remained rather closed systems with respect to uranium and to most of the fission products, with the exception of alkali metals, alkaline earth elements, rare noble gases and some heavy metals such as Mo, Ag and Te;
- Under certain chemical conditions, fluid movement through uranium oxide has selectively removed and transported a fraction of plutonium generated during criticality;

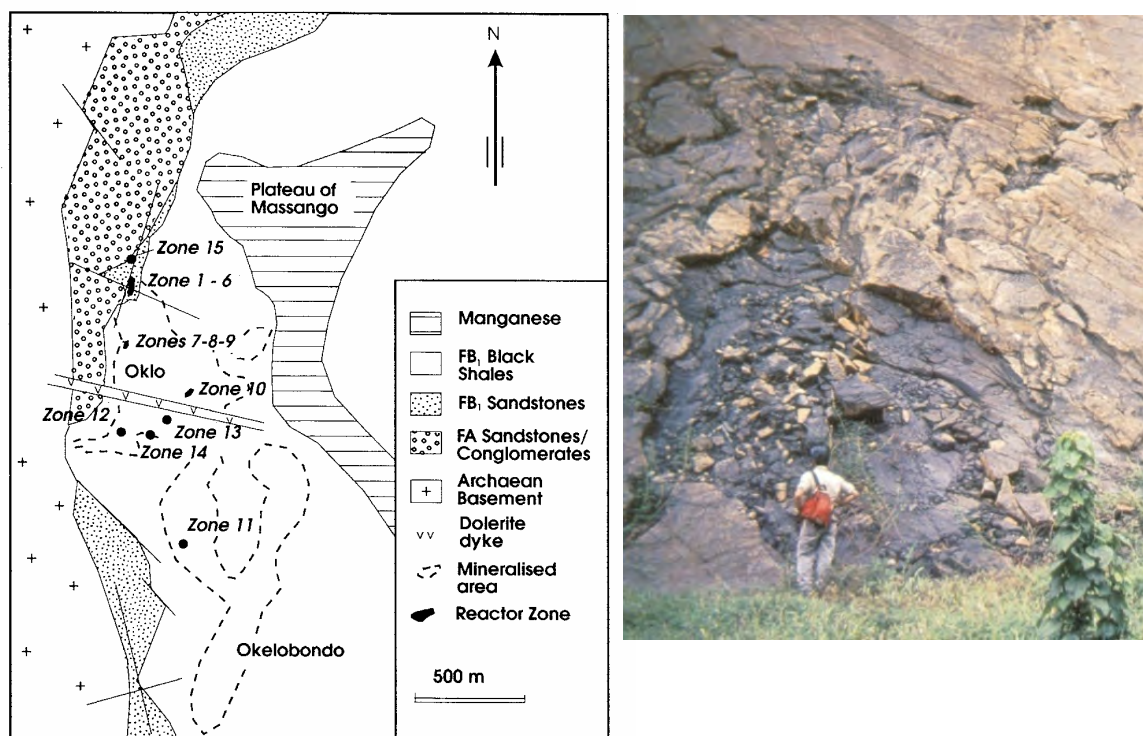


Figure 8-3. Left, simplified geological map of the Oklo and Okélobondo uranium deposits showing locations of fossil reactor zones 1–15 (from Gauthier-Lafaye et al. 1989). Right, imprint of a former reactor core (now mined out) on the underlying rock, showing the limited dimensions of these features (from Miller et al. 2000).

- Migration of elements was largely due to the circulation of hydrothermal fluids around the core of the reactors;
- The fission process appears to have been adversely affected or halted by the gradual or rapid collapse of the overlying strata resulting from the extensive and prolonged hydrothermal desilicification. This effectively reduced the porosity, closed off the source of the water (i.e. moderator) and, in some cases, disrupted the fissile U-235 material.

Although no other ‘natural reactors’ have been discovered to date, there are no physico-chemical reasons against their existence. Overall, this analogue study (which focused on the spent nuclear fuel stability analogy) has demonstrated that criticality due to uranium-235 transport outside the canister would require dissolution under oxidising conditions and deposition of uranium under reducing conditions. There is no credible mechanism to achieve both oxidising and reducing conditions in the near field of a deep geological repository in the long term. Criticality due to plutonium outside the canisters is also shown to be impossible because of the lack of a mechanism to dissolve and transport plutonium out of the canisters and then accumulate it in the necessary geometry under any reasonable disposal conditions. Thus, out-of-canister criticality has a vanishingly small probability of occurrence (Oversby 1996, 2000).

Similar conclusions were reached by SKB (2010), stating that the probability of criticality inside or outside the canister is considered to be negligibly small based on process, site and material understanding.

Even in an oxidising environment such as that around Yucca Mountain, Nevada, external nuclear criticality would not be a concern, in spite of the potential elevated percolation rate around the spent nuclear fuel packages, unfavourable material degradation rates favouring actinide release, and the high host rock porosity values (Nicot 2008).

As in the previous section, it is emphasised that the geometry of the fuel and its reactivity may be significantly different from uranium ores, thus criticality in relation to analogues is perhaps even less transferable than other processes.

8.2 Canister materials stability

The canister corrosion processes considered in the Finnish programme include:

- general corrosion under oxic and anoxic conditions;
- localised corrosion;
- microbially influenced corrosion;
- stress corrosion cracking (SCC).

A review of copper corrosion processes in conditions relevant to those at the Olkiluoto site has been compiled by King et al. (2011). Copper corrosion strongly depends on the redox conditions of the near field. During the early evolution, while oxic conditions prevail, rather high corrosion rates are expected (of the order of 10s of $\mu\text{m/a}$ initially). Soluble copper oxide corrosion products (mainly Cu_2O) will be formed and dissolved Cu(II) chloride and carbonate species will predominate. Localised corrosion will result

in a roughened surface rather than the classic pitting corrosion, according to observations and natural analogues. The corrosion rate will substantially decrease once anoxic conditions are established, at which point sulphide from groundwater becomes the main corrosion agent.

Detailed laboratory analyses have been performed relating to general and localised corrosion and these can be supported by natural analogue data. Studies carried out to date include Denison & Romanoff (1950), Tylecote (1979), Araki et al. (1989), Mitsui et al. (1996a,b), Sumiyama et al. (1997), Saheb et al. (2008) and Yoshikawa et al. (2008, 2009). Few data exist for the *long-term* behaviour of such materials, a review by the IAEA (2005) lists various archaeological analogue studies on EBS degradation, citing a total of four studies on iron/steel and seven on copper/bronze.

While microbially-influenced sulphate reduction is likely (e.g. Pedersen 2000, Pitkänen et al. 2004), studies so far have suggested that the environment in highly compacted bentonite is too extreme (i.e. the water activity is too low and the pore size of the clay at full compaction is too small) for sulphate-reducing bacteria (SRB) from the groundwater to survive and to actively produce sulphide within the buffer itself. Short-term URL experiments show very low microbial populations and activity in compacted bentonite (e.g. Pedersen et al. 2000a,b; Chi & Athar 2008). Natural analogue studies of microbial populations in natural bentonites are rare (e.g. Fukunaga et al. 2005), but they also indicate very low microbial populations (much lower than in soils or marine sediments), generally supporting the URL data, even though the physical form (density, porosity) of the natural bentonite differs from that in a repository. Thus, the most likely microbially-produced sulphide in the near field available to affect the canister is at the backfill-rock interface where lower bentonite density areas may persist. The sulphide produced in these peripheral areas has to diffuse through the buffer in order to reach the canister (see *Performance Assessment* for a fuller discussion).

SCC is believed to be unlikely because the maximum concentration of SCC agent, and the corrosion potential lie below the respective threshold values for SCC, or because the creep rate will exceed the crack growth rate (King et al. 2011).

Other potential corrosion products are being considered, such as copper corrosion by water in anoxic environment but so far the results do not support the claim that the canister durability will be significantly affected by this process, even if it were to occur (*Performance Assessment*).

8.2.1 Copper archaeological analogues

The Kronan Cannon

The Kronan was a Swedish warship built in 1668 which heeled over, exploded and sank (Figure 8-4) in June 1676 during the Battle of Öland. At the time, she was one of the largest warships in the world and was armed with 126–128 bronze cannon, most of which were salvaged in the 17th century. More were recovered from the mud into which they had sunk (Figure 8-5) in the 1980s (Figure 8-6) as part of one of the biggest archaeological studies in Swedish history (Lanitzki 1989).

One cannon in particular was investigated in detail as an analogue for the corrosion of the copper canisters planned to be used in the Swedish spent nuclear fuel repository design (Hallberg et al. 1987). This cannon had remained partly buried in a vertical position, muzzle down, in the mud since the ship sank.



Figure 8-4. Contemporary painting of the explosion by Danish artist Claus Möinichen.



Figure 8-5. Skull and bronze guns from the Kronan in situ (image © the Kronan Project and Bengt Grisell). Note muzzle down, vertically oriented cannon, right background.

The Kronan cannon had a high copper content (96.3 %) and the clay (which included montmorillonite), was tightly packed and water-saturated. Due to the ingress of oxidants from the seawater above, the mud porewaters nearest the top of the sediments were more strongly oxidising. Corrosion products include Cu_2O , confirming a generally oxidising environment. A corrosion rate of $0.15 \mu\text{m/a}$ was calculated (see Table 8-1), and was constant over the cannon surface, although inclusions of CuO slag in the bronze weathered more rapidly (Hallberg et al. 1987). That the cannon had suffered only minor corrosion, despite the oxidising conditions, increases confidence in the suitability of copper as a canister material. Indeed, the corrosion of a copper disposal canister should be slower than that of the cannon due to the less harsh repository environment (reducing rather than oxidising) and higher quality copper production (no inclusions).

Overall, this is a good archaeological analogue of copper corrosion and, although there are important differences between it and the repository (e.g. composition and compaction of the clay, porewater composition etc.), other information such as the known age of the cannon's burial make this a convincing dataset.

King's (1995) re-analysis of the original work on the Kronan cannon is also worthy of mention. As noted above, the cannon was originally used as supporting evidence for SKB's copper canister design but, in this study, the data were re-examined in the light of the Canadian design for a spent nuclear fuel repository and the original data were compared with more recent laboratory experiments. It was shown that a corrosion mechanism developed from the results of short-term copper corrosion experiments could be used to describe the observed reaction sequence on the cannon. Despite this being an ideal study where the relevance of the archaeological analogue data to the long-term behaviour of a copper canister (and associated clay) is clear and where a corrosion pathway which is relevant to various coastal sites (i.e. with brine groundwaters) was produced, neither the new data nor the new model have been used in any safety assessment. Nevertheless, both the original analysis by Hallberg et al. (1987) and the re-analysis by King (1995) support the proposed long lifetime of a copper canister under likely repository conditions.



Figure 8-6. One of the cannon on the deck of the dive ship, immediately after recovery (image courtesy of Kalmar County Museum/The Kronan Project).

Other examples

In both the Japanese H12 and H17 safety assessments (JNC 2000a and 2005, respectively), corrosion data from archaeological analogues were used to increase confidence in the data produced in short-term laboratory corrosion tests for copper canisters (see Table 8-1). To these data, additional information that was not included in the H12 assessments, or which did not exist at the time, have been added from other archaeological analogue studies. The newer data from the IAEA (IAEA 2005) suggest that the assumptions made in H12 and H17 are certainly conservative.

Localised corrosion has been covered to a much lesser degree, with the studies of Bresle et al. (1983) and Mattson (1983) being two rare examples. As noted in Miller et al. (2000) *“The copper and copper alloy materials used in these studies included several archaeological artefacts such as Roman coins and vases, objects from the Swedish Bronze Age, 17th Century coins, buried lightening conductor plates and lumps of native copper. The objects were all of different age, came from a wide range of environments and had differing compositions. All samples (excluding the native metal) indicated pitting factors of less than 3; the native metal had a pitting factor of 2 to 6. Comparison with pitting factors used in performance assessment (a factor of 25 was used in the KBS-3 assessment) would suggest that the performance assessment values are overly conservative.”* Pitting factors are no longer used in safety assessment; rather, a corrosion allowance of about 100 micrometres is taken into account due to a “surface roughening” effect, rather than pitting.

Table 8-1. Comparison of copper corrosion depths cited in H12 with existing and new archaeological analogue data for copper/bronze artefacts.

Form of data	Corrosion depth (per 1000a)	Reference	Comments
Short-term lab	13 mm	JNC (2000a)	Uniform corrosion of copper
Archaeological analogue	<3 mm	Range of studies cited in JNC (2000a)	Uniform corrosion of copper and bronze
Archaeological analogue	0.26–39 mm	Bresle et al. (1983)	Pitting corrosion of copper
Archaeological analogue	0.025–1.27 mm	Tylecote (1979), Johnson & Francis (1980)	Uniform corrosion of mixed artefacts
Archaeological analogue	0.15 mm	Hallberg et al. (1987)	Kronan cannon
Archaeological analogue	0.13–1.13 mm	Appendix A in IAEA (2005)	Bronze artefacts from a river
Archaeological analogue	<0.27 mm	Appendix B in IAEA (2005)	Bronze artefacts in soils
Archaeological analogue	0.4–1.2 mm	Appendix D in IAEA (2005)	Copper artefacts in flood plain soil
Archaeological analogue	0.01–1.91 mm	Appendix D in IAEA (2005)	Bronze artefacts in flood plain soil

Overall, if data from less relevant environments (e.g. oxidising soils etc.) are excluded, the results broadly support the thesis that copper canisters will be long-lived. However, as noted in Miller et al. (2000), “Further analogue studies on metal corrosion processes and rates are probably only warranted if more relevant systems can be identified. In particular, metal compositions should be as close as possible to canister metals.” This remains the case today.

8.2.2 Copper geological analogues

Elemental (or ‘native’) copper has persisted for millions of years in a range of geological environments such as (Marcos 1989):

- In sedimentary rocks: Keweenaw Peninsula, Lake Superior region, Michigan, U.S.; Corocoro, Bolivia; South Devon, United Kingdom;
- In basaltic lavas: Keweenaw Peninsula; Appalachian States from central Virginia to southern Pennsylvania, U.S.; Coppermine River area, NWT, Canada; Dalane, Norway;
- In granitic rocks: Hyrkkölä and Askola, Finland;
- In the oxidised zones of sulphide deposits (many places in the world, including Finland, Chile, Japan etc.).
- Here, two examples are presented to show the durability of native copper in a range of environments.
- Littleham Cove Native Copper
- Native copper from the Permian Littleham Mudstone Formation, at Littleham Cove, in South Devon in southwest England (Figure 8-7), has been studied as a copper natural analogue. The copper is 99.9 % Cu, occurring as plates up to 160 mm diameter and up to 4 mm thick (Figure 8-8), closely associated with uraniferous-vanadiferous concretions and reduction spots within red mudstone. Each plate is a composite of several stacked thin sheets of copper, varying in thickness from <0.1 to 2 mm.

The native copper has suffered some alteration and corrosion, which can be broadly divided into four stages (Milodowski et al. 2000, 2002):

- Stage 1: early oxidative alteration, during which the copper was corroded and partly replaced by copper oxides;
- Stage 2: involved subsequent overgrowth and partial replacement of the copper and copper oxides by a complex assemblage of copper, nickel, cobalt and iron sulphides and arsenides;
- Stage 3: after initial diagenetic reaction, the native copper sheets were then preserved within the clay matrix of the Littleham Mudstone Formation without further alteration for over 170 Ma, until uplift and erosion exposed the copper plates to the current weathering environment;
- Stage 4: following uplift and exposure on the current surface, very late-stage oxidation alteration of the native copper, sulphide and arsenide minerals to secondary copper oxyhydroxide, copper carbonate and copper sulphate minerals.

In this particular case, after early corrosion and alteration of copper during burial diagenesis, the residual copper (representing an estimated 30–80 % of the original copper mass) effectively remained inert and isolated from further corrosion within the

naturally-compacted mudstone matrix for at least 170 Ma, until uplift and erosion exposed the native copper to alteration by the present near-surface weathering environment. Unlike a purpose-built bentonite clay barrier in a repository, the natural clay matrix of the Littleham Mudstone Formation is not smectite-rich nor has it been compacted to ensure a low permeability buffer around the copper. Nevertheless, the copper survived for much longer than is required in the Finnish repository design, providing further indication of the likely durability of a copper canister.



Figure 8-7. Littleham Cove in Devon, southwest England (Milodowski et al. 2002).

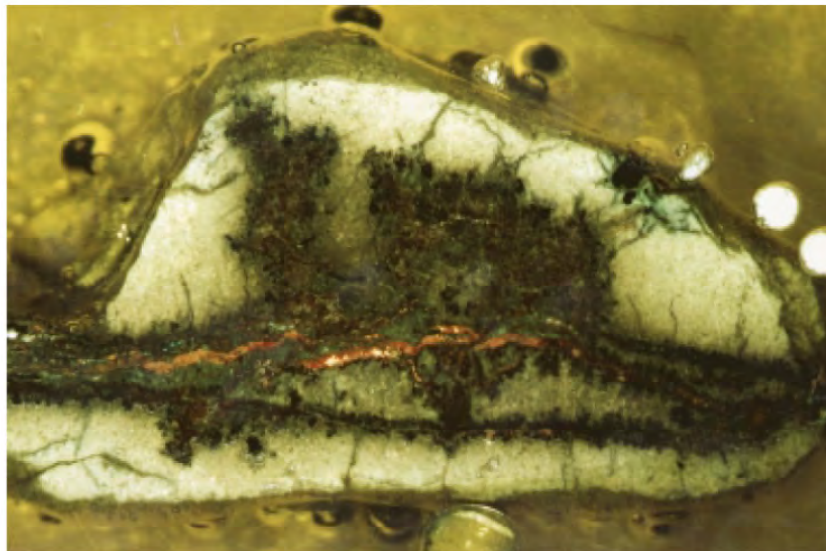


Figure 8-8. Native copper embedded in sediment (nodule ca. 6x8 cm) (Milodowski et al. 2000).

Hyrkkölä and Askola, Finland

At the 1700 Ma U-Cu mineralisation at Hyrkkölä, near the Palmottu natural analogue site in Southern Finland, native copper and Cu sulphides occur in open fractures in crystalline rocks (Marcos & Ahonen 1999). This has allowed the study of sulphidation and corrosion processes under conditions somewhat analogous to a repository.

The Hyrkkölä analogue shows that, in the past, corrosion in both oxidising and reducing conditions has occurred. Corrosion in reducing conditions by sulphide ions (sulphidation) is identified as the key canister corrosion process during the post-closure evolution of the repository. Although sulphidation is no longer occurring in the Hyrkkölä analogue, the mineral assemblage gives insights to the conditions in which it occurred. Most of the copper remained in its native state, indicating a slow copper corrosion rate, although the duration of sulphidation is not known. Oxidation in contact with groundwater containing dissolved oxygen (0.4 to 4 mg/L) is still ongoing. The process started at least 10,000 to 100,000 years ago (based on indirect observations from other fracture-filling materials) and has been shown to proceed at a very slow rate, possibly due to passivation mechanisms (see e.g. Figure 8-9).

8.2.3 Iron analogues

The main perturbations potentially associated with the massive iron insert are hydrogen gas production due to anaerobic corrosion and redox changes around the damaged copper-iron canister (e.g. Nagra 1994). As a check on the long-term performance of an iron or steel as potential canister materials, both Nagra and JNC (now JAEA, Japan Atomic Energy Agency) carried out natural and archaeological analogue studies of iron artefacts from a range of environments (Figure 8-10) that can be applied to the corrosion processes of the iron insert.

Despite the fact that most material studied came from oxic to sub-oxic environments and would therefore be expected to corrode to a much greater extent than in an oxygen-free repository environment, a maximum corrosion depth of 10 mm in 1000 years was calculated by Nagra (Figure 8-11), so increasing confidence in the results from the short-term experimental data and in the conservatism of the Base Case corrosion depth of 29 mm (Table 8-2). For JNC, a maximum corrosion depth of 15 mm in 1000 years was calculated, again comparing well with the Base Case assumption of 31.8 mm in H12 (Table 8-2). The maximum corrosion depth was dropped to <10 mm in JNC (2005), even though this included data from aerobic environments (Figure 8-12).

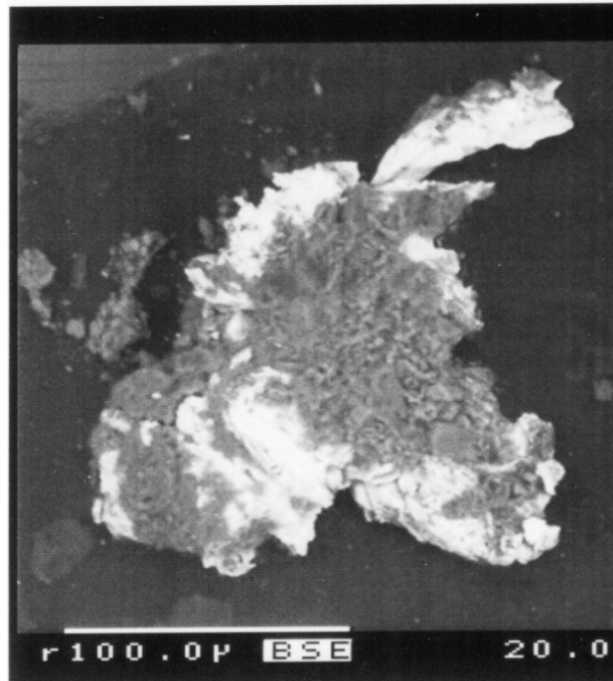


Figure 8-9. Native copper plate covered with smectite from a groundwater-conducting fracture at Hyrkkölä, SW Finland (Marcos 2002).



Figure 8-10. Nagra used a range of archaeological analogues to provide supporting evidence for their safety assessment Base Case corrosion depth. Left, 2000 year old Roman nails which were disposed in a pit (right) as the Romans abandoned Inchtuthil, Scotland, the most northerly Legionnaires Fortress in the Roman Empire. Courtesy of Nagra.

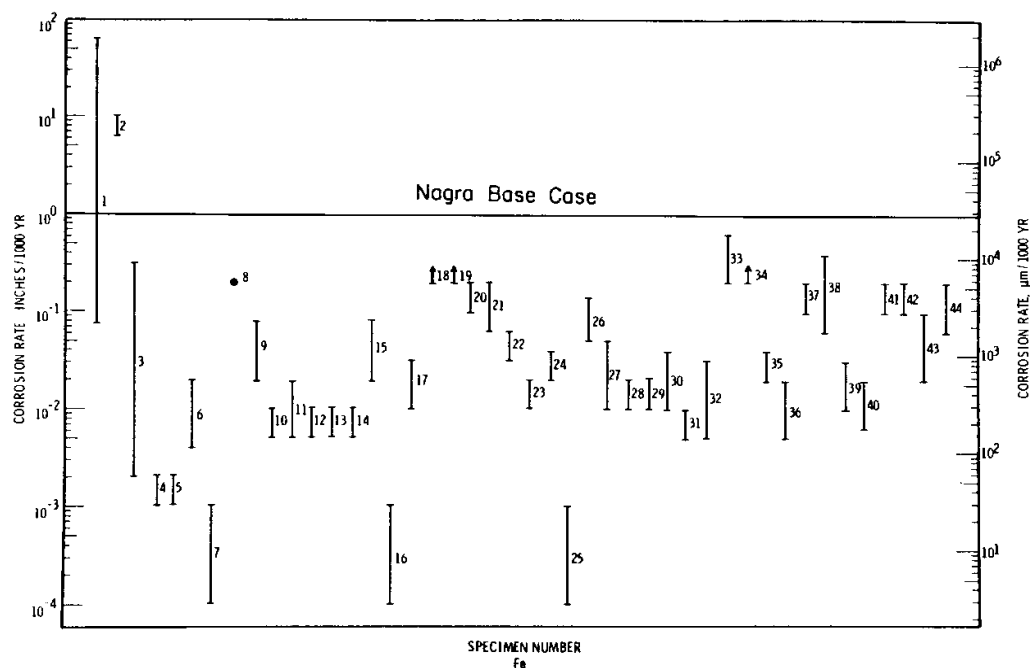


Figure 8-11. Corrosion rate data from natural and archaeological analogue studies (from Miller et al. 1994). The corrosion rates for the archaeological artefacts range from 0.1 to 10 $\mu\text{m/a}$ (note that samples 1 and 2 are from oxidising marine conditions; details of all other samples included in Johnson & Francis 1980). The Nagra base case corrosion rate for steel canisters from Projekt Gewähr is also shown for comparison.

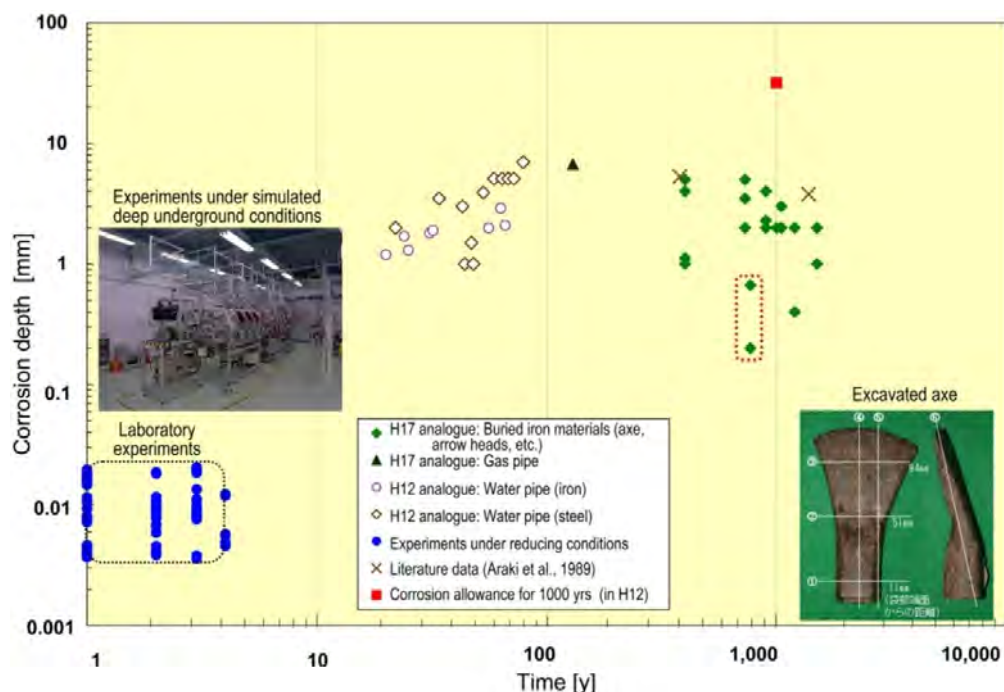


Figure 8-12. Integration of iron/steel corrosion data from laboratory experiments and several natural analogue sources (details in JNC 2005). Note that the H17 analogues surrounded by the red dots are believed to have come from an aerobic environment.

Table 8-2. Comparison of steel corrosion depths cited in the H12 (JNC 2000a), Kristallin-1 (Nagra 1994) and H17 (JNC 2005) safety assessments with a range of archaeological analogue data for steel/iron artefacts.

Form of data	Corrosion depth (per 1000 a)	Reference	Comments
Short-term laboratory	31.8 mm	JNC (2000a)	Uniform corrosion of carbon steel. Base Case value
Short-term laboratory	29 mm	NWGCT (1984)	Conservative corrosion rate, including an allowance for pitting. Base Case value
Natural analogue	$0.09 \cdot 10^{-3}$ mm	Hellmuth (1991)	Weathering of native iron in basalt (Disko Island). These are very low values which may reflect the site complexity. See discussion in Hellmuth (1991) and Miller et al. (2000).
Archaeological analogue	10 mm	Range of studies cited in Nagra (1994)	Uniform corrosion of iron and steel
Archaeological analogue	<15 mm	Range of studies cited in JNC (2000a)	Uniform corrosion of iron and steel
Archaeological analogue	0.1 – 10	David (2001)	Literature review of archaeological samples
Archaeological analogue	<10 mm	Range of studies cited in JNC (2005)	Uniform corrosion of iron and steel

Conclusions

Both archaeological and natural analogue studies suggest that the corrosion rates for copper and steel assumed in the safety assessment are generally conservative (and this is generally supported by long-term laboratory experiments, e.g. Smailos et al. 2004). This is especially the case when it is considered that the majority of materials studied to date have come from environments that are likely to have been more oxidising than is expected in the repository near field. The available studies of localised corrosion of copper suggest that the assumed corrosion allowance is also conservative. To date, no natural analogue studies of localised corrosion of iron/steel are available.

8.3 Buffer and backfill analogues

Studies of the long-term stability and behaviour of buffer and backfill material (bentonite and other swelling clays) have focused on a range of issues, including:

- Thermal alteration of the bentonite by heat from the canister;
- Mechanical deformation (e.g. from canister sinking and tilting, fracturing or gas out-bubbling);
- Chemical alteration from saline water or brines, cementitious leachates or canister corrosion products;
- Hydraulic barrier function;
- Freezing and thawing;
- Chemical erosion.

Laine & Karttunen (2010) have reported a wide-ranging review of bentonite natural analogues so here, once again, only a few, relevant examples are given along with novel data on potential chemical alteration of bentonites. Unfortunately, only a few of these issues are truly amenable to natural analogue studies and so few useful data exist.

8.3.1 Thermal alteration

In the case of thermal alteration, cementation of the bentonite can occur due to illitisation, where silica is released at higher temperature (and later re-precipitated). Miller et al. (2000, 2006) and Laine & Karttunen (2010) note a significant number of natural analogue papers in the literature, but most examples suffer from the same problem, namely the conditions are not truly analogous to those of a repository. In most cases of contact metamorphism studied, the temperatures have been much too high (800–900 °C rather than the repository relevant maximum of 100 °C in the Finnish design, cf. Figure 8-13). In the case of diagenetic illitisation, a number of natural analogue studies have been carried out in the Gulf of Mexico, USA (e.g. Eberl & Hower 1976, Robertson & Lahann 1981), and elsewhere (Pusch & Karnland 1988). Although the conditions are not truly analogous to those of the repository environment, these studies suggest the illitisation rate in the natural environment is considerably slower than that predicted by kinetic models, due to the fact that the process depends on the rate of supply of potassium, which may often be limited, as is likely to be the case at Olkiluoto.

Pusch & Karnland (1988) investigated bentonite from the Busachi site in Sardinia (Figure 8-14) and ran a series of calculations to assess the likely temperature at different places in the bentonite following burial by rhyolitic lava (Table 8-3). Based on these observations and calculations, they provided evidence that significant heat-induced dissolution of smectite occurred at 150 to 200 °C (i.e. much warmer than is likely in the buffer at Olkiluoto) and precipitation of siliceous material occurred during cooling. This siliceous cementation was found to have measurably affected the rheological properties of the bentonite, in a manner which might adversely affect the containment of radionuclides if it occurred in a repository environment. However, the smectite content was never less than 60 % in any part of the bentonite. The authors concluded that the rate of alteration was primarily dependent on temperature up to about 150 °C, with the supply of potassium from the groundwater as the rate determining factor.

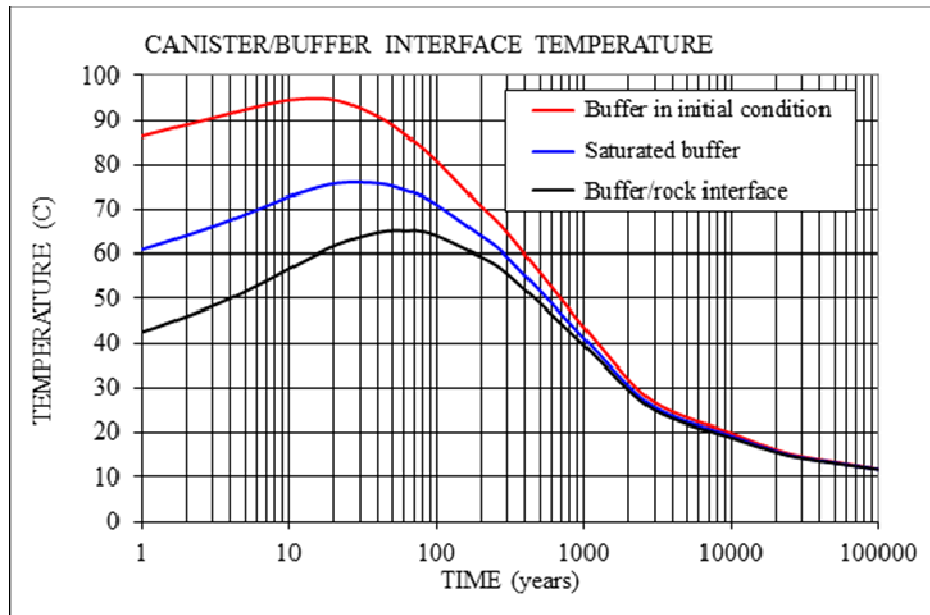


Figure 8-13. Canister surface temperature estimates and temperatures at the buffer-rock interface in the repository (central area) as a function of time since emplacement using the two extreme saturation degrees for the bentonite buffer. OL3 canister, average burnup 50 MWd/kgU, canister distances in repository 10.5 m (between deposition holes)/ 25 m (between deposition tunnels), buffer conductivity is 1.0 W/m/K in the initial condition and 1.3 W/m/K in saturated condition. In the initial condition, there is a 10 mm air gap between the canister and the buffer, in the saturated condition the gap is closed. The outer 50 mm gap between buffer and rock is assumed to be filled with bentonite pellets that have conductivity of 0.2 W/m/K in the initial condition and 0.6 W/m/K when saturated. Based on the results of Ikonen & Raiko (2012).

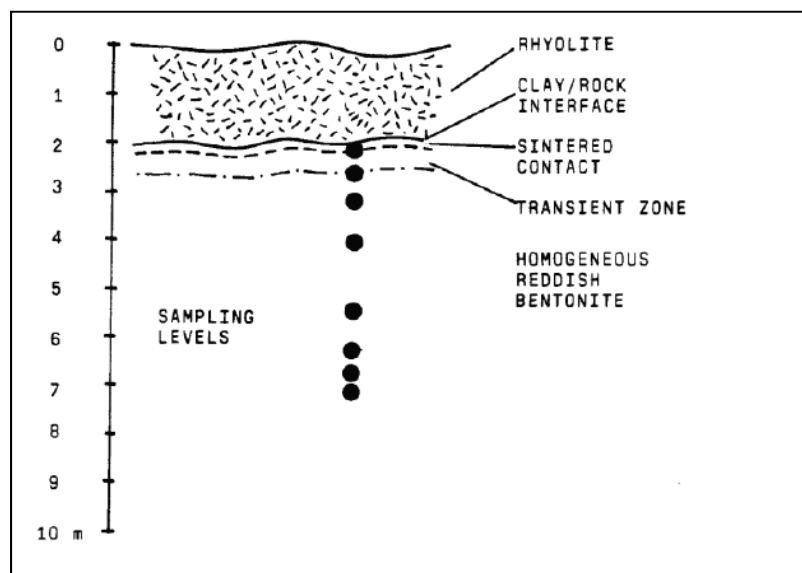


Figure 8-14. Stratigraphy of the Busachi site, Sardinia, showing sample positions (Pusch & Karnland 1988).

Table 8-3. Calculated temperature profile for the Busachi bentonite (based on three different boundary conditions). From Pusch & Karnland (1988).

Depth below rock boundary [m]	Dry crust case T (°C)	Wet case (30 °C) T (°C)	Wet case (75 °C) T (°C)
0.2	800	750	700
0.7	300	280	250
1.2	170	155	150
2.1	80	65	60
3.5	50	35	25
4.3	35	25	20
4.7	25	20	20
5.1	20	20	20

Key:

Dry crust case: assumes the bentonite was dry when engulfed by the rhyolite

Wet case 30 °C: assumes the bentonite was under seawater at 30 °C when engulfed by the rhyolite

Wet case 75 °C: assumes the bentonite was under seawater at 75 °C when engulfed by the rhyolite.

Pusch et al. (1987) studied seven natural clays with smectite contents ranging up to 25 %. Although these clays ranged in age up to several hundred million years old and had experienced slow mineralogical alteration processes in relatively near-surface active environments, most still possessed swelling and rheological properties which would be adequate for buffer performance. As such, this qualitatively suggests that the bentonite buffer in the relatively stable, low flow conditions of a repository near field would be likely to retain its barrier functions for at least similar periods of time.

Interestingly, two of the clays studied by Pusch et al. (1987) showed indications of cementation, something which could decrease the bentonite's ability to self-seal if a sufficient mass of the buffer was affected. Several natural analogue studies (e.g. Pellegrini 1999, Woods et al. 2000) have since tried to address this point by examining bentonite cementation. Here, smectite-bearing clays which had been penetrated by dykes were examined and, at distances from the dyke where the temperature gradients were repository relevant, mineralogical changes were associated with subsequent fracturing of the cemented clays as the interstitial waters were driven out. Retrograde or 'back' reactions during the temperature decrease following the thermal event were suggested as the reason why new smectite was produced. Unfortunately, these observations cannot directly be applied to safety assessment because the full thermal, chemical and pressure histories experienced by these clays have not yet been adequately characterised.

Laine & Karttunen (2010) reported on the Ishrini bentonite body in Libya (Kolaříková & Hanus 2008). Here, the impact of a basalt dome and dykes on the bentonite has been assessed by examining isotopic and mineralogical changes. It would appear that the minimum temperature experienced by the bentonite was probably >190 °C and bentonite from outside this zone would appear not to have been sampled. In conclusion, Laine & Karttunen (2010) noted that the probable impact of raised temperatures in the bentonite in the deposition hole is minimal (i.e. some local cementation occurs, but the

majority of the bentonite remains unaltered), but that a truly representative analogue has not yet been examined.

8.3.2 Mechanical deformation

With respect to bentonite deformation, again, few data exist in the natural analogue literature because it is basically very difficult to find an appropriate natural analogue. The issue was discussed as far back as 1994 (NAWG 1996) and highlighted again more recently in Miller et al. (2000) but, despite this, all attempts so far to address the issue have failed. In one rare report on the process, Keto (1999) examined two bentonite deposits (from the MX-80 source in Wyoming and from FEBEX bentonite in Almeria, Spain; Figure 8-15) which are planned to be used in actual repository programmes (rather than just a generic bentonite). This report is an academic study of existing literature on bentonites along with the production of new data on mineralogy and water content allied with field observations on bentonite deformation and weathering. An attempt is made to tie in the work to the behaviour of bentonite during canister sinking, but most of the results are no more than qualitative.

The evidence of bentonite plasticity is interesting, but this topic is not treated quantitatively enough to warrant use in safety assessments.

8.3.3 Chemical alteration

In the area of chemical alteration of bentonite, Villar et al. (2006) noted that, for two particular sites, Cala de Tomate and Cortijo de Archidona deposits (Figure 8-15), post-formational reaction of the bentonite with seawater had changed the presumed original Ca-smectites to Na-Mg smectites following seawater interaction. Interestingly, where meteoritic waters had then interacted with the upper part of the deposits, the Na-Mg smectites became Mg-Ca-(Na) smectites. Frustratingly, no information is provided as to whether these changes had any subsequent effects on the bentonite other than, at Cortijo de Archidona, where it is stated *“In any case, the bentonite has preserved a high concentration of solutes and its mineral stability”* but no data to support this point are reported. Martinez et al. (2007) also reported an alteration front in bentonite at the El Toril site (Figure 8-15), but concluded that it probably represented meteoric water alteration (with possible dissolution of pyrites in the bentonite).

Few other natural analogue studies have attempted to look at saline/brine water alteration of bentonite. In one rare example, Smellie (2001) examined Wyoming bentonites from a basin where it is believed the deposit had slowly become more saline over several million years, finally becoming a brine. Despite this, there is no evidence (either mineralogical or geochemical) for any post-depositional alteration of the bentonite. It was shown that the Wyoming bentonite had, in fact, been isolated from interaction with marine waters by thick, relatively impermeable marine clays. However, it should be noted that, to date, the bentonite/impermeable clay boundary has not been examined in detail for any indication of diffusive fluxes of Na, Cl etc. across the boundary. Although this boundary shows no visible alteration, possibly indicating that the bentonite is, in fact, resistant to saline water interaction, however, this observation has not been checked analytically. Another example is that of Karnland et al. (2004), who studied bentonite from San José (see Figure 8-15 above) as a natural analogue exposed to high saline groundwater. The observed smectite structure and the measured

properties of the San José bentonite were similar to those found in Wyoming bentonite and no obvious effects of the harsh saline environment were observed.

Interaction between the bentonite and the canister could cause a loss of bentonite isolation properties as, for example, metal ions from the canister exchange with cations in the clay. In the case of a copper canister, Pusch (1982) showed that Cu (II) could potentially replace all the sodium in bentonite, so increasing the bentonite permeability¹⁵. An attempt was made to evaluate this process by examining the sediments around the Kronan cannon (Section 8.2.1). Here, Hallberg et al. (1987) showed that copper from the cannon had diffused 4 cm into the clay over the 300 years of contact (see Figure 8-16). Unfortunately, there is little information regarding the actual products of the copper interaction with the clay.

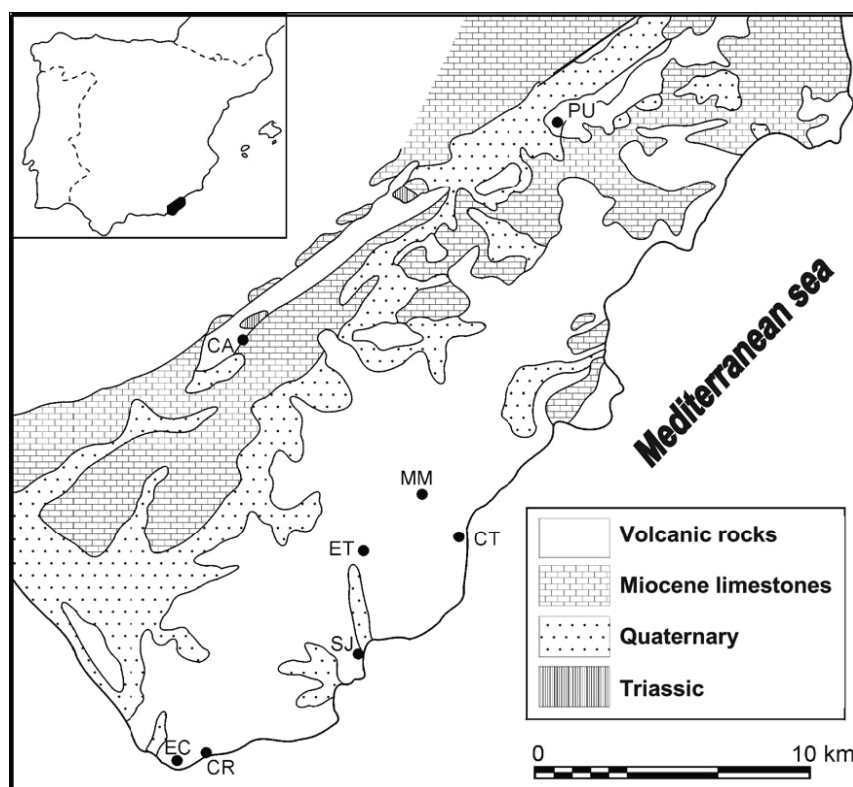


Figure 8-15. Geological map of the Cabo de Gata region, southern Spain, showing the bentonite deposits selected for ENRESA's natural analogue studies: PU: Pozo Usero, MM: Morrón de Mateo, CT: Cala del Tomate, ET: El Toril, SJ: San José, EC: El Corralote, CR: Cala Rajá, CA: Cortijo de Archidona (from Villar et al. 2006).

¹⁵ In the case of Cu (I), solubility control on the copper will probably limit the amount of copper available for uptake on the bentonite.

Interestingly, King (1995) compared the copper diffusion (in bentonite) profiles from 3 laboratory 180 day experiments with the Kronan cannon diffusion data from Hallberg et al. (1987, 1988) and noted they were very similar, despite differences in experimental duration and temperatures. It was also noted that the apparent diffusion coefficient of copper in the clays was highly temperature dependent (although this is contradicted by Carlsson 2008), possibly due to the temperature dependent reduction of Cu (II) to Cu (I) by iron present in the bentonite. However, other than noting that “*Compacted clay...is shown to severely limit both the supply of O₂ to the corroding surface and the rate of diffusion of dissolved Cu (II) away from it*”, no other analysis of the impact of the copper on the properties of the clay was attempted. In the recent study summarising the available reported information on the possible interaction of copper on bentonite King & Wersin (2012) concluded that the impact of Cu(I) on the performance of the bentonite buffer is expected to be very limited. Diffusion experiments reveal that the interactions of Cu(I) with the clay are much weaker than those of Cu(II). Further support for weak or no uptake of Cu(I) is provided by chemical analyses of natural bentonites. Some interaction processes between Cu(I) and the clay, e.g. the possibility of oxidation to Cu(II) by structural Fe(III) and the concomitant reduction of Fe(III) to Fe(II), cannot be ruled out.

In the case of iron-bentonite interactions (note the iron insert in the canister), useful natural analogue data are very limited (cf. Marcos 2003; Wersin et al. 2007), but Marcos (2004) assessed a site (Cortijo del Aire, Spain) where bentonite was affected by iron-rich solutions coming from faults or other discontinuities (Figure 8-17).

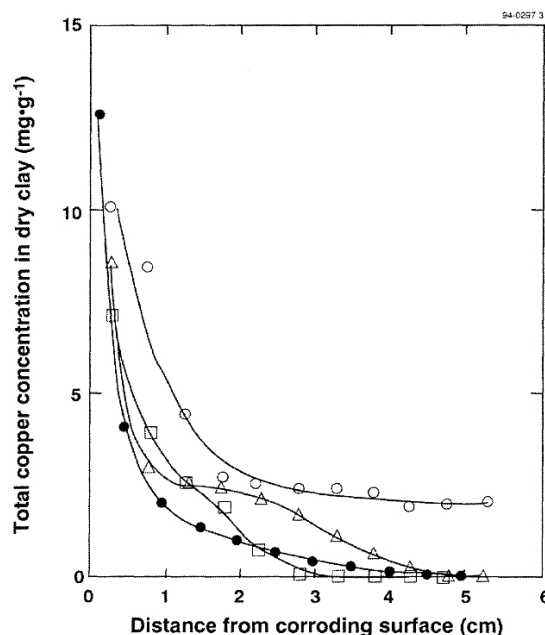


Figure 8-16. Comparison of copper concentration profiles in sea sediments adjacent to a submerged bronze cannon and a profile observed experimentally in compacted bentonite-sand buffer material in contact with a copper coupon (from King et al. 2001). The sea-sediment profiles (open symbols: O, □ and Δ) developed over a period of 310 years at a mean ambient temperature of 7 ± 5 °C compared with an exposure period of 180 days at 100 °C for the buffer material profile (●).

Marcos (2004) noted that, in the samples analysed, the content of iron was greater (almost two-fold) in the redder samples than in the greyer ones. The origin of the hematite pigment was unclear (although migration of structural Fe from smectite was noted to be a possibility). However, apart from a preliminary reconnaissance, no further work was conducted on the samples as the smectite content of the bentonite was deemed to be too low (maximum 15 %) for the material to be of relevance to the Olkiluoto design.

More recently, Fukushi et al. (2010) reported alteration of bentonite at the Kawasaki bentonite deposit in Zao, Japan (Figure 8-18). The bentonite consists of Na-Ca smectite with subordinate opal-CT, quartz and zeolite (Takagi et al. 2005) and includes greenish hydrothermal veins. They are composed of mixed-layer minerals consisting of smectite and glauconite, pyrite and opal. The mineral assemblages of the greenish veins and their surroundings indicate that the hydrothermal activity had most likely taken place at a temperature of less than 100 °C and that the pH and Eh conditions of the reacted solution were neutral to alkaline pH and reducing. The bulk (unaltered) bentonite is composed mainly of Al smectite and opal whereas the bentonite in contact with the greenish veins appears mineralogically and petrographically altered and consists of discrete opal grains and dioctahedral Al smectite containing Fe.

The authors contend that the conditions of alteration are similar “...to those in the geological disposal environment...” and therefore were worth study. However, although superficially of relevance, it should be noted that the secondary clay and opal were formed by dissolution and subsequent precipitation from the interaction of the original bentonite with the hydrothermal solution. This is not what would be expected in the Olkiluoto EBS and, as such, this occurrence is not considered further here.



Figure 8-17. View of the bentonite sampling site at Cortijo del Aire, Almería, Spain (left). The red staining of the bentonite is clear during sampling (right). From Marcos (2004).

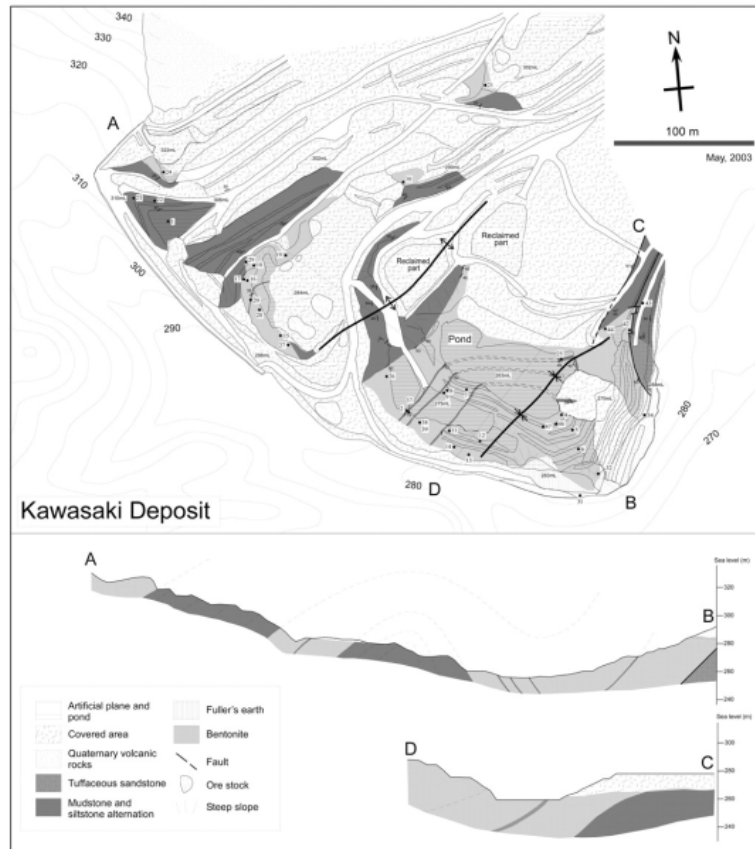


Figure 8-18. The Kawasaki bentonite deposit, Zao, Japan (from Takagi et al. 2005).

8.3.4 Hydraulic barrier functions

The bentonite should isolate the canister from the host rock groundwaters and act as a colloid (and microbe) filter for very long time periods and there are several natural examples which show very well that this is possible. The best known is the 2 Ma preserved Sequoia trees at Dunarobba, near Todi in central Italy (Figure 8-19). In the Dunarobba and Cava Topetti quarries, the trees were trapped in their upright position by a flood of lacustrine clay which completely enclosed them. Above the clay is a layer of coarse sand and there is a large difference in hydraulic conductivity between them, with the clay layer having a hydraulic conductivity in the range $2 \cdot 10^{-13}$ to $2 \cdot 10^{-10}$ m/s compared with an estimated hydraulic conductivity for the sand deposits of 10^{-4} m/s (Lombardi & Valentini 1996). This hydraulic contrast has almost certainly minimised ingress of oxygenated groundwaters into the clay, so protecting the wood from lithification.

The wood remains in almost perfect condition and can be cut and burned as though it had been only recently felled. Organic leachates from samples of the Dunarobba wood were compared with leachates obtained from modern oak, oak from the Roman period and 150 Ma old lignites (Chapman 1990). The total organic carbon (TOC) content produced from the Dunarobba wood was comparable to the steady state values from cellulose breakdown in the modern oak sample, clearly showing how well preserved are the Dunarobba trees.

Another, illustrative, example of the isolation capability of clays is that of the 2100 year old cadaver from a Chinese tomb (Lee 1986). Here, a series of barriers (not unlike a repository EBS) including clay (Figure 8-20) preserved the body of Xin Zhui, who died between 178 and 145 BC. Xin Zhui was not mummified before being buried and her tomb included silk, wood, meat and vegetables.

After exhumation, the cadaver (Figure 8-21) was found to be well-preserved, with the skin complete and retaining some of its elasticity whilst the abdominal organs were intact and some of the joints were partially movable. The burial objects were in an equally good state of preservation, the meat and vegetables showing only partial decomposition, and this has been attributed to the thick clay layer around the coffin (Figure 8-20) providing an air-tight seal.



Figure 8-19. The Dunarobba Sequoia forest, Italy. The massive trees were preserved upright by a sudden flood of lacustrine clay (courtesy Chris Weiss).

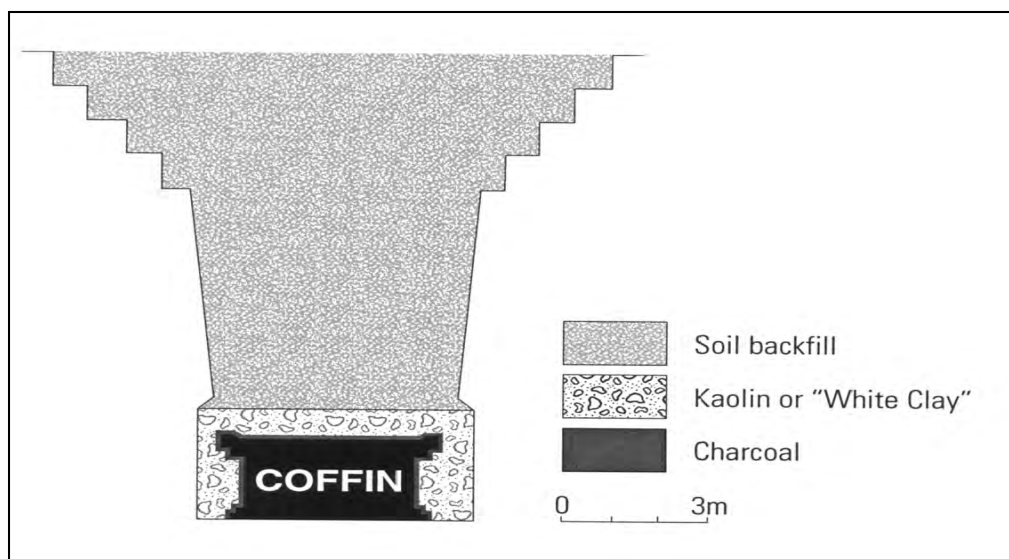


Figure 8-20. Cross-section of the Xin Zhui tomb (after Lee 1986).



Figure 8-21. *The extremely well-preserved cadaver of Xin Zhui.*

8.3.5 Freezing and thawing

In the USA there are several deposits of bentonite being commercially exploited (see e.g.: <http://geology.com/news/images/bentonite.gif>). The Wyoming bentonites, of which one product is MX-80, are located near the border of the Laurentide Ice sheet (Figure 8-22) and as such, the area was subject to permafrost periods during the last ice age and also during previous ice ages (e.g. Mears 1981 and references therein).

The overburden covering the bentonite deposits varies in thickness from less than one to four or more metres, and thus part of the bentonite deposits were within the active layer that freezes and thaws every year. Over the last 740,000 years eight glacial cycles are known to have occurred in North America (and the Northern Hemisphere), thus the bentonite deposits have been subjected to a great many freeze-thaw cycles over this period.

In addition, laboratory experiments on freezing and thawing of compacted bentonite have shown that there are no major consequences for the behaviour of bentonite after even more extreme temperature variations that can be found in nature (Schatz & Martikainen 2010).

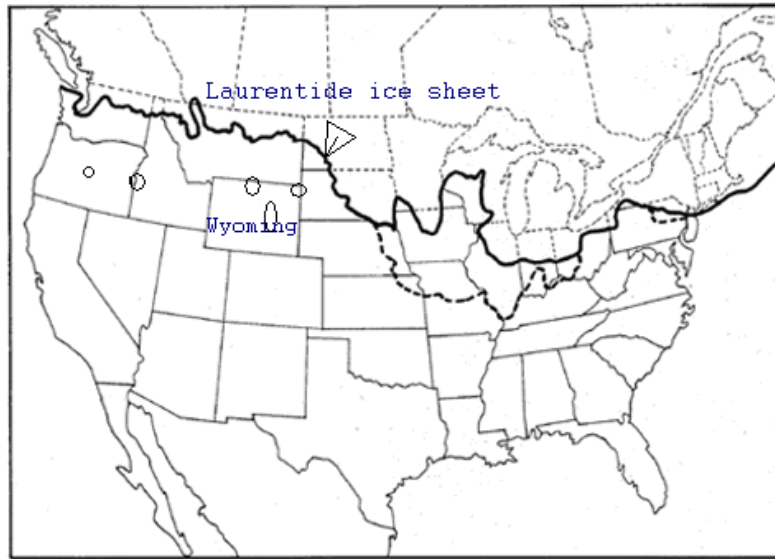


Figure 8-22. The extent of the Laurentide ice sheet and position of bentonite deposits in Wyoming and Oregon permafrost areas (modified from figure retrieved from website of <http://www.uleth.ca>).

8.3.6 Chemical erosion

Chemical erosion of bentonite is the erosion (i.e. mass loss of bentonite) caused by the interaction of low ionic strength (dilute) water with bentonite (see process 5.2.4 in *Features, Events and Processes*). The presence of soluble calcium minerals, e.g. calcium sulphate (gypsum), will put Ca ions into solution, which hinders the formation of colloids from the smectite. Other less soluble materials such as quartz and feldspar will be left behind as the smectite is lost and this residue may act as a filter in thin fractures to slow down further smectite loss. There are no natural analogues as such to support or discount this process, although it has been observed experimentally (e.g. Neretnieks et al. 2009), but it is certainly related to the availability of dilute water at repository depth (see Section 7.3.2 of this report). It should be noted that chemical erosion does not occur without active water flow supplying the dilute waters and removing the chemically (and possible physically) eroded particles of bentonite. In spite of the lack of strict natural analogues, it is possible to use observations of natural occurrences of bentonite to assess the extent of occurrence of this process.

It is well known that weathering generally improves bentonite quality (e.g. Elzea & Murray 1990), reducing the smectite particle size and “cleaning up” impurities. Thus it may be argued that bentonite-water (usually rain water of low ionic strength) interaction alone does not erode or “dissolve” bentonite, but that physical forces are needed as well. It can also be argued that chemical erosion is limited by the occurrences of impurities or accessory minerals in the bentonite (e.g. gypsum). In many cases, a thin layer of soil at the top of bentonite is enough to “buffer” the dilute water, thus preventing chemical erosion. In repository conditions, there is a barrier of about 400 m of rock between the soil and the bentonite and backfill in the EBS, which would buffer any dilute rain or glacial water long before reaching repository depth.

8.4 Limited impact of cementitious materials

Cement and concrete are extensively used in the construction of repositories for low- and intermediate-level radioactive wastes (L/ILW) in, for example, underground silos such as in Finland and Sweden and much of the waste is conditioned with cement-based materials and is packed in concrete containers. Even in many spent nuclear fuel and HLW repositories, cement and concrete will be used in paving tunnels and drifts, shotcreting tunnel and drift walls, grouting of fractures etc. and this will be the case at Olkiluoto. However, only low-pH cementitious materials will be allowed at the repository level and no grouting will be allowed in the deposition holes. While it is foreseen (e.g. Karvonen 2011) that all non-essential cementitious material will be removed before closure, a significant volume of cementitious material will still remain in place (e.g. the largest volume of concrete in the deposition tunnels is in the low-pH concrete plugs).

Although a relatively old review, McKinley & Alexander (1992) noted that there were few good natural and anthropogenic analogues of cementitious systems and recent literature searches carried out by the author supports this even today. Some new material (on low pH cements) is presented in Section 8.4.2 and, otherwise, the main focus of new work has been in the area of CCS (carbon capture and sequestration), some of which is presented below. Otherwise, most information has been produced in the Maqarin (Jordan) Natural Analogue Project which was initiated in 1989 with Phase I, continuing with Phase II in 1991 and Phase III in 1993. Phase IV started in 1999 and the results have just been published (see below).

8.4.1 Cement analogues

Malinowski & Garfinkel (1991) described the use of a lime-based concrete in the floors of a Neolithic (ca. 7000 BC) construction in Galilee. The type of material used here indicates that Neolithic man had technology for the burning and calcining of limestone. Prior to this discovery, it was thought that the first inorganic cements were made from gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$). It is generally agreed that Egyptians used gypsum in the construction of the Great Pyramid of Cheops built between 2613 and 2494 BC (Lea 1970). Thomassin & Rassineux (1992) reviewed some of the literature on Gallo-Roman cement-based materials and noted that one of the most impressive examples is the 1700 year old Roman mortar used in Hadrian's Wall (Figure 8-23) which still contains substantial amounts of CSH (calcium silicate hydrate) compounds. These mortars were studied (e.g. Jull & Lees 1990) specifically with the behaviour of an ILW repository in mind. The origin of the CSH compounds in Hadrian's Wall is thought to be from calcining of siliceous limestones to produce lime or by the inclusion in the mortars of larnite from metamorphosed cherts found locally in limestones. The formation of the CSH compounds reduced the porosity and permeability of the cement which helped to ensure the wall's preservation for the last 2000 years in this notoriously wet part of northern England.

More recently, samples were collected from the Gallo-Roman amphitheatre in Béziers (Figure 8-24) as an analogy to the long-term behaviour of a surface repository (Miller et al. 2002). Although the age of the cements is well known, the site has suffered from

variable saturation and so it is difficult to draw firm conclusions on the implications of the high degree of carbonation observed.

The natural cements at Maqarin, northern Jordan (Figure 8-25) are part of a widespread terrain which stretches from Syria in the north, through Israel and Jordan to Saudi Arabia in the south. These cements were formed by the combustion of organic-rich limestones, a process which continues today (Kamei et al. 2010). The oldest reported cements in this area are some 2 Ma old (Alexander 1992) and, although usually heavily fractured into blocks (Figure 8-26), reaction is very much restricted to the outer edge of the blocks. This is possibly because the natural material is of low porosity and permeability (generally, the cements act as an aquiclude in Jordan and Syria) and the secondary reaction products naturally seal any flowing porosity (see discussion below). Reaction only occurs following fracturing and exposure of fresh surfaces (usually following earthquakes)



Figure 8-23. A close up of Hadrian's Wall (top) showing the mortar binding the stone blocks with (bottom) a section of core drilled through the wall (from Jull & Lees 1990).

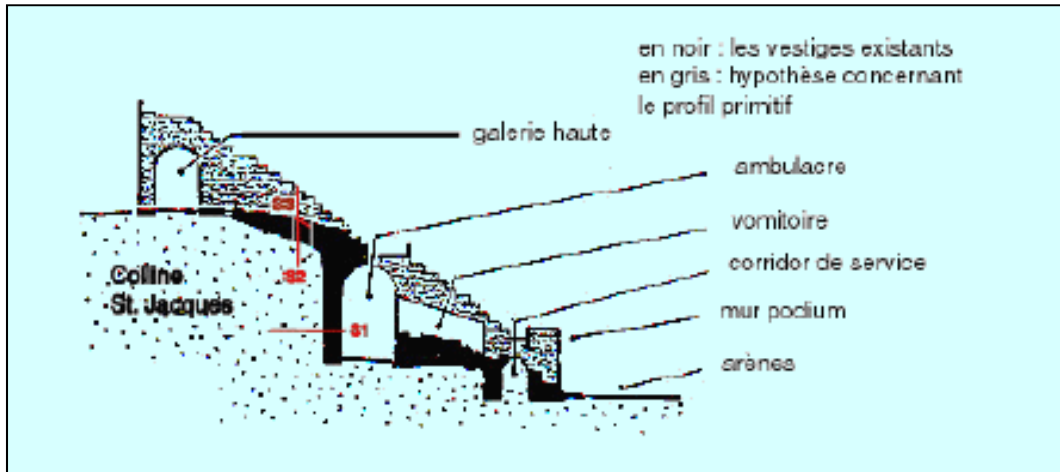


Figure 8-24. Section through the amphitheatre in Béziers, France. The black zone survives today (Miller et al. 2002).

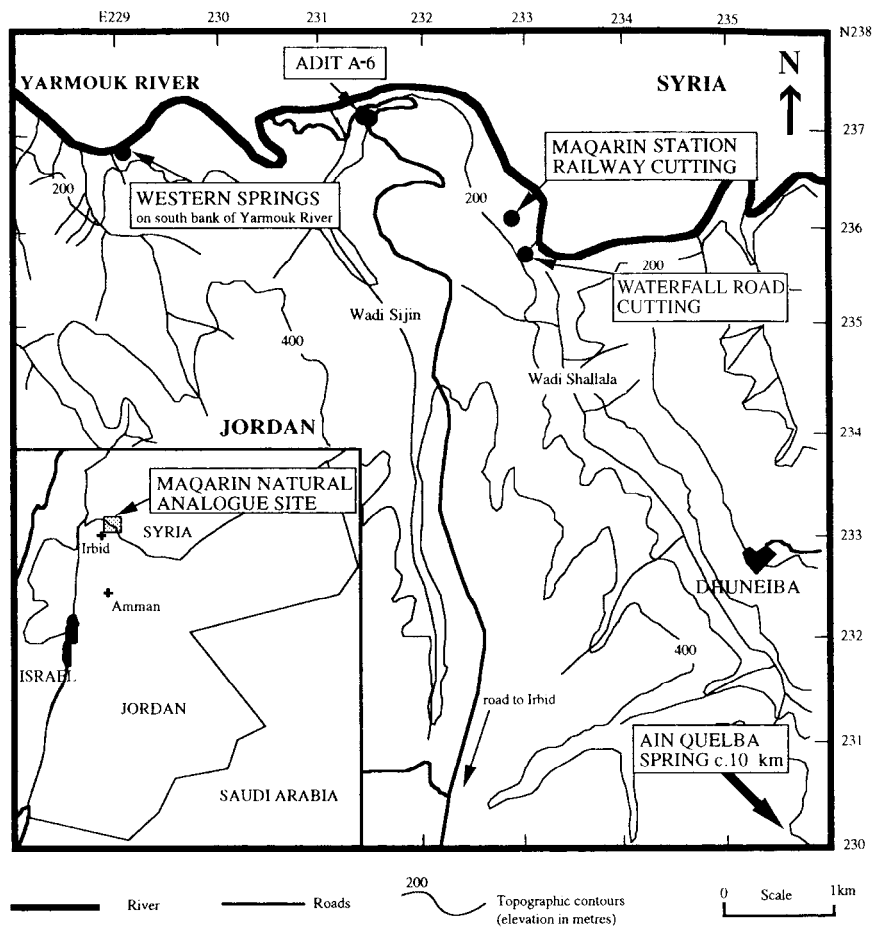


Figure 8-25. Map of Jordan and the Maqarin Site (Khoury et al. 1992).



Figure 8-26. Small aperture sealed fractures in the natural cements at the Maqarin site, Jordan (for scale, see hammer in bottom right corner). From Pitty & Alexander (2011).

Milodowski et al. (1989) also reported the presence of unreacted natural cements from the Scawt Hill and Carneal Plug sites in Northern Ireland (Figure 8-27). These phases were produced during the thermal metamorphism of the host limestone and are estimated to be some 58 Ma old.

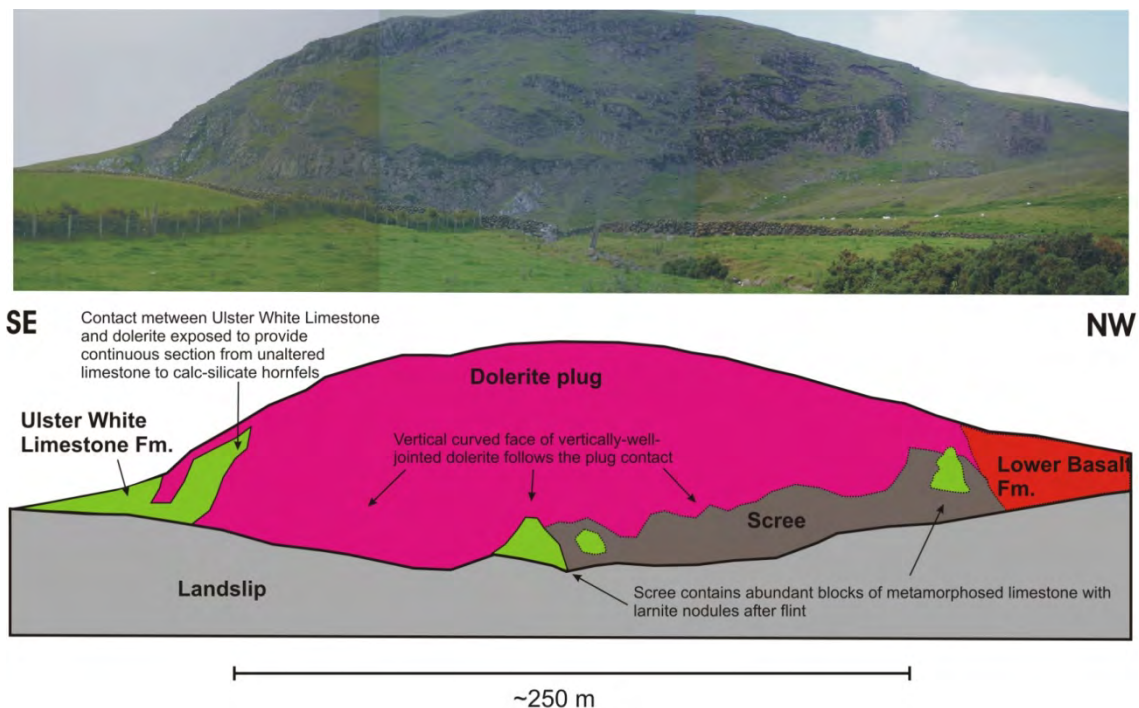


Figure 8-27. The Scawt Hill site in Northern Ireland (top). The natural cements are in the contact zone between the Ulster White Limestone and the dolerite plug (bottom). From Milodowski et al. (2009).

As with the cements in Jordan, these natural cements in Northern Ireland remained unchanged until accessed by groundwaters in the last 10–20 ka (Milodowski et al. 2011).

Conclusions

All studies of archaeological mortars so far have been focused on the similarities with modern OPC (Ordinary Portland Cement) as they were carried out before the current interest in low pH cements, making much of the information difficult to extrapolate to the low pH cements. Although the ages of many of the structures studied are impressive and are of direct relevance to near-surface repositories for very low and low level wastes, none of the material studied has been from an environment of relevance to a deep geological repository, so once again, extrapolation of data is difficult.

Despite the differences in manufacturing technique, the many examples of Roman concrete structures still in existence provides very strong qualitative evidence of the durability of cement-based materials. The writings of Marcus Vitruvius Pollio recorded in his treatise *De Architectura* (27 BC) make clear that the Romans exercised effective quality control during cement manufacture and this will have helped their preservation¹⁶.

There are many reports of long-lived natural cements with the oldest studied to date being some 58 Ma old, but where enough supporting information exists to assess why they have survived alteration, it appears that isolation from significant groundwater fluxes is of prime importance. With respect to Olkiluoto, although the overall groundwater flux will be low, it should not be forgotten that cementitious material will be predominantly used in areas where fluxes might be expected to be higher (e.g. as grouts in water conducting fractures etc.). As such, these cements might be expected to have a shorter life span, although this is currently under investigation elsewhere (see, for example, the discussions in Alexander & Neall 2007 and Sidborn et al. 2012).

8.4.2 Cement-bentonite interaction

Bentonite – especially the swelling clay component (smectite) that contributes to its essential barrier functions – is unstable under higher pH conditions (e.g. Savage et al. 2010), so there is potential for bentonite degradation in the case that cementitious porewaters (which have an initial pH of up to about 13: see, for example, Atkinson 1985) leach out of any concrete used in the Olkiluoto repository. Recently, therefore, there have been extensive efforts to better understand the interactions of hyperalkaline fluids with bentonite (e.g. Karnland et al. 2005, 2007), coupled with studies aimed at reducing the risk by development of low pH cement formulations (e.g. Mihara et al. 2008) which will have lower pH leachates. The greatest challenge is bringing the information produced by laboratory (conventional and underground rock laboratories, URLs) and modelling studies (e.g. Soler 2010) together to form a robust safety case.

¹⁶ Of course, examples of poor quality mortar from antiquity are rare as the usual processes of weathering act on the mortar leading to loosening of the masonry and collapse of the structure, possibly with the original stonework removed by local populations to new building and uses.

This is complicated by the inherently slow kinetics of such reactions and the commonly observed persistence of metastable phases for geological time periods (for a good overview of the issues involved, see Metcalfe & Walker 2004). Clearly, this is an area where analogues could play a valuable role – bridging the disparity in realism and timescales between laboratory studies and the systems represented in repository performance assessment (see also discussion in Alexander et al. 1998 and Miller et al. 2000).

As indicated in Figure 8-28, the cement leachate is simulated by natural hyperalkaline water, which, if the timescale of interaction can be determined, allows the models that are being developed to quantify the specific processes shown in the figure to be tested.

Data on the impact of hyperalkaline groundwaters on bentonite are restricted and the information is ambiguous in some cases. In the Jordan Natural Analogue Study, several sites across the country were examined to assess a range of mechanisms and processes of relevance to cementitious repository systems (see Kamei et al. 2010 for an overview). The natural cements which are the source of the alkaline leachates are produced by combustion of organic-rich limestones (Figure 8-29). At one site, Khushaym Matruk (Figure 8-30) in central Jordan, it is believed that the cement leachates diffused down into the underlying clay-rich sediments (see Figure 8-31) and reacted with them. Techer et al. (2006) state that the clays have clearly reacted with the alkaline solutions, dissolving smectites and leaving zeolites as reaction products. However, Pitty & Alexander (2011) modulate these conclusions, noting that “*The observed transformations in the samples from the Khushaym Matruk site were mainly secondary recrystallisation in joints and mineralogical evolution in the clay-rich matrix near the contact with the alkaline cement zone ACZ.*” They appear to post-date the combustion processes and, consequently, they might be attributable to the circulation of hyperalkaline solutions from leaching of the overlying ACZ (although other processes, such as weathering, cannot yet be ruled out). Zeolites have been found in the joint fillings and in foram tests in the matrix and the weight of evidence suggests that these phases have been produced by interaction with hyperalkaline fluids. However, it is not yet possible to rule out the matrix zeolites being a primary diagenetic feature of the sediments without further isotopic analysis.

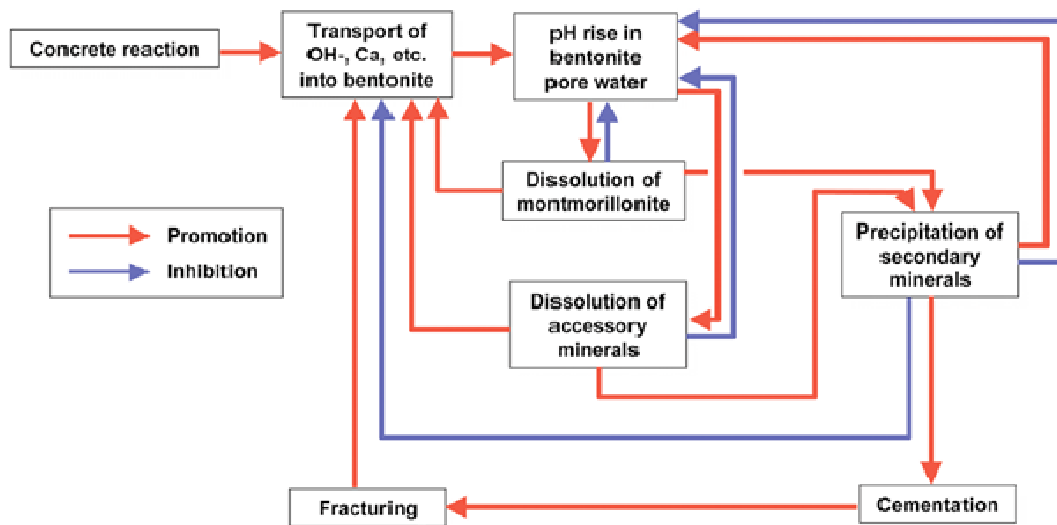


Figure 8-28. Some of the important processes involved in bentonite alteration (from Metcalfe & Walker 2004); at the natural analogue sites in Cyprus, natural hyperalkaline groundwaters simulate the concrete leachate.

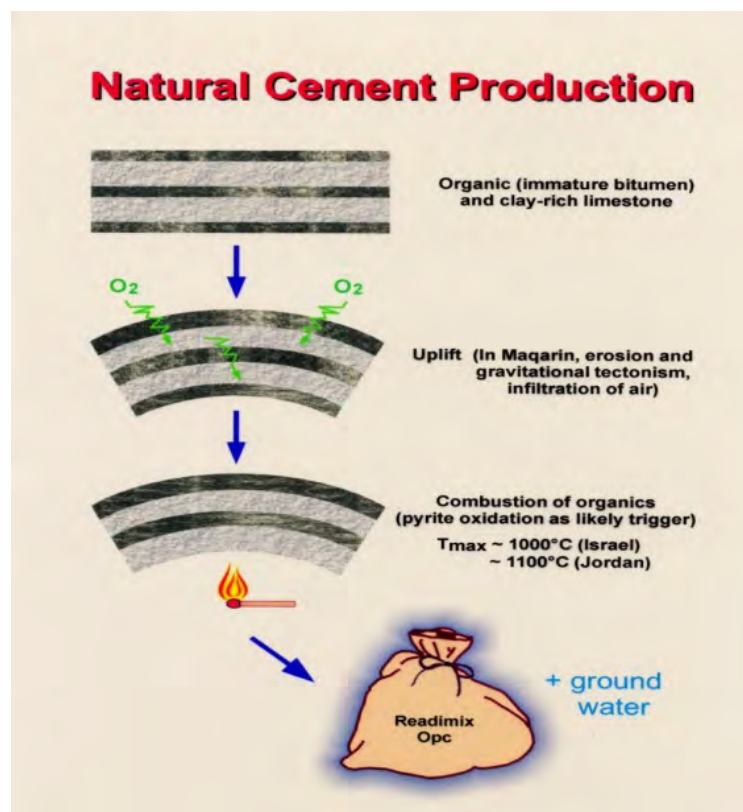


Figure 8-29. The natural cements in Jordan are produced by combustion of organic-rich limestones (from Alexander et al. 2005).



Figure 8-30. Khushaym Matruk site in Jordan. The sampling area is at the base of the peak, left background.

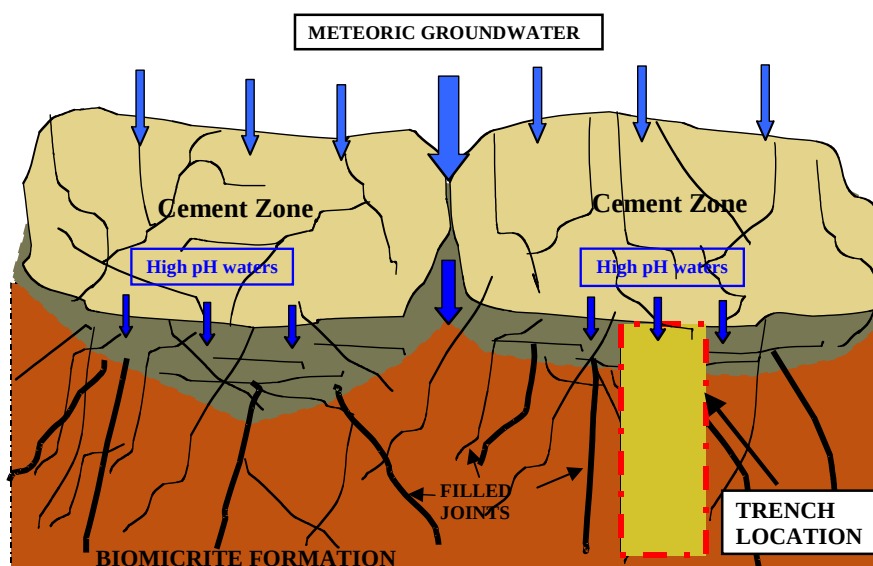


Figure 8-31. Sampling at Khushaym Matruk was by trenching. The trench begins at the cement zone and moves away downslope into the underlying clay-rich sediments. The thermal aureole from the cements extends for the length of the trench (Pitty & Alexander 2011).

As noted above, much effort has been invested recently in the development of low pH cements (also known as low alkali cements) in the belief (based on laboratory and modelling studies) that the lower pH of the cement leachates (usually pH 10–11) would be less deleterious to any bentonite in the vicinity. Unfortunately, the reaction kinetics involved are very slow and so the reaction is difficult to study in the laboratory (cf. Karnland et al. 2005). Consequently, two new natural analogue studies on bentonite interactions in low pH cement leachates are currently on-going in the Philippines and Cyprus (see Alexander et al. 2008a and Alexander & Milodowski 2011, respectively, for details).

These studies differ from Jordan insofar as the analogue cement leachates are not produced by natural cement, but rather by the serpentinisation of ophiolites. The exact reactions involved are highly variable (see Alexander & Milodowski 2011 for details), but the hyperalkaline groundwaters produced by reaction in the igneous layers of an ophiolite (Figure 8-32) are reasonable analogues of low pH cement leachates (see Table 8-4).

Bentonites are integral parts of the ophiolites (see Figure 8-32) and the hyperalkaline groundwaters move upwards through the ophiolite (Figure 8-33) until reaching the base of the overlying bentonites (Figure 8-34).

Table 8-4. *Hydrochemistry of hyperalkaline groundwaters: examples from around the world for comparison with the low pH cement leachates. All data in ppm.*

Location	pH	Na	K	Ca	Mg	Cl
Cyprus¹						
Cyprus 3a	11.5	385	15.1	1.0	0.3	420.0
Cyprus 3b	11.2	163.0	1.2	93.0	0.5	190.0
Worldwide²						
Greece	11.3	24.0	1.0	34.0	0.3	15.0
Bosnia	11.7	35.0	1.5	29.0	7.0	20.0
Oman	11.5	132	4.8	34.0	1.3	127.5
PNG	10.8	15.0	3.0	14.0	2.3	22.0
Western USA	11.5	19.0	1.0	40.0	0.3	63.0
Philippines³						
Manleluag1	11.1	28.0	0.5	18.6	0.2	17.4
Manleluag 2	10.4	20.6	0.4	18.1	---	15.8
Poon Bato	10.9	18.4	0.9	33.1	0.05	20.9
Narra 1	10.8	158	0.9	3.1	0.0	95.0
Narra 3	10.3	157	0.9	2.4	0.1	80.0
Cement leachate⁴						
ALL-MR f63	11.0	42	7.3	20	<0.5	52
OL-SR f63	10.0	4400	150	4300	0.56	13000

Notes:

- 1: Neal & Shand (2002)
- 2: Neal & Stanger (1983)
- 3: Alexander et al. (2008b)
- 4: Vuorinen et al. (2005)

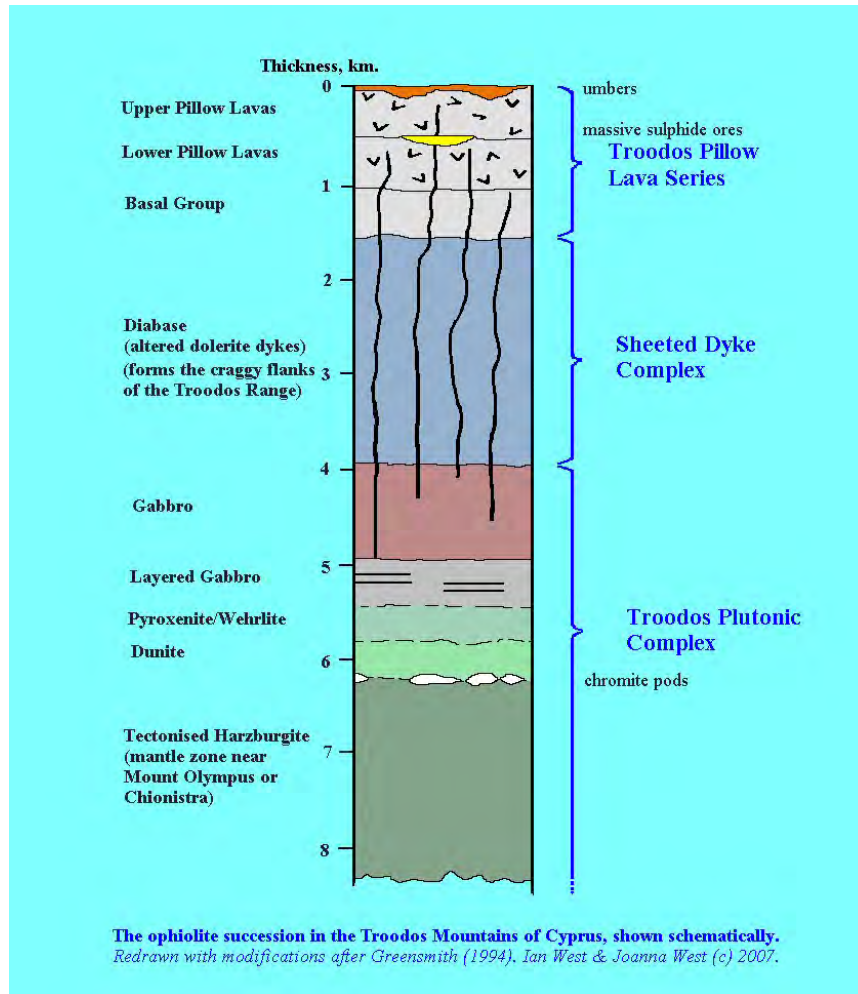


Figure 8-32. Schematic lithographic section of the Troodos ophiolite in Cyprus (from West 2007). The ‘umbers’ at the top of the sequence include bentonites.



Figure 8-33. Hyperalkaline groundwater flowing from a fracture in the gabbro of the Zambales ophiolite, Philippines. The groundwater here has a pH of 11.1 and releases a mixture of hydrogen (up to 3500 ppm) and methane (up to 5000 ppm) bubbles (misty area in centre of photo).

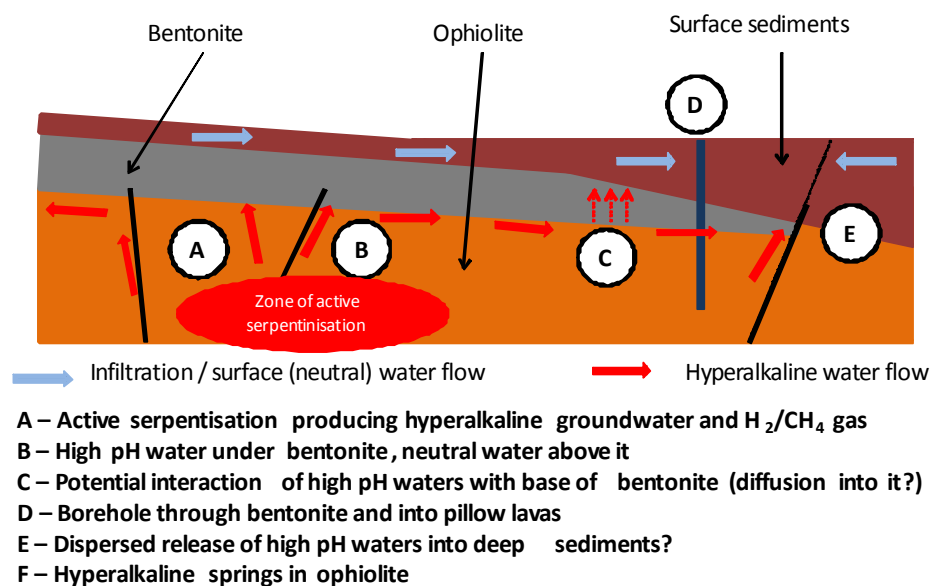


Figure 8-34. Conceptual model for alkaline groundwater/bentonite reaction at Cyprus (from Alexander & Milodowski 2011).

Although these projects are still ongoing, preliminary results (e.g. Fujii et al. 2010) from Philippines indicate that bentonite reactions would appear to be minimal and that the bentonite retains its swelling capacity and acts as a seal, effectively trapping the alkaline groundwaters below it (Figure 8-33). In Cyprus (Figure 8-35), both the base of the bentonite and fracture faces in sparse fractures in the bentonite show a very small degree of reaction, with the smectites producing palygorskite as the reaction product (see Alexander et al. 2011 for details). This is consistent with observations from other natural alkaline systems such as soils (e.g. Singer 1979) and limited laboratory data. These laboratory experiments indicate that palygorskite can develop relatively rapidly and in a closed system which is perhaps not unlike that present below the bentonite in Cyprus, suggesting a common reaction sequence which would be worth investigating further. Of importance is that less than a few percent of the bentonite has been reacted to palygorskite over a period of several hundred thousand years and that, otherwise, the bentonite appears to have retained its isolating properties regardless of the reaction. Recent work from the open (and advective) system at Searles Lake (Savage et al. 2010) reports different reaction products, such as Fe-illite, analcime and K-feldspar (although palygorskite is also abundant in the area), but this may be a reflection of the very low solid/liquid ratio of this system.



Figure 8-35. *Drilling through the bentonite to access the reaction zone at the base. Parsata, Cyprus.*

8.4.3 Cement-host rock interaction

The Maqarin site in northern Jordan appears to be unique in that the hyperalkaline groundwaters in the area are the product of on-going leaching of an assemblage of natural cement minerals produced as a result of high temperature-low pressure metamorphism of marls and limestones which is highly analogous to the process of producing ordinary Portland cement in a cement kiln. When this raw material is rehydrated by groundwaters, it effectively produces an analogue of concrete. Later groundwater interaction with this material produces hyperalkaline leachates (i.e. groundwater) directly analogous to the leachates produced when repository concrete/cement interacts with the host rock groundwater. Long-term studies at the site (see Pitty & Alexander 2011 for details) have shown that:

- reactions between hyperalkaline waters and the host rock mostly have positive reaction volumes and thus fractures are sealed by the precipitation of secondary phases;
- interaction between hyperalkaline waters and the host rock occur extensively and (small aperture) fracture sealing occurs within short timescales (years to hundreds of years);
- once a fracture is sealed by secondary phases, no further alteration can take place unless new pore-space is created by fracture reactivation (e.g. by earthquakes) –

while this is relevant at the Maqarin site, it is unlikely to be a feature of the Olkiluoto site (see discussion in Section 7.4);

- before the fractures fully seal, the altered rock matrix appears to be accessible to diffusion of aqueous species to a depth of several centimetres.

Clearly, any blocking of fractures in a repository host rock will have an impact on the site hydrogeology and evidence from laboratory tests (e.g. Bateman et al. 1995, 1998; Adler et al. 2001) and one URL experiment (the HPF experiment at the Grimsel Test Site; see Mäder 2011, for details) indicate that sealing of fractures ‘disrupts’ the pre-existing flow conditions. In the HPF (Hyperalkaline Plume in Fractured Rock) experiment, formation of secondary phases in the experimental shear zone induced a decrease in fracture transmissivity and a focussing of flow in the remaining open porosity. However, the data from HPF must be treated with caution as the pressure in the groundwater injection system was continuously increased to maintain constant flow conditions. In the case of a repository, the hydrogeological system will probably be largely dominated by the relative elevations of the recharge and discharge regions, and these elevations can be assumed to remain largely constant over the timescales of interest¹⁷. This situation is best represented by constant gradient conditions, which means that when permeability decreases (at constant gradient), flux will also decrease. In the case of the HPF supporting laboratory studies on a fractured rock core (see Mäder et al. 2004, 2006), a constant gradient was applied during infiltration of hyperalkaline leachates and the production of secondary phases led to a decrease in hydraulic conductivity of 20-fold (over a 9 month period).

As noted in Alexander (2010), the quantitative implications of cement leachate/host rock interaction on the long-term performance of repository in fractured crystalline rock have barely been examined. Two examples exist where preliminary assessments of the implications of the hyperalkaline plume on specific repository sites have been carried out. In the first, Alexander & Mazurek (1996) considered the likely impact on the fractured marls of the Wellenberg site in Switzerland. Focus was on application of the data collected in the Jordan natural analogue studies and the main conclusion was that it was necessary to conduct a detailed assessment of the likely long-term impact of gradual fracture sealing at the site due to the host rock reacting with the hyperalkaline fluids. In the second, Vieno et al. (2003) assessed the likely impact on the EBS of hyperalkaline leachates (predominantly from fracture grouts) in the proposed Olkiluoto repository. The results suggested that little impact would be expected but, although Vieno et al. (2003) made use of hydrogeological data on the site, no attempt was made to assess the impact of the plume on the host rock. More recently, Posiva implemented an RTD (research, development and technical design) project in 2006 with the aim of developing an acceptable grouting method before the ONKALO excavation extended beyond the low-gradient fracture zone known as R20 (now called HZ20). The project’s results led to a recommendation to the use of low pH grouting cement in ONKALO (Arenius et al. 2008).

¹⁷ Obviously, this will not be the case in an area of significant uplift or erosion or at coastal sites (see McKinley & Alexander 2009, for discussion)

Conclusions

Several studies (natural analogue, laboratory and URL) indicate that the reaction of cement leachates on a fractured crystalline host rock is likely to impact the site hydrogeology. However, only two preliminary studies on the likely effects have been reported to date.

8.4.4 Carbonation

Neall & Johnson (2006) noted that, although the carbonation mechanism can be viewed as mainly a favourable phenomenon for safety which could “...to a large degree mitigate the potential for high pH alteration of the buffer...” it has generally been omitted in safety assessments to date. It may be worth conducting scoping calculations (cf. Pfingsten 2001) to assess if carbonation by groundwater HCO_3^- could play a significant role in minimising hyperalkaline leachate production at Olkiluoto. Currently, no directly relevant natural analogue studies of carbonation exist, but there is potential to study the process at Maqarin (Alexander & Neall 2007).

Based on $\delta^{13}\text{C}$ data, a period of CO_2 uptake has been identified at the site, but no more than a preliminary assessment was originally made. In addition, new work in support of carbon capture and sequestration (CCS), such as the natural analogue work of Pearce (2006) or the on-going NASCENT project (<http://www.bgs.ac.uk/nascent/home.html>), hold the potential to elucidate the likely impact of carbonation on cementitious systems.

Conclusions

Although no direct natural analogue evidence currently exists for carbonation effects, existing sites such as Maqarin could be studied to gain an understanding of the likely (positive) impact of carbonation on the cementitious materials at Olkiluoto.

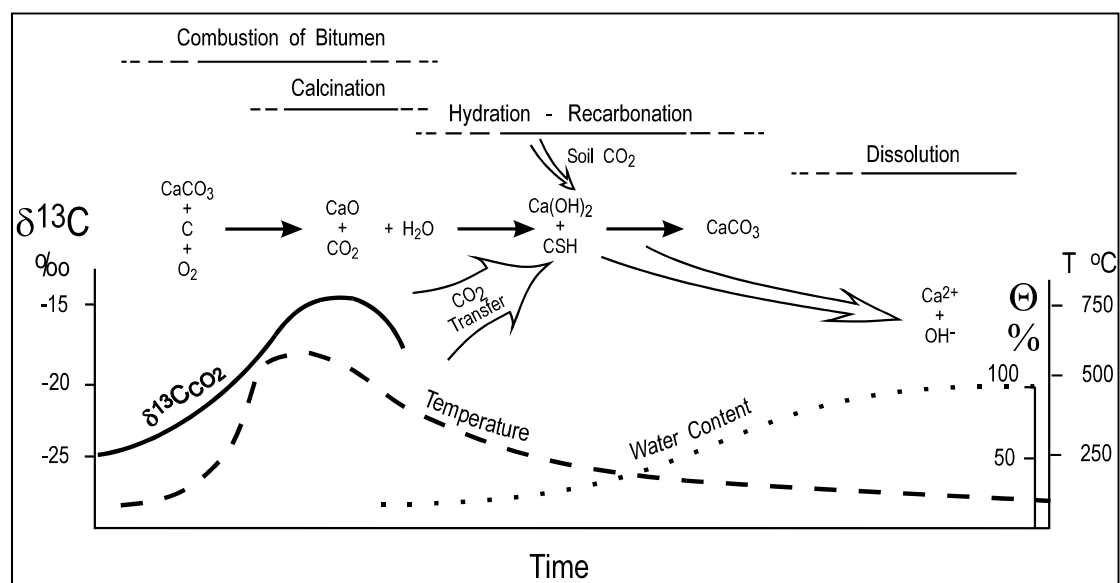


Figure 8-36. Conceptual model showing sequence of thermal metamorphic and retrograde alteration reactions with changes in temperature and humidity in the reaction zone at Maqarin (from Clark et al. 1994).

8.5 Silica sol analogues

Silica sol has been developed as a grouting material for fractures in situations, such as the deposition tunnels of a spent nuclear fuel repository, where it is desirable to avoid the use of conventional cementitious grouting materials (Axelsson 2004, Funehag 2005, Butrón et al. 2007). Silica sol grout consists of inorganic colloidal silica with an accelerator, usually NaCl, thus its use avoids introduction of organic additives that might be required with cement-based grouts.

Grouts based on colloidal silica were developed for use in engineering applications where the key problem is very small fracture apertures into which the coarser-grained cement-based grouts could not penetrate (e.g. Emmelin & Funehag 2005). Questions arise with respect to their use in a repository context, e.g:

- whether they are more benign than cement-based grouts as they lack the high pH source material;
- whether their use will introduce other potentially detrimental interactions with the EBS, such as affecting bentonite stability;
- their long-term durability;
- whether silica colloids are released from the silica sol and whether they could affect radionuclide transport.

To evaluate long-term durability, natural cherts (which can be produced from colloidal silica solutions; e.g. Folk 2002, Rogers & Longman 2002) in clay-rich sediments could be examined for indications of silica-clay interactions and sol durability. Appropriate information could perhaps also be sought from the extensive literature on silica diagenesis in clay-rich sediments. However, such natural analogue studies are still to be started and there is little relevant information currently available upon which to base any statements about the long-term behaviour of the silica sol.

9 EVIDENCE FOR LIMITED RATES OF RADIONUCLIDE MIGRATION IN THE REPOSITORY SYSTEM

To understand radionuclide retardation in general and in the geosphere in particular, it is necessary that all relevant processes and all structures are represented in an appropriate manner. This is the case whether transport in the rock formation is predominantly controlled by diffusion (e.g. in a salt diapir or in a consolidated claystone) or by advection in porous media (e.g. in a sandstone aquifer or fluvial gravel deposit) or in fractured media (e.g. in a limestone formation or in crystalline rock as in the Olkiluoto area). In addition to the direct processes of advection, dispersion and diffusion, a range of coupled processes, including thermal, chemical and electrical osmosis, thermal diffusion, hyperfiltration and electrophoresis, can transport porewater and radionuclides in solution in response to gradients in temperature, pressure, solute concentration and electrical potential. Although such coupling is generally negligible for most practical applications, some of these processes, including chemical osmosis and hyperfiltration, can be significant in argillaceous sediments, where the overlapping diffuse double layers of clay platelets result in the rock acting as a semi-permeable membrane. This has direct bearing on the behaviour of the bentonite buffer and the clay backfill in the drifts and tunnels.

The complete Onsager matrix of direct and coupled processes is given in Table 9-1.

A number of chemical retardation mechanisms have been identified in natural systems, as shown diagrammatically in Figure 9-1c and d:

- adsorption
- ion-exchange (and isotope exchange)
- precipitation (and co-precipitation)
- mineralisation.

Table 9-1. *The Onsager matrix of direct (diagonal) and coupled (off-diagonal) transport processes (from Horseman et al. 1996).*

	POTENTIAL GRADIENT			
FLUX	Temperature	Hydraulic	Chemical	Electrical
Heat	Thermal conduction (Fourier's Law)	Thermal filtration	Dufour effect	Peltier effect
Fluid	Thermal osmosis	Advection (Darcy's Law)	Chemical osmosis	Electrical osmosis
Solute	Thermal diffusion or Soret effect	Hyperfiltration	Diffusion (Fick's Law)	Electrophoresis
Current	Seebeck or Thompson effect	Rouss effect	Diffusion and membrane potential	Electrical conduction (Ohm's Law)

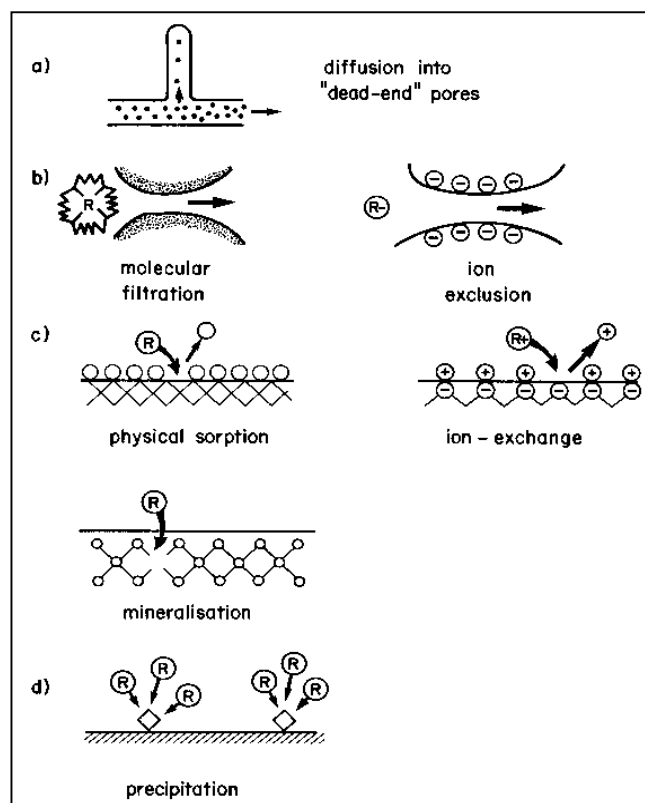


Figure 9-1. The retardation mechanisms that may affect radionuclides in the geosphere (after McKinley & Hadermann 1984).

Finally, radioactive decay and ingrowth are, of course, important processes to be taken into account in evaluating radionuclide retardation as the decay process can produce progeny nuclides whose geochemical characteristics, and hence retardation properties, differ markedly from those of the parent.

Between the study of natural analogues and the examination of radionuclide retardation in URLs, it is generally felt that all significant processes are understood and, here, a few examples from natural analogues which either support or illustrate this position are presented.

9.1 Migration in the buffer and backfill

A significant retardation process in compacted and plastic clays such as bentonites is that the lack of groundwater flow in fractures (also because of the lack of fractures) or matrix porosity reduces any transport to diffusive. As discussed below, this in itself will already bring a significant reduction to any radionuclide releases due to radioactive decay, especially if the diffusion times are significantly greater than the half-life of the radionuclide. For radionuclides susceptible of retardation, sorption, including ion exchange, on the individual minerals which make up the clay will clearly add to this process. In Table 9-2, a few examples of natural analogues of physico-chemical retardation in clays are noted. Although many examples are cited, few have been (or can

be) used in a quantitative manner, but they nevertheless serve to indicate the potential barrier function of clays.

More recently, the CLAYTRAC project (NEA 2009b) has been examining natural tracer profiles in clay formations to obtain a better understanding of long-term retardation processes. The rationale for CLAYTRAC was:

- Studies of tracer profiles refer to natural large-scale and long-term experiments;
- Usefulness for upscaling of laboratory and numerical experiments;
- Identification of relevant transport processes;
- Development of arguments on hydraulic (ir)relevance of faults;
- Consistency with independent lines of evidence (hydraulics, mineralogy).

The results of two case studies are briefly presented here: the Opalinus Clay (OPA) at Mont Terri in Switzerland and Couche silteuse at Marcoule, France.

9.1.1 Mont Terri

The Opalinus Clay study was carried out in the Mont Terri URL in St Ursanne, Switzerland (see Figure 9-2).

The main features of the site are:

- Jura Mountains are a thin-skinned fold belt, 10 to 3 Ma old;
- Exhumation and activation of aquifers over- and underlying the Opalinus Clay;
- Porewater Cl contents up to 14 g/L with marine Cl/Br ratio, so the porewater is assumed to have a connate component.
- Studies of natural tracers across the Mont Terri have shown that they display smooth, regular profiles with depth, with some profiles being more symmetric than the others (see Figure 9-3). When modelling the profiles, it was assumed that all transport was diffusive and that the Opalinus Clay porewaters began with a marine salinity (i.e. 19.3 g/L). In addition, following the erosion pattern, it was assumed that the upper aquifer was activated first, followed by the lower.

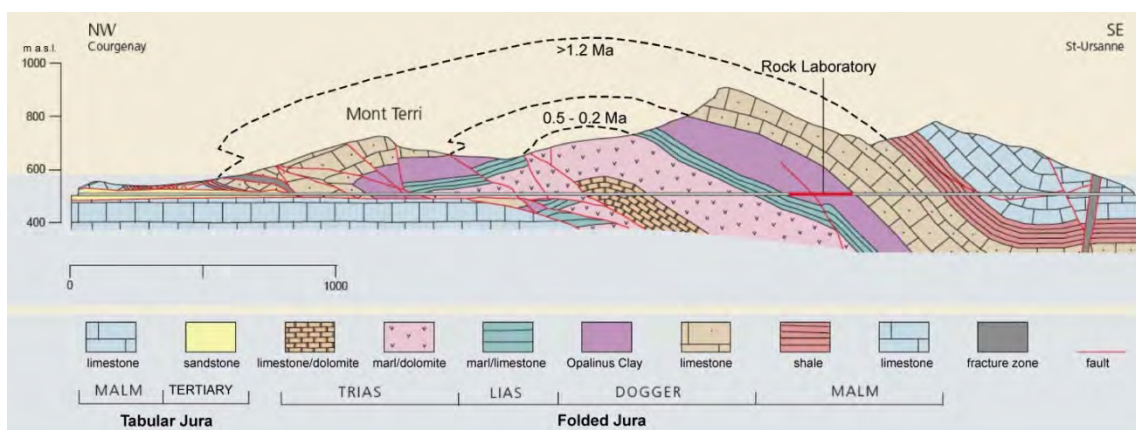


Figure 9-2. Cross-section of Mont Terri with the URL shown in red (from http://www.mont-terri.ch/pdf/geological_profile.pdf).

Table 9-2. Examples of natural analogues of physico-chemical retardation processes.

Site (and main references)	Clay type ¹	Hydraulic conductivity (m/s)	Temperature domain (°C)	Retardation	Comments
Clay halo around U ore, Cigar Lake, Saskatchewan, C. (Cramer 1994; Cramer & Smellie 1994)	illite/kaolinite enriched (30–80 %) sandstone	10^{-9} to 10^{-6} (in unfractured clay, 10^{-9} to 10^{-8})	150–200, has increased illite crystallinity	Isotope data indicate effective isolation of 10^4 – 10^5 a. Colloids in ore 'sealed in' by clay halo.	Clay-rich halo surrounds U ore. Possible that clay-poor sandstone (hydraulic conductivity of 10^{-5} m/s), around the halo acts like a hydraulic cage, but this was not examined.
Natural bentonite, Avonlea, Saskatchewan, C. (Oscarson et al. 1990)	bentonitic clay-shale	10^{-12} to 10^{-11}	diagenetic		Despite very high soluble salt content (34 g/L), clay maintains swelling pressure (>600 kPa) when wet.
Koongarra, Australia (von Maravic & Smellie 1994)	illite and montmorillonite in weathered zone			Preferential retention of U^{VI} by illite and montmorillonite.	Irrelevant to deep disposal programme.
Kronan cannon, Baltic Sea, S. (Hallberg et al. 1987)	Unconsolidated marine clay (montmorillonite)	no data	early diagenetic (<15)	Cu leached from cannon migrated maximum 0.06 m in 300 a.	Assumed to be diffusive but mechanical effects (bioturbation, wave action) cannot be discounted. No real quantitative assessment of migration carried out (Cu corrosion main area of interest, see comments in Section 8.2). Data were used by Apted & Engle (1991) for code testing.
Fracture smectite at Hyrkkölä, Finland (Marcos et al. 2000)	Smectite in fractured crystalline rock	Water conductive fracture; no data on hydraulic conductivity		Retention of Th and U^{VI} and U^{IV} .	Elevated U content (57 ppm) was observed in fracture smectite sampled from the surface of water-carrying fracture in granite pegmatite at a depth of 70 m.
Fossilised trees, Dunarobba, Italy (Benvegnú et al. 1988; Ambrosetti et al. 1992; Lombardi & Valentini 1996)	fluviolacustrine clay	$2 \cdot 10^{-13}$ to $2 \cdot 10^{-10}$	unknown but probably less than 25	Dead tree trunks enveloped in clay can still be sawn and burned after about 2 million years of burial. No data on clays but tree isotopic signatures similar to modern trees.	Probable that sand layers (hydraulic conductivity of 10^{-4} m/s), above clay act as a hydraulic cage, keeping oxygenated waters away from the clay and trees.

Table 9-2 continued. *Examples of natural analogues of physico-chemical retardation processes.*

Site (and main references)	Clay type ¹	Hydraulic conductivity (m/s)	Temperature domain (°C)	Retardation	Comments
Fossilised trees, Florennes, B. (De Putter & Charlet 1994, De Putter 1996)	Late Eocene marine clays and Neogene organic rich fluviolacustrine sediments	no data	no data	Branches, roots, ash deposits etc. preserved in clays.	Probable that surrounding karstic system acted as hydraulic cage.
Matrix diffusion, Opalinus clay, CH. (Mazurek et al. 1996)	Opalinus clay	at the site, unknown	surface weathering (<25)	Natural decay series disequilibria indicate matrix diffusion to at least 0.08 m from the fracture surface.	Fracture system non-typical for OPA at repository depth (as near-surface fracturing heavily influenced by glacial offloading and weathering).
Matrix diffusion, Palfris Formation, Wellenberg, Switzerland (author's unpublished data, quoted in Miller et al. 2000)	limestones, biomicrites and clay biomicrites			Natural decay series disequilibria indicates matrix diffusion to at least 0.06 to 0.07 m from the fracture surface.	Of particular interest is that fracture faces with calcite coatings still displayed evidence of matrix diffusion in the rock.
Matrix diffusion, Tono U prospect, Japan (Yoshida 1994)	fluviolacustrine, lignite-bearing sediments	10^{-12} to 10^{-15}	10	Significant grain boundary accumulation of U and associated diffusion into the grains.	
Reduction spots, northern Switzerland. (Hofmann 1990, 1992, 1999)	Permian redbeds, Buntsandstein-Lower Muschelkalk, USM all studied.	No data	no data	Complex element enrichments around redox fronts.	Existence of reduction spots at depth in the rock matrix indicates existence of connected porosity at distance from fractures. Long-term process with some spots estimated to take 10^6 a to form.
Near-surface PAH contamination, Chattanooga, USA (Vulava et al. 2007)	Clay-rich superficial floodplain	$2.8 \cdot 10^{-7}$ to $1.3 \cdot 10^{-3}$	Early diagenetic	Despite 4–5 m of thick, clay-rich (75 %) sediments, the PAHs have penetrated to the base of the sequence.	Appears to be diffusive transport with some advection along fractures and other discontinuities.

¹ Terminology of the authors is used here without comment.

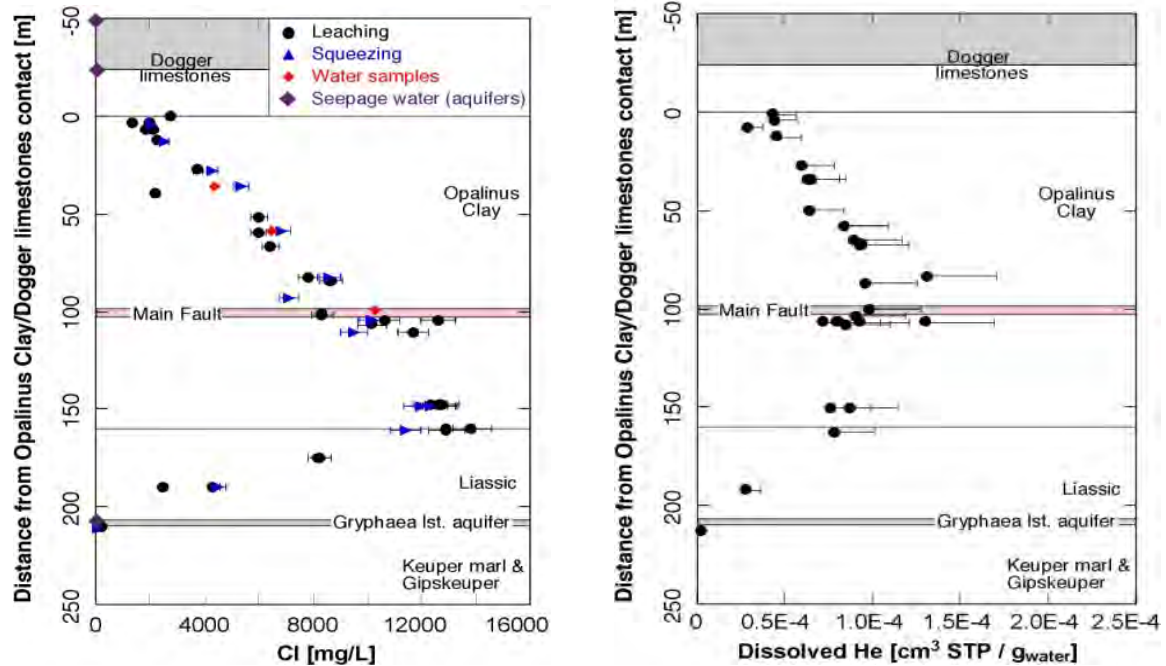


Figure 9-3. Cl (left) and He (right) profiles across Mont Terri (from NEA 2009b).

The results are shown in Figure 9-4 and show an excellent model fit for activation of the upper aquifer at 6.5 Ma and lower aquifer at 0.5 Ma (times which are within the geologically plausible range). The laboratory-derived diffusion coefficients also apply on the formation scale (providing an excellent example of upscaling, cf. Mazurek et al. 2007). The same model scenario explains the asymmetry of the Cl profile and the symmetry of the He profile (due to different values of the diffusion coefficient).

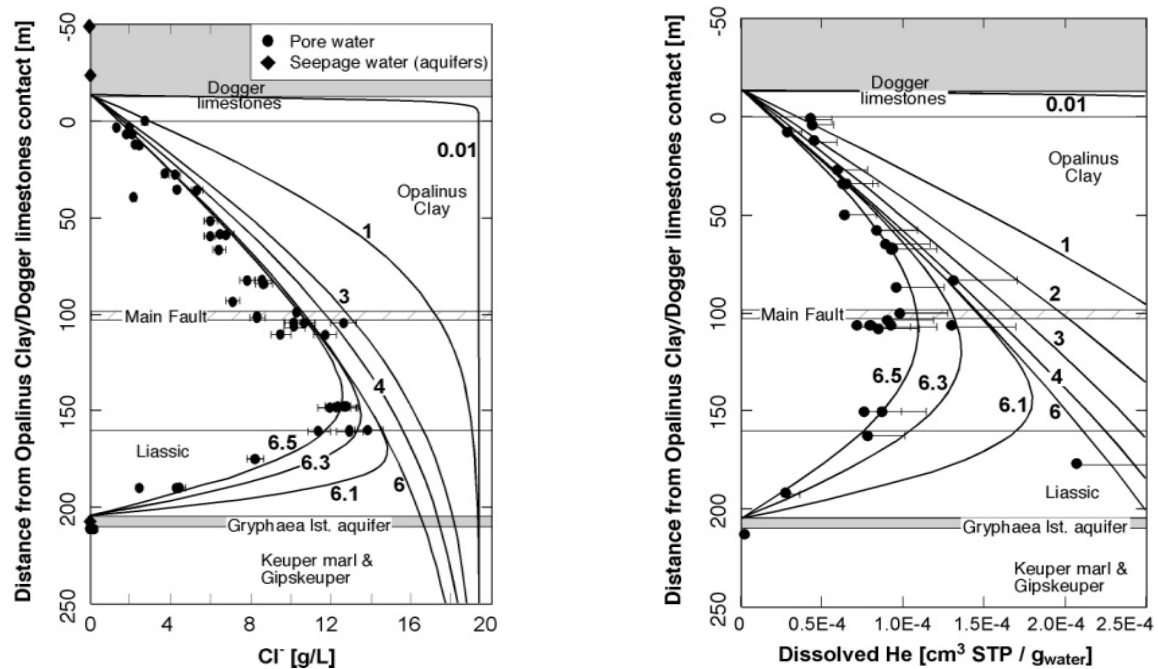


Figure 9-4. Model fits for Cl (left) and He (right) profiles across Mont Terri (from NEA 2009b).

9.1.2 Couche Silteuse

The argillaceous sediments at the Couche Silteuse at Marcoule, France (Figure 9-5) are marine silty shale 100 Ma old. The geological conditions are slightly more complicated than at Mont Terri with a significant erosion event happening 5.85–5.3 Ma ago producing deep canyons which refilled under marine conditions ~3 Ma before final emergence.

Once again, the natural tracer profiles in the porewaters (Figure 9-6) were modelled, but note that the Cl concentrations, although generally exhibiting smooth diffusive transport profiles, contrast significantly between boreholes. The highest Cl concentrations are at depth in borehole MAR 203 and are near to the full marine value. The question asked was: can the same model and the same palaeohydrogeological scenario explain all three profiles?

The authors claim that the results indicate that diffusion times of 3 Ma are consistent with data in boreholes MAR 203 and 501, tying in nicely with the estimated time of final emergence. However, this thesis needs to be better tested with more data from MAR 501. The diffusion time of 1.5 Ma obtained for borehole MAR 402 indicates delayed activation of the aquifers. However, this is plausible due to the greater depth of the formation here (Figure 9-7) and an increased distance from the infilled canyons (Figure 9-5). So, once again, diffusional transport alone can explain the natural tracer profiles in the clays at Marcoule.

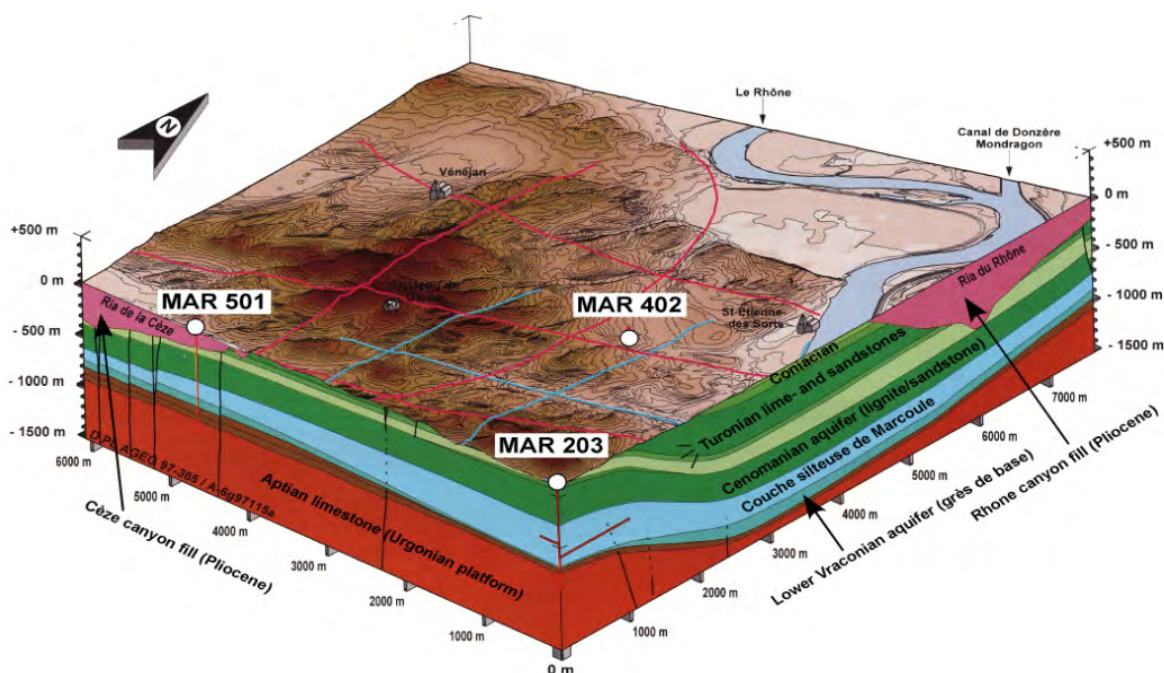


Figure 9-5. Block diagram of the Couche Silteuse with the three boreholes marked (MAR). The canyon fill is marked in pink in the figure (at the NE & SW corners of the block). After Mouroux & Brulhet (1999).

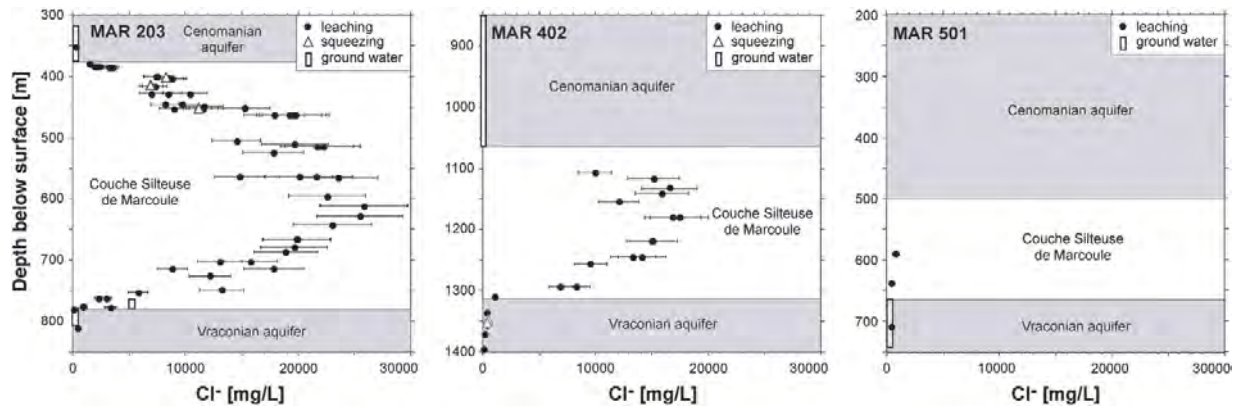


Figure 9-6. Distribution of Cl^- contents in pore and ground waters of boreholes MAR203, MAR402 and MAR501 penetrating the Couche Silteuse de Marcoule. Grey areas indicate aquifers. From Mazurek et al. (2011).

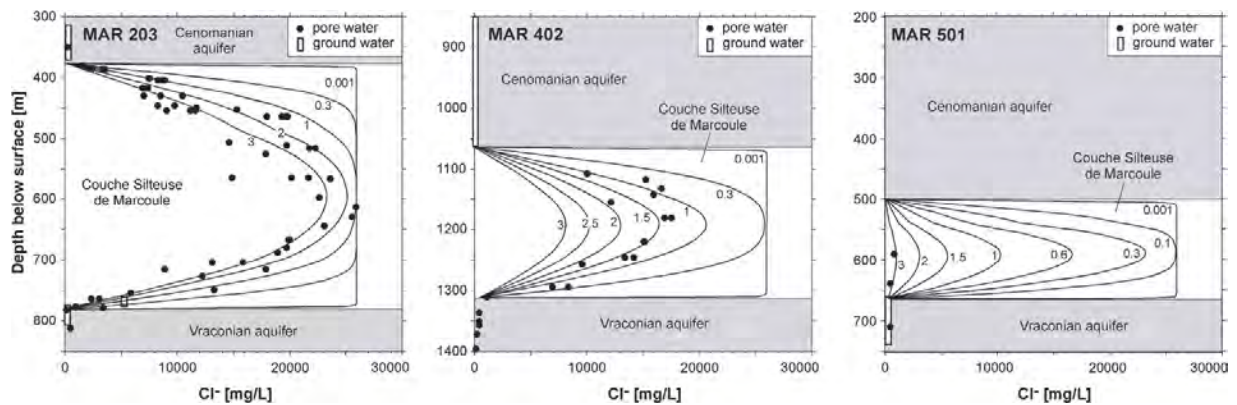


Figure 9-7. Base-case model for the out-diffusion of Cl^- at Marcoule considering an initial concentration of 25.9 g/L (the maximum observed Cl^- concentration in the Couche Silteuse – see Figure 9-6). Numbers adjacent to model curves indicate evolution times in Ma since activation of the aquifers. From Mazurek et al. (2011).

Conclusions

The two examples presented here show that:

- Several case studies (of which only two were described here and others presented in Table 9-2) indicate that transport in argillaceous rocks which are analogous to the bentonite buffer in the EBS is diffusion-dominated;
- Laboratory-derived parameters, such as D_e (effective diffusion), are applicable at the formation scale and this is presumed to be the case for bentonite;
- In the case of formation-scale studies like these, consideration of several tracers in parallel adds confidence in the interpretation;
- Similarly, quantification of tracer profiles requires a good understanding of the site palaeohydrogeology, something which is typically the case for repository host formations, but not always for analogue sites.

9.2 Migration in the host rock

The basic retardation processes shown in Figure 9-1 are also of direct relevance to fractured crystalline rocks. The main difference is that the principal method of solute transport in fractured rocks is advection through hydraulically active channels within fractures or fracture networks. Today, after decades of examination of likely processes in the laboratory, URLs and in natural analogue studies (see discussion in Alexander et al. 2003, for example), there is reasonable confidence that all relevant processes have been identified (see Figure 9-8), if not fully quantified (see comments in Section 9.3).

9.2.1 Retardation in fractures

Smellie (2011) noted that most natural analogue studies on radionuclide retardation in fractured rocks focus on matrix diffusion and this is addressed in Section 9.2.2. For example, in the Palmottu study, Smellie (2011) noted that *“All samples were selected away from the vein-type orebodies based on the assumption that radionuclides have been transported along the fractures from the mineralised localities by the flowing groundwater, from which the subsequent diffusion into the rock matrix has taken place.”* This is generally typical of most studies of radionuclide retardation in fractured systems, but at least one natural analogue study has tried to address retardation in the fracture system itself.

In the Klipperås natural analogue study (Landström & Tullborg 1990), a complex suite of fracture minerals was recorded, including chlorite, calcite, siderite, quartz, epidote, muscovite, illite, hematite, pyrite, goethite, kaolinite, mixed-layer clays, gibbsite, plagioclase, and potassium feldspar. In addition to the detailed mineralogy, analyses were performed on samples of fracture-filling minerals, the host rock and the associated groundwater. These data indicated that redistribution of certain elements occurred in response to alteration (clay formation) of the host rock by groundwater.

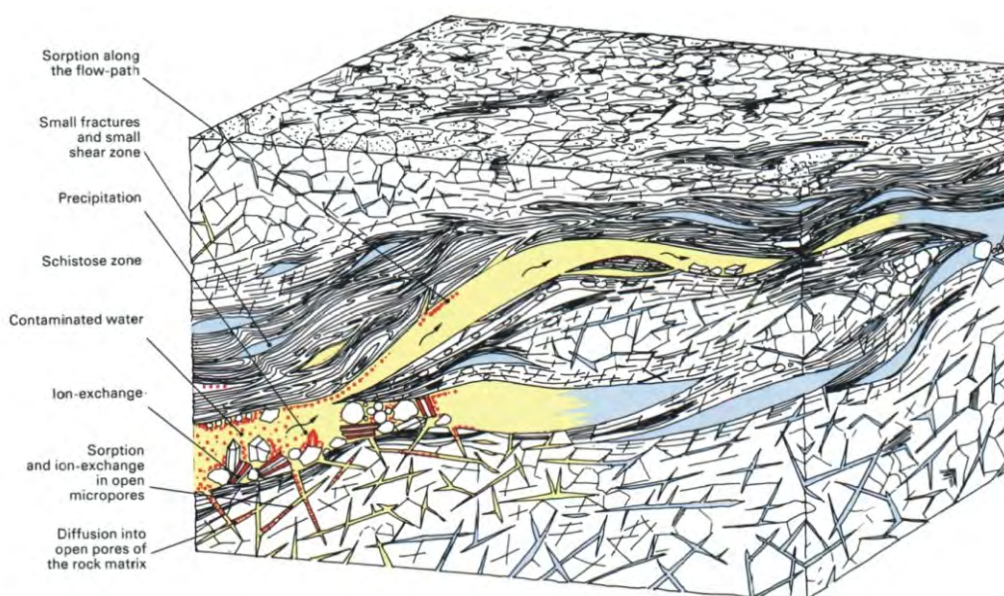


Figure 9-8. Some of the more significant radionuclide retardation processes in fractured crystalline rocks (image courtesy Nagra).

Uranium, for example, appears to have been very mobile and is associated with most fracture materials, preferentially with iron-rich phases. Similarly, thorium was found to have been mobile and strongly associated with iron-rich phases.

In general, this study confirmed the association of remobilised elements with iron-rich phases (principally secondary iron oxyhydroxides). However, the thorium data from this study show the problems associated with trying to separate the effects of recent, low-temperature processes from earlier high-temperature events. Thorium is generally immobile under low temperature conditions, but may migrate in association with colloids (Section 9.3). The thorium mobility described at Klipperås by Landström & Tullborg (1990) is, therefore, most likely to be an artefact of colloid transport or a previous high-temperature event, rather than recent low-temperature transport.

More recently, the study of retardation in fracture systems has been addressed in URLs, rather than natural analogue studies, but the reasoning behind this is probably questionable. For example, in a recently reported study of work in the Grimsel URL (also known as the Grimsel Test Site, GTS) in Switzerland, Alexander et al. (2010) noted that both traditional natural analogue studies and in-situ radionuclide migration tests tend to leave the flow system under-defined. Hautajärvi et al. (1997) stated that *“The general rationale of performing tracer tests has been, and will probably be also in the future, to characterise properties of the medium in question regarding flow and transport of solutes to various degrees of detail. Transport of solutes is, however, more dependent on certain details of the water flow-paths than the pressure field and average flow rates. It is necessary, therefore, to study also these details in addition to the hydraulic testing.”*

This is effectively what the GTS Excavation Project (EP) experiment was set up to address at the end of the extensive Migration Experiment (MI) programme. As McKinley et al. (2001) noted when reviewing the status at the end of MI: *“...all three (radionuclide transport) models are insensitive to the finer details of the in situ flow system. None of the three models considered the effects of channelling, multiple flow-paths or unlimited matrix diffusion...”*

and further

“...that all three codes examined appear to be insensitive to features of the flow system, which could play a role in radionuclide retardation in a repository host rock, requires further consideration. These results also indicate the limitations of the approach of process identification by examination only of the form of breakthrough curves...”

This clear lack of sensitivity of the transport codes was, arguably, partly due to a lack of appropriate data on flow systems for the models to address – and this became the focus of GTS EP. The thinking at the time can be well summarised in Figure 9-9: in MI, the input to the experimental shear zone (on the left – radionuclide activity, groundwater flow rates, tracer input function) were well known as were the outputs (on the right – radionuclide breakthrough curves). But the processes and mechanisms on-going inside the shear zone (the “black box” in the middle) were basically unknown and so were represented in the transport codes in a relatively simple and stylised manner. Thus the

focus of GTS EP became “opening the black box” and, by doing so, the aim was to elucidate as many of the then unknown processes and mechanisms as was possible.

Safety assessment relevant radionuclides were injected directly into a controlled flow system in a much more highly transmissive shear zone in the GTS tunnels (see Figure 9-10) than would be expected in a repository via a borehole injection system and, following a period of transport along the shear zone, the flow system was closed down and sections of the rock recovered to ascertain radionuclide transport distances and sites of retardation (in 3D).

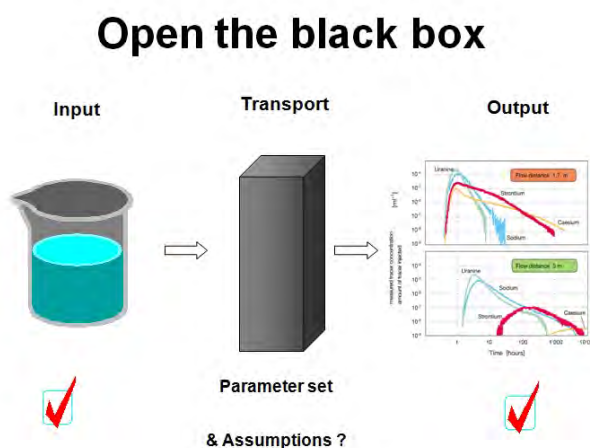


Figure 9-9. In traditional URL in-situ radionuclide tracer tests, the geosphere is a modeller’s “black box” from which a range of parameters may be deduced indirectly for input to transport codes (Alexander et al. 2010).

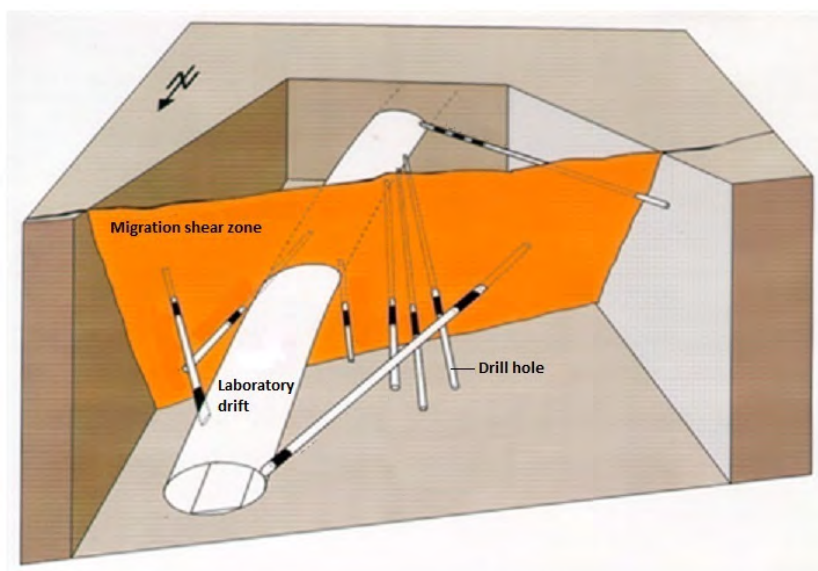


Figure 9-10. Schematic layout of the experimental shear zone in the GTS (from Frick et al. 1992).

The shear zone was highly friable and so, in order to define the sites of retardation of the radionuclides, it was necessary to inject a specially developed UV fluorescent resin into the rock to immobilise the entire system (see Figure 9-11). The target zone could then be overcored and the cores sliced and sub-sampled for later radiochemical analysis in the laboratory.

This showed clear retardation of radionuclides on the fracture faces and in fracture fault gouge (Figure 9-12). In addition to radiochemical analyses, the actual flow space geometry was analysed, using the presence of the fluorescent resin as an indication of open channels and pore spaces. After the flow-field geometry had been investigated in detail and the retardation behaviour of the different radionuclides studied, it was necessary to bring both data sets together to develop a conceptual model of groundwater and radionuclide transport in the geosphere.

The structural model of the experimental shear zone presented in Figure 9-13 is based on:

- The geological information gained from 8 successive core slices which had been digitised under UV light;
- Augmented with observations from the preceding structural analysis of the entire experimental shear zone; and
- Data on the distribution of radionuclides throughout the flow-field.

The bottom part of the figure therefore shows the open, water-filled volume within the shear zone and the upper part the distribution of the radionuclides along the length of the flow path. This not only allowed comparison of relative retardation of the radionuclides (they were found to approximately follow the retardation coefficient values assigned to them based on Nagra's then geosphere retardation database), but also allowed mineral-specific retardation to be defined along with the effect of the flow path structure.

Overall, the EP experiment allowed detailed tests of the conceptual models of radionuclide retardation in fractured crystalline rock and this greatly increased the confidence in these models and in the belief that all relevant retardation mechanisms have been identified. Furthermore the conclusions are in agreement with those of tests carried out elsewhere, such as the TRUE Block Scale international project carried out at Äspö (Winberg et al. 2003).

Conclusions

Appropriate natural analogue studies of radionuclide retardation in fractured rock are rare as most systems are under-defined. Nevertheless, both those relevant studies available in the literature and the few URL studies that have been undertaken indicate that the conceptual models of radionuclide retardation in the geosphere include all the most significant mechanisms and processes.

Immobilisation and sampling of a shear zone (schematic)

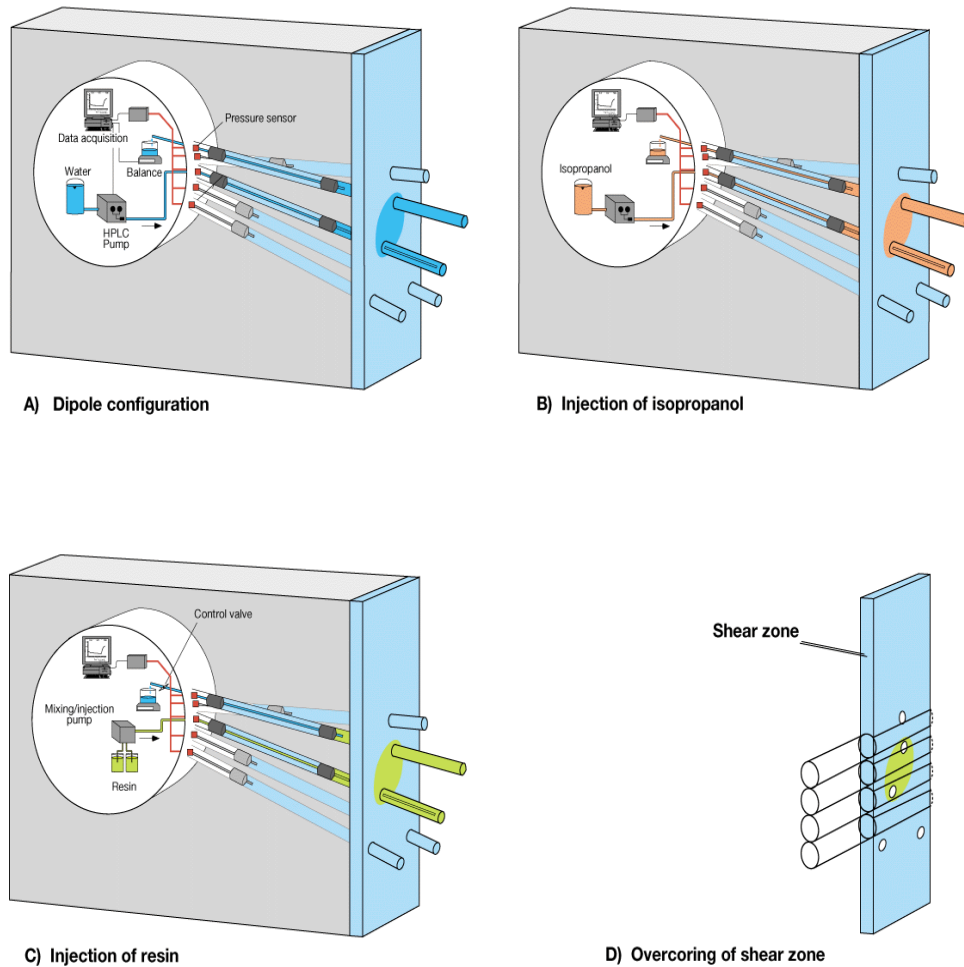


Figure 9-11. Excavation of the experimental shear zone (see Figure 9-10) during the EP experiment. A: injection of the radionuclides into a dipole flow-field. B: the water in the dipole is replaced by isopropanol to ensure adequate wetting of the rock by the resin which follows. C: specially developed resin is injected into the experimental shear zone and allowed to polymerise and harden. D: the immobilised (radioactive) part of the shear zone is recovered for analysis by overlapping triple barrel drill cores, drilled parallel to the shear zone (from Alexander et al. 1996).

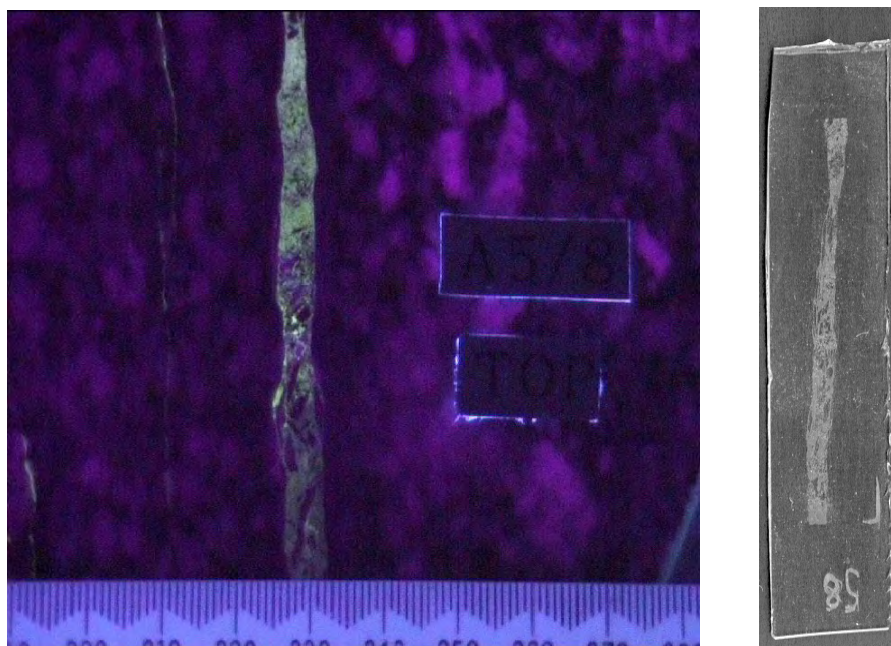


Figure 9-12. Clear examples of radionuclide diffusion into and sorption onto fracture infill and the fracture surface. The sample has been impregnated in situ with fluorescent resin and then over-cored and sawn into slices for examination. a) On the left: fault filled with fault gouge (yellowish colour) and fragments of wall rock (black) floating in the fault gouge. UV light, partial scale below in mm divisions. b) On the right: alpha-autoradiograph of part of the sample on the left showing uptake of radionuclides in the fault gouge (white material) and on the fracture surface (or wallrock) and on the surface of the rock fragments (grey areas). Note that neither image shows any indications of significant matrix diffusion as the experiment only ran for several months (Alexander et al. 2003).

However, there is a slight twist to the tale as Alexander et al. (2010) noted:

“During the original planning (see Alexander & Kawamura 1992), it was foreseen that safety relevant radionuclides would be injected into the experimental shear zone and, following natural retardation processes in situ, the flow-field would be recovered and taken to the laboratory. Here, the sites of radionuclide retardation would be defined and any mineral-specific or flow system-specific mechanisms elucidated. The data would then be used to improve the representation of radionuclide retardation in the geosphere in the then current generation of transport codes.

“Unfortunately, nature refused to play along with these plans and it rapidly became apparent that the natural solubility limits of the suite of radionuclides chosen for study would severely restrict the project aims. In other words, it simply was not possible to inject enough radionuclide mass (or activity) into the flow-field so that any surface or radio-analytical methods available at the time would be able to detect the sites of in situ radionuclide retardation. ...even assuming a best case scenario where the majority of the radionuclides would be concentrated in a very small volume (of several cm^3), it could be shown that only a couple of the radionuclides would be detected by then current radio-analytical methods.

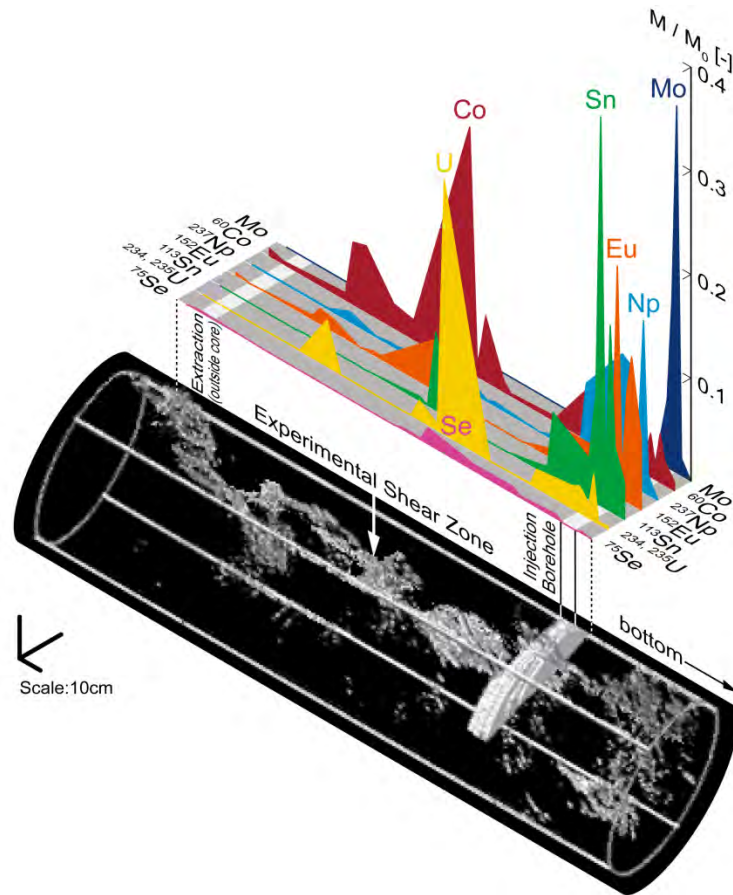


Figure 9-13. Conceptualisation of the water-filled porosity in the shear zone (bottom) along with the relative retardation of the injected radionuclides (top). Note that the injection borehole is on the right, so the radionuclides travelled right to left along the shear zone (Alexander et al. 2010).

“However, it was realised that a major part of the original aims could still be fulfilled, namely that flow-field-specific retardation could be studied by injecting radionuclides at concentrations above the natural solubility limit. By keeping the radionuclides in solution in acid, the geochemistry of the system would obviously be of little relevance to the majority of repository environments, but the radionuclide retardation sites would provide clear indication of the active flow-paths in the overall flow-field.”

9.2.2 Matrix diffusion

The term matrix diffusion is applied to the process by which a solute, flowing in distinct fractures, penetrates the surrounding rock. Diffusion into this rock occurs in a connected system of pores or microfractures, and diffusion through the solid phase is insignificant by comparison. The importance of matrix diffusion in the context of a radioactive waste repository is that it greatly enlarges both the area of rock surface (for sorbing radionuclides) and pore volume (for non-sorbing radionuclides) in contact with advecting radionuclides from just the fracture surface (and fracture mineral porosity) to a portion of the bulk rock (Neretnieks 1980, Grisak & Pickens 1980 and Ota et al. 2003).

The matrix diffusion theory proposes that dissolved radionuclides will diffuse from a water-filled fracture, through any fracture coating layer, into the rock matrix and, if they are reactive, sorb onto the inner surface of the pores or else remain dissolved within the immobile pore-water. This process can be envisaged as an extreme case of a dual porosity medium, in which advective flow occurs entirely within the fracture system, or primary porosity, whereas all solute transport in the bulk rock, the secondary porosity, takes place by diffusion (see, for example, Barenblatt et al. 1960, Grisak & Pickens 1980). The porosity of the rock matrix can be very small (about 0.1 to 1 %) in many hard rocks and diffusion into the matrix is likely to be slow. The impact of matrix diffusion will, as a consequence, not be significant along short or rapid flow paths but, for the longer and slower paths likely in a repository host rock, it may result in a significant reduction in the maximum concentration of radionuclides in the groundwater entering the biosphere. This process is particularly important if the resulting transport time to the biosphere is greater than the half-life of the radionuclide, since the total release can be reduced by several orders of magnitude (see, for example, Neretnieks 1980 for discussion).

In addition, pulse releases can be spread over longer time periods, thereby decreasing release concentrations by a process of temporal dilution (Nagra 1985). For non-sorbed radionuclides, this process also represents an important retardation mechanism, since these radionuclides would otherwise be transported at the advection rate of the groundwater. Thus the importance of matrix diffusion lies in the fact that it provides a mechanism for enlarging the rock surface in contact with the advecting solute, from that of the fracture surfaces and their infills, to a much larger portion of the bulk rock.

The theoretical basis for matrix diffusion is thus fairly well established and it has since been verified both in laboratory experiments (e.g. Garrels et al. 1949, Heath 1995), URL experiments (e.g. Birgersson & Neretnieks, 1990, Möri et al. 2003) and site characterisation studies (e.g. Ohlsson et al. 2001, Löfgren 2007). As always, both these approaches are relatively short-term, so much effort has been expended in examining long-term matrix diffusion via natural analogues. The usual approach is to carry out studies on natural tracers in rock where perturbations to the average chemical composition of the bulk rock are used to indicate the presence of previous rock-water interaction. The distribution of a range of indicator elements is examined along a profile, usually away from a known (or suspected) water-conducting fracture in the bulk rock. In an ideal case, matrix diffusion would result in a smooth concentration profile away from the fracture but heterogeneity in the rock and the effects of different geological events on the system will complicate this picture.

Usually, the natural decay series, rare-earth elements and a suite of other redox-sensitive elements are studied with the intention of covering a range of geochemical behaviour, thus increasing the chances of obtaining evidence of rock-water interaction. In early studies (e.g. Smellie et al. 1985) a rock core would be taken at an appropriate site and samples along the length of the core would be examined (cf. Figure 9-14) and assessed for indications of disturbance due to rock-water interactions. At the time, matrix diffusion depths of 100s of mm were being reported for fractured crystalline rocks, with depths of up to 500 mm in hydrothermally altered rocks (see discussion in Miller et al. 2000).

Matrix diffusion was also investigated as part of the Palmottu natural analogue study. Radionuclide concentration profiles, from a fracture into the rock matrix, were examined from several different drill cores (Suksi & Ruskeeniemi 1992). Alpha-autoradiography clearly showed the distribution of radionuclides decreasing from the fracture surface into the rock in an almost exponential fashion. Diffuse alpha activity could be identified some 80 mm into the rock matrix in one sample. The alpha-autoradiography also indicated that the radionuclides were associated with clay minerals associated with iron oxyhydroxides, formed from feldspar alteration, and biotite grains. Selective leaching experiments showed that most of the radionuclides that had diffused into the rock matrix were only loosely bound to the iron-rich phases. In some samples, activity correlated to microfractures indicating that fluid flow was channelled even at the microscopic level, whilst, in another sample, diffusion was predominantly along grain boundaries.

Studies were also carried out on fractures at Palmottu located only a few metres away from the mineralisation (Kumpulainen et al. 1992). A range of elemental concentration profiles were measured in rock adjacent to the fractures and these indicated significant alteration over a depth of around 25 mm in one sample but very little alteration in another, highlighting the spatial variability of matrix diffusion depths in natural systems that results from natural heterogeneity in the rock matrix structure and, possibly, the extent of groundwater flow in the fractures due to channelling.

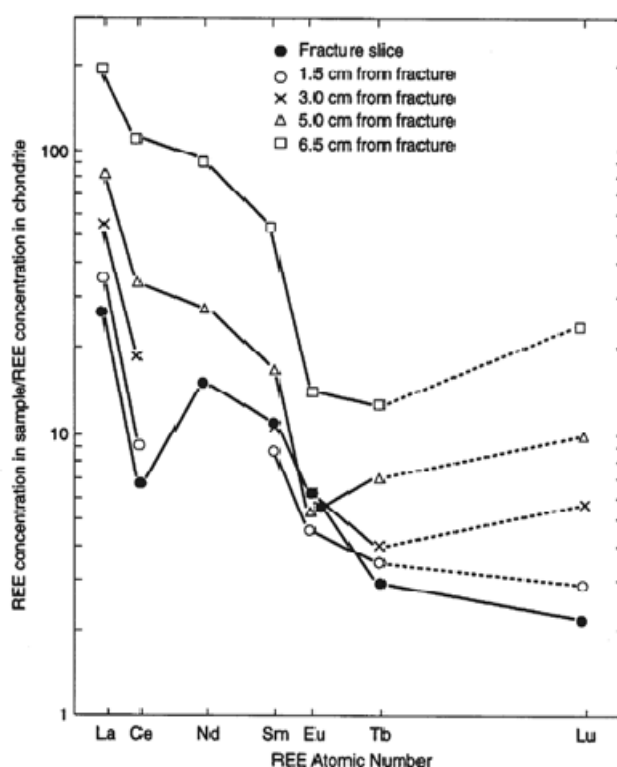


Figure 9-14. Example of early matrix diffusion studies – a sample from the north crystalline basement of Switzerland showing an increased depletion of certain REE from the rock matrix as the fracture is approached (from Alexander et al. 1990).

Conclusions

The rock matrix of fractured crystalline rock is most certainly connected, to some degree or other, throughout and, by slow, diffusive exchange between the porewater and the neighbouring groundwaters, remains in a state of constant dynamic flux. In effect, the majority of the rock matrix porewater can be assumed to be accessible to matrix diffusion as long as a concentration gradient exists between the groundwaters in the fractures and the rock matrix porewaters. Numerous natural analogue and URL studies now exist that confirm matrix diffusion will occur at tens (to hundreds) of metres from a water-conducting feature, in contrast to earlier safety assessments which assumed limited access (e.g. JNC (2000a) assumed a limited diffusion depth of 10 mm).

9.3 Limited impact of natural groundwater colloids

Colloids (that is particles in the size range 1–1000 nm¹⁸) have the potential to enhance the migration of radionuclides away from a deep repository owing to their ability to transport sorbed radionuclides. For colloids to play a significant role, they must be present, stable in the groundwater both chemically (with respect to dissolution) and colloidally (with respect to aggregation and sedimentation processes), and mobile over significant distances through the groundwater system (Figure 9-15). There must also be a significant association of radionuclides with colloids.

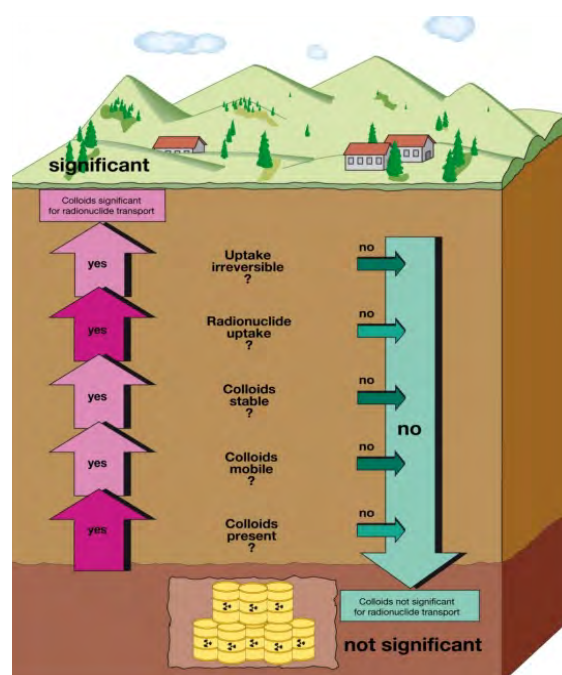


Figure 9-15. The colloid ladder: for colloids to be relevant to geosphere transport of radionuclides, they have to fulfil all of the steps on the left hand side. Just one ‘no’ is required to send them tumbling back down the ladder. After Chapman et al. (1993).

¹⁸ Although inaccurate, the term ‘nanoparticle’ is currently finding favour in the scientific literature (Alexander et al. 2011).

Colloid-facilitated radionuclide transport is potentially important for radionuclides of low solubility that will also sorb strongly to mineral surfaces (such as thorium, americium and plutonium[IV]) and whose retardation in the geosphere may be reduced by association with mobile colloids.

Generally, colloid composition reflects that of the host rock locally – predominantly primary silicates and rock weathering products such as clays etc. – and so the colloids are relatively insoluble in the associated groundwater. Measured deep groundwater colloid populations range across seven orders of magnitude (typically from 10^{-5} to >100 mg/L) and there is no straightforward distinction in concentrations between rock types. Inorganic colloids tend towards aggregation and rock surface attachment (surface charge, PZC (point of zero charge) at low pH etc.). In deep groundwaters, organic colloid populations are generally low other than near formations of biogenic origin (for example oil reservoirs and coal sands) (Stevenson 1994 and Alexander et al. 2011).

Considering the number of deep, research-oriented boreholes that exist worldwide, there are few datasets for groundwater colloids. Those that do exist come from three main sources:

- repository site characterisation;
- URL programmes;
- natural analogue studies.

These data have been summarised in Table 9-3 (from Alexander et al. 2011). In comparison, the colloid concentration at Olkiluoto is between 10^{-6} and 0.2 mg/L (Järvinen et al. 2011). In addition to the scarce data on deep groundwater colloids, much work has been focused on shallow groundwater colloids (see, for example, the review of Bradford & Torkzaban 2008), but this area will not be addressed here as it is of little relevance to a deep geological repository. Similarly, the extensive literature on the colloids from the clean-up of the various nuclear research sites in the US has been omitted as the work generally deals with the shallow (i.e. tens of metres deep) systems. In the same vein, due to the massive differences between the environment of the Yucca Mountain site in the USA and Olkiluoto, information from the former site has also been excluded. However, although the vast majority of the work done on the Gorleben site in Germany (e.g. Plaschke et al. 2002) is focused on the shallower groundwaters, these data have been included as they are some of the few that relate to organic colloids.

As noted by Degueldre et al. (1996), colloids display an inverse correlation between colloid populations and groundwater ionic strength (see Figure 9-16) and the size distribution shows inverse power law scaling behaviour down to 10 – 100 nm colloid diameter (although the contribution of the smallest sizes is difficult to estimate due to analytical limitations – see discussion in Alexander et al. 2011).

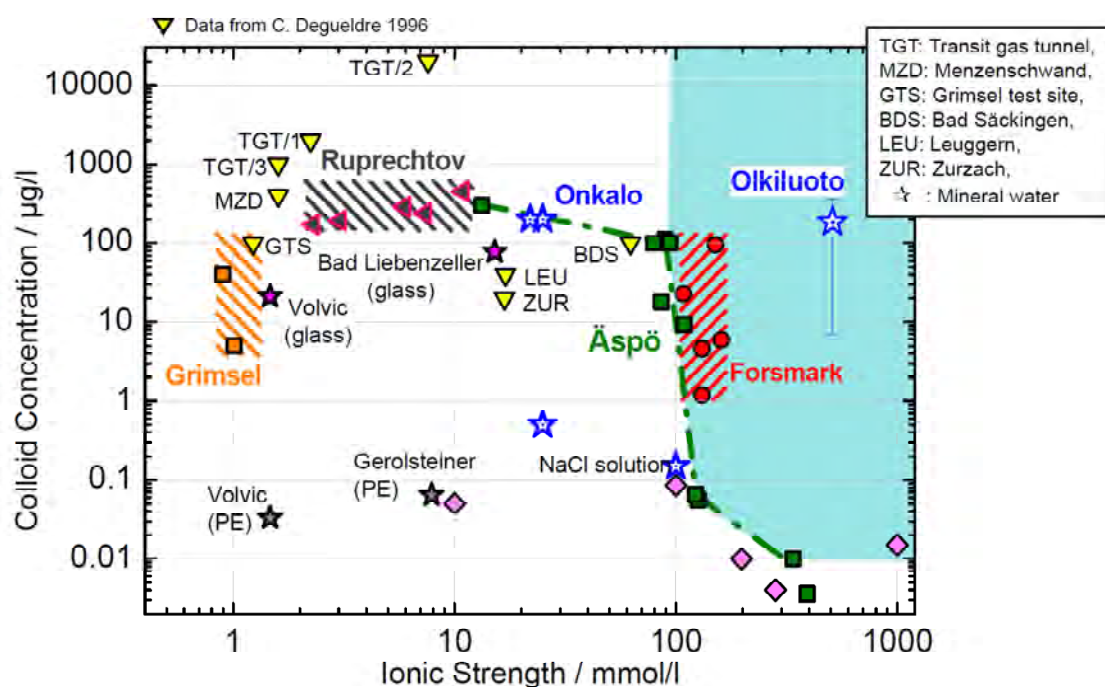


Figure 9-16. Comparison of inorganic colloid concentration in different types of natural groundwater, mineral water and synthetic NaCl-solution versus ionic strength. Data from Degueldre et al. (1996) is represented in yellow filled inverted triangles; sampling sites are - TGT: Transit Gas Tunnel, Haslital (Switzerland), MZD: Menzenschwand uranium prospect (Germany), GTS: Grimsel Test Site, BDS: Bad Säkingen borehole (Switzerland), LEU: Leuggern borehole (Switzerland), ZUR: Zurzach borehole (Switzerland). Data from Hauser et al. (2007) based on laser induced breakdown detection (LIBD) are shown as symbols and hatched areas are giving the variation limit: Grimsel (Switzerland); orange hatch and orange squares- Ruprechtov (Czech Republic); grey hatch and rotated pink triangles with gray fill- Forsmark site (Sweden); red hatch and red filled circles- Äspö URL (Sweden); green hatched trend line and green filled squares. For comparison, data on colloid concentration of mineral water purchasable from Gerolsteiner: Gerolsteiner mineral water plant (Germany) and Volvic: Volvic mineral water plant (France) filled in glass bottles (purple filled stars) and PE (Polyethene) bottles (gray filled stars) and synthetic NaCl salt solutions of different ionic strength (purple diamonds) are inserted. Full sampling details in Degueldre et al. (1996) and Hauser et al. (2007). Additionally inserted is data from Olkiluoto (Laaksoharju et al. 1993) and ONKALO (Järvinen et al. 2011); blue stars. The uniformly blue shaded area indicates the ionic strength range, where destabilisation of natural inorganic colloids is expected.

In general, under steady-state conditions, the majority of colloids in a rock are immobile – but these could be mobilised by physical and chemical disturbance (colloid sampling in boreholes being a leading contender). Consequently, Alexander et al. (2011) noted that the vast majority of data reported so far must be assumed to include artefacts and so to generally represent maximum concentrations. Proven, artefact-free sampling is possible (see site 5 in Table 9-3: Laxemar, $2\text{--}12 \cdot 10^{-3}$ mg/L), but the effort involved is so great that such data are currently so rare as to be statistically meaningless.

With respect to EBS-derived colloids moving into the geosphere, the following points can be made based on natural analogue studies:

- Cementitious system colloids are unlikely due to the high ionic strength of the leachates, with dissolution in the geosphere likely for any that exist (Wetton et al. 1998, Swanton et al. 2010);
- But colloids derived from cementitious leachate-host rock interaction are more problematic to define due to the ever-changing chemical conditions; appropriate data are very rare (Swanton et al. 2010);
- HLW/spent nuclear fuel colloids are unlikely as long as the bentonite functions effectively as a colloid filter and diffusion barrier (cf. Cramer & Smellie 1994, Voegelin & Kretzschmar 2002);
- For colloids generated at the bentonite buffer/host rock interface, little is known. Very few natural analogue data exist (e.g. Kuno et al. 2002) and laboratory results are contradictory (Alexander et al. 2011). Little of relevance exists from URL studies to date – although in the Äspö Colloid Project, bentonite colloids were shown to be unstable in the high ionic strength deep Äspö (and Olkiluoto) groundwaters (Laaksoharju & Wold 2005) and so the populations were also as low as the natural groundwater colloids. Bentonite colloids in low ionic strength groundwaters are currently being addressed in the CFM (Colloid Formation and Migration) project in Grimsel (see www.grimsel.com).

Given the significant number of studies of colloid transport (Table 9-3), remarkably little evidence exists for the long-distance transport of inorganic colloids. Long-distance transport of colloids in the Krunkelbach uranium prospect in the small Black Forest village of Menzenschwand was noted in Degueldre et al. (1996). While there was evidence of transport of colloids from the nearby Feldberg gneiss/metasediment formation some 5 km away, it must be remembered that:

- the Krunkelbach uranium prospect seriously disturbed the local groundwater flow system, drawing in up to 2000 L/min, equivalent to 14 % of the precipitation in the mine's surficial catchment area;
- the main fracture sampled was ~10 cm wide and contained no fault gouge – such a wide-open fracture would be unusual in most repository host rocks;
- as only four samples were analysed, the results are statistically weak.

The Morro do Ferro site in Brazil has a long record of investigations (e.g. Eisenbud et al. 1982), mainly because it is the most naturally radioactive place on the Earth's surface. The advantage of Morro do Ferro is that the Th-REE ore body (Figure 9-17) sits at the top of a hill, about 140 m above a local stream and flow from the ore to the stream was thought easy to intercept, providing a clear-cut case of colloid mobility or retardation. In fact, no colloids from the ore body were found in the lower boreholes nor in the stream at the base of the hill and no clay colloids were observed in the lowest borehole (MF12, Figure 9-17[right]), despite being observed in the groundwaters sampled in the upper boreholes. As noted in Alexander et al. (2011), the colloids analysed also showed little evidence of prolonged transport, all of which indicates efficient colloid retardation in the groundwater flow system at the site.

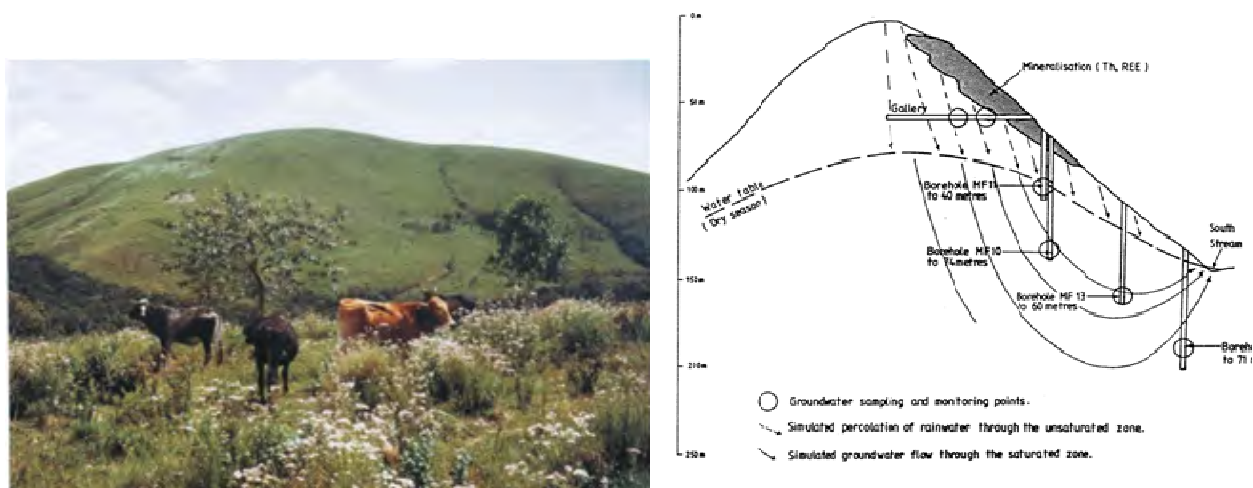


Figure 9-17. Morro do Ferro (the Hill of Iron). Left, although the most naturally radioactive place on earth, the hill is farmed by a local family. Right, cross-section through Morro do Ferro showing the ore body, gallery and the boreholes (from Miekeley et al. 1991).

The Cigar Lake orebody is described briefly in Section 8.1 and illustrated in Figures 8-1 and 8-2. Colloid concentrations in the Cigar Lake groundwaters are low (0.03 to 2.21 mg/L, see Table 9-3, site 19) and consist primarily of clay minerals (illite, chlorite and kaolinite), amorphous Fe-Si precipitates, quartz and organic material and were found to be fairly uniform throughout the deposit. The uranium concentration of colloids down-flow of the ore body was no higher than the concentrations upstream, indicating the absence of any uranium ‘plume’ in the sandstone. The colloids collected within the illite halo, in contact with the ore body, were of a similar composition to those collected in the sandstone, but had higher uranium concentrations, providing further support for the thesis that the clay halo (even though it is illitic and not montmorillonitic) provides an efficient barrier to colloid transport.

Conclusions

Although a wide range of colloid populations have been reported in the literature for deep groundwaters (Table 9-3), most values almost certainly reflect sampling artefacts. Few proven artefact-free data exist, so the values are statistically weak. The few relevant examples of colloid transport indicate that retardation of colloids (and associated radionuclides) appears to be efficient – but, it must be emphasised, there is currently little mechanistic understanding of the processes (straining/sieving, attachment to fracture faces etc.) involved (see Swanton et al. 2010, Alexander et al. 2011 for a detailed discussion).

Table 9-3. Examples of groundwater colloid populations in various host rocks and groundwaters (from Alexander et al. 2011, where detailed information on the sampling and analytical techniques are reported in full).

Site	Rock type and groundwater	Flow system	Colloid number (l)	Size and form	Sampling and analytical methods	Colloid composition	Comments
1. Canadian Shield (Whiteshell Research Area), Canada	Canadian Shield (crystalline) with deep saline (pH 6.1–7.7) or shallow Na-Ca-HCO ₃ type (low ionic strength), pH 7.4–8.9 groundwaters.	Fractures	Range 0.04 to 14 Mean 0.34 ± 0.34	10–450 nm	Sequential ultrafiltration (colloid concentrations calculated from compositions)	Iron oxides, aluminosilicates (probably clays) and carbonates	URL and so flow conditions disturbed. No significant difference in populations between shallow and deep waters. No observable ionic strength effect.
2. Canadian Shield (Atikokan), Canada [i]	Canadian Shield (crystalline) with deep saline or shallow Na-Ca-HCO ₃ type (low ionic strength).	Fractures	Mean 2.4 ± 0.3		Sequential ultrafiltration		Suggested that highly fractured and altered rock has greater surface area to produce colloids than at Whiteshell URL.
3. Fennoscandian Shield (Äspö), Sweden [ii]	Granitoids with basic sills and xenoliths and dykes of fine-grained granite.	Fractures and deformation zones	<0.1 [232] <0.3 [iii] $0.01 \cdot 10^{-3}$ to $1.00 \cdot 10^{-3}$ [iv]	19–993 nm	PCS Ultrafiltration (SEM/EDS) LIBD	Organics, inorganic colloids (clay, calcite, ironoxo-hydroxide) and microbes	URL and so flow conditions disturbed.
4. Fennoscandian Shield (Forsmark), Sweden	Granite with muscovite/biotite, feldspar, quartz and clays (chlorite and illite). Groundwater Na-Ca-Cl type, pH=7.53 Eh=–203 mV.	Fractures and deformation zones	0.14 to $1.8 \cdot 10^{-3}$		Sequential ultrafiltration and SEM/EDS	Clay	From borehole KFM11A, at 391 m depth.

Table 9-3 continued. Examples of groundwater colloid populations in various host rocks and groundwaters (from Alexander et al. 2011, where detailed information on the sampling and analytical techniques are reported in full).

Site	Rock type and groundwater	Flow system	Colloid number (l)	Size and form	Sampling and analytical methods	Colloid composition	Comments
5. Fennoscandian Shield (Laxemar), Sweden [v]	Porphyritic granitoids and red/grey granites to quartz monzodiorite. Groundwater pH 8.2–8.5, Eh -277 mV.	Fractures and deformation zones	6–12·10 ⁻³ 2–5·10 ⁻³		Sequential ultrafiltration, with mass calculation from the filter chemistry LIBD	Illite, calcite, iron and manganese hydroxide (colloid) compositions calculated from the filtrate chemistry)	Very careful groundwater and colloid sampling.
6. Fennoscandian Shield (Oikiluoto), Finland	Migmatite granite with tonalite and granodiorite with Na-Ca-Cl ground-water, anoxic, pH–8.5.	Fractures and deformation zones	0.020–0.450 0.028–0.142 0.039–0.091	20–450 nm 50–2500 nm 2–1000 nm	Ultracentrifugal deposition, ultrafiltration and AF4 with SEM/EDS	Clays, silica, pyrite, goethite and magnesium oxide	URL so site is disturbed by tunnels. High salinity groundwater will induce colloid agglomeration.
7. Fennoscandian Shield (Palmottu), Finland [vi]	Mica gneiss and granite, groundwater is split between shallow Ca-HCO ₃ to Na-HCO ₃ types and deep Na-SO ₄ and Na-Cl types. pH 7.0–8.5 (100 m) to 9.0–9.5 (300 m), anoxic. I=30 mmol/L at depth.	Fractures and deformation zones	0.05 0.89	290 nm 217–229 nm	Ultrafiltration for PCS and EDAX	Silica gel and organics	Although uranium ore is present at the site, the only disturbance is from sampling boreholes. In the deepest, stagnant water (>400 m), no colloids were detected.
8. Grimsel Test Site, Switzerland [vii]	Granite and granodiorite, low salinity Na-Ca-HCO ₃ -F groundwater, sub-oxic, pH 9.6.	Fractures filled with fault gouge	Range 0.13 to 13 4.2·10 ⁶ to 3.9·10 ¹⁰	Angular to rounded, 20–450 nm	Suite of methods used	Silica, phyllosilicates (illite/muscovite/biotite) and calcium silicates (probably plagioclase)	Low salinity water induces colloid stability. URL and so flow conditions disturbed.

Table 9-3 continued. Examples of groundwater colloid populations in various host rocks and groundwaters (from Alexander et al. 2011, where detailed information on the sampling and analytical techniques are reported in full).

Site	Rock type and groundwater	Flow system	Colloid number (l)	Size and form	Sampling and analytical methods	Colloid composition	Comments
9. Transitgas Tunnel, Switzerland	Granite and granodiorite, gneiss and mafic amphibolite, low salinity Na-Ca-SO ₄ -HCO ₃ groundwater, oxic, pH 8–9.6.	Fractures (no detailed description)	$<2 \cdot 10^{-4}$ to $<2 \cdot 10^{-3}$	10–1000 nm	Cross-filtration, bulk water for SPC and colloids on membrane for XRD	Clay (including illite), silica	Collected from seeps in a tunnel, therefore drawdown disturbance.
10. Menzenschwand, Germany	Granite intruded into gneiss and metasediment, Ca-HCO ₃ water, oxic, pH 6.5.	Large aperture fracture	$<4 \cdot 10^{-4}$	10–1000 nm	Cross-filtration, bulk water for SPC and colloids on membrane for XRD	Clay, silica	Uranium prospect, heavy pumping of water (up to 2000 L/min) therefore significant disturbance of the local groundwater system. Evidence of long-distance (1–5 km) transport of colloids.
11. Leuggern, Switzerland	Mica-granite, Na-HCO ₃ -Cl-SO ₄ water, strongly anoxic, pH 7.8	Not described, assumed to be fractures	<20	10–1000 nm	Cross-filtration, bulk water for SPC and colloids on membrane for XRD	Silica, clay	Water pumped 1700 m to the surface, so degassed.
12. Fanay-Augères, France [viii]	Granite with Na-Ca-HCO ₃ -SO ₄ -Cl groundwater.	Fractures	$<0.1 \cdot 10^{-3}$	100–1000 nm		Silica and organics	Former uranium mine, so significant disturbance to the system.

Table 9-3 continued. Examples of groundwater colloid populations in various host rocks and groundwaters (from Alexander *et al.* 2011, where detailed information on the sampling and analytical techniques are reported in full).

Site	Rock type and groundwater	Flow system	Colloid number (l/l)	Size and form	Sampling and analytical methods	Colloid composition	Comments
13. El Berrocal, Spain	Two-mica granite (with leucogranite and pegmoaplites), groundwater Na-Ca-HCO ₃ -Cl type to Ca-Na-HCO ₃ -Cl type, pH 6.2 to 7.3, oxic, I= 6–11 mmol/L.	Fractures	<0.1 10 ¹² 10 ¹⁰ 10–10 ⁸	down to 1 nm down to 50 nm 208 to 342 nm [239]	Tangential flow ultrafiltration with TEM and PCS/SEM	Quartz/silica, iron oxy-hydroxides and alumina gels Albite, apatite, clays, calcite, other carbonates and Al- and Fe-oxyhydroxides. Some organics too [239]	Shallow (S1 and S7) boreholes in a former uranium mine, so heavily disturbed site [ix]. Boreholes were left open for a long time and "...geochemical stabilisation...may not have been reached at all....sampled water may represent mixtures...". Majority of U and Th in solution.
14. Sellafield, UK	Borrowdale Volcanic Group.	Fractures			Tangential flow ultrafiltration and STEM/EDAX	Fe-oxyhydroxides	Borehole 9A, drilled specifically for geochemical sampling. No detailed analysis carried out due to interference from Fe-oxyhydroxides on filters.
15. Cigar Lake, Canada [x]	Zone of altered crystalline basement, Na-Ca-HCO ₃ -Cl groundwater, oxic to anoxic, pH 6.1 to 7.8	Heavily damaged zone, crushed rock Fracture in basement	7.78 ± 4.14 0.03 to 2.21		Tangential flow ultrafiltration and colloids on membrane for XRD and SEM/EDX	Clays (illite, chlorite, kaolinite), amorphous Fe-Si oxides, quartz and organics	U deposit has been explored with several hundred boreholes, so significant disturbance.

Table 9-3 continued. Examples of groundwater colloid populations in various host rocks and groundwaters (from Alexander et al. 2011, where detailed information on the sampling and analytical techniques are reported in full).

Site	Rock type and groundwater	Flow system	Colloid number (l)	Size and form	Sampling and analytical methods	Colloid composition	Comments
16. Osamu Utsumi, Brazil [xi]	Phonolite, K-Fe-SO ₄ , oxidising and weakly to strongly acidic groundwaters, oxic.	Fractures (with extensive redox front development near surface)	<0.5 (generally 0.1 to 0.2) by filtrate chemistry		Sequential (cascade) cross-flow ultrafiltration (filter size 450 to 1.5 nm) with ESCA, SEM, XRD	DOC (80–90 %), Fe-oxyhydroxides (2–20 %) and clay±chalcedony (<2 %)	U deposit extensively mined, therefore highly disturbed site. Groundwater flow from depth to surface, the opposite of the state before mining.
17. Morro do Ferro, Brazil	Phonolite, groundwater varies from the bottom of the hill, which resembles the water from Osamu Utsumi (above), to the ore body at the top of the hill which is similar but less mineralised and more oxidising, representing unsaturated conditions.	Fractures in heavily weathered host rock	0.01 to 0.10 by SEM 0.26 by filtrate chemistry $3.4 \cdot 10^7$	Large (0.5–1.0 µm), irregular/angular shaped particles, agglomerates (5–10 µm) of these particles and long (humic) chains (2–3 µm)	Sequential (cascade) cross-flow ultrafiltration (filter size 450 to 1.5 nm) with ESCA, SEM and XRD	DOC (80 %), Fe-oxyhydroxides (18 %) and clay (2 %)	Th, REE deposit has been explored with a drift and several boreholes, so some disturbance. Colloid concentration range represents the same borehole, but different samples.
18. Wellenberg, Switzerland [xii]	Tectonised marl (clay-rich limestone).	Heavily tectonised fractures		Colloids are cylinders 30–100 nm diameter and 5–15 nm long. Mean diam. 72 nm	Cross-filtration and colloids on membrane for AFM (atomic force microscopy) LIBD [206]	Clay	Pumped from depth, but borehole was supposedly hydrochemically 'stable'.

Table 9-3 continued. Examples of groundwater colloid populations in various host rocks and groundwaters (from Alexander et al. 2011, where detailed information on the sampling and analytical techniques are reported in full).

Site	Rock type and groundwater	Flow system	Colloid number (l/l)	Size and form	Sampling and analytical methods	Colloid composition	Comments
19. Cigar Lake, Canada	Sandstone and ore (within clay halo), Na-Ca-HCO ₃ -Cl groundwater, oxic to anoxic, pH 6.1 to 7.8.	Porous matrix	Mean 0.6±0.09 to 1.52±0.29	10 nm to 20 µm, but most in 100–400 nm range	Tangential flow ultrafiltration and colloids on membrane for XRD and SEM/EDX	Clays (illite, chlorite, kaolinite), amorphous Fe-Si oxides, quartz and organics	No significant U association with the colloids found downflow of the ore body.
20. Sellafield, UK	St Bees Sandstone Formations.	Porous matrix	0.5 8·10 ⁻¹⁰	100–200 nm	Tangential flow ultrafiltration and STEM/EDAX	Silica, coated with iron oxyhydroxide associated with an illitic clay mineral. Significant amounts of Ca, S, Zn and As also associated with these colloids	Borehole RCF3 (NB long-term pump test on-going, so likely to be disturbed).
21. Oklo and Bangombé natural reactors, Gabon [xiii]	Sandstone, conglomerates and clays, Na-Mg-Ca-HCO ₃ groundwater, pH 6–7, slightly negative Eh.	Porous matrix	0.03 to 0.92 2·10 ⁵ to 2·10 ⁷	>100 nm	Colloids on membranes for SEM/EDAX and ICP-MS, bulk water samples for SPC	Silica, clay and iron oxyhydroxides	Sampled from 4 shallow (<105 m) boreholes below the reactor zone. Bacteria and organics examined in parallel, but no attempt to link the results.
22. Zurzach, Switzerland	Sandstones and marls directly above crystalline basement, Na-HCO ₃ -Cl-SO ₄ water, strongly anoxic, pH 8.0.	Not defined, possibly fractures in sediments	<10	10–1000 nm	Cross-filtration, bulk water for SPC and colloids on membrane for XRD	Silica, clay	Pumped from 470 m deep so disturbed. Note similarity of water type to Leuggern; likely that groundwater from the crystalline basement is being drawn in.

Table 9-3 continued. Examples of groundwater colloid populations in various host rocks and groundwaters (from Alexander *et al.* 2011, where detailed information on the sampling and analytical techniques are reported in full).

Site	Rock type and groundwater	Flow system	Colloid number (l/l)	Size and form	Sampling and analytical methods	Colloid composition	Comments
23. Ruprechtov, Czech Republic [xiv]	Clays (altered tuff), lignite and altered granite, Ca-HCO ₃ groundwater, I=0.003 to 0.02 mol/L, oxic to anoxic, pH 6.2–8.0.	Zones of higher hydraulic conductivity in the clay lignite [xv]	0.003 to 0.744	Most >45 nm	LIBD	No data	Samples from shallow boreholes (max. depth 37 m), some questions regarding Eh status of system (varies with time).
24. Alligator Rivers, Australia [xvi]	Quaternary sands over clay-rich zone of intensely-weathered schist grading into weathered schist. Mg-Ca-Si-HCO type groundwater, pH~7, oxic, I=1–1.5 mol/L.	Not described in detail, seems to be permeable zones in the sands and clays.	<0.05 102 to 105	Platelets, spheres, crystals (quartz, Fe-oxide) up to 1 µm diameter	Hollow-fibre ultrafiltration and direct ultrafiltration for ICP-MS and SEM/TEM/EDS	Quartz, Fe-rich colloids and clays. In highly weathered zone, colloidal clay is kaolinite, in less weathered zones, chlorite. Also uranyl silicate and lead	Uranium ore not mined, but over 100 exploration boreholes drilled. Site also experiences significant seasonal fluctuation of the water table.
26. Tsukinuno, Japan	Bentonite beds intercalated with shales.	Not described, but fractures likely.	None detected, presumed to be due to groundwater chemistry		Ultrafiltration and ICP-AES (colloid concentrations calculated from [Si, Mg and Al])	Assumed to be bentonite, but not presented	Bentonite deposit is mined, so highly disturbed site. Colloid detection limit is not presented.

Abbreviations:

AFM Atomic force microscopy
 BSE-SEM Backscattering electron scanning electron microscopy
 DOC Dissolved organic carbon
 EDX (EDAX) Energy dispersive X-ray analysis
 ESCA Electron spectroscopy for chemical analysis
 ESEM Environmental scanning electron microscopy
 EXAFS (XAFS) Extended X-ray absorption fine structure (spectroscopy)
 ICP-AES (ICP-OES) Inductively coupled plasma-atomic (optical) emission spectroscopy
 ICP-MS Inductively coupled plasma-mass spectrometry
 LIBD Laser-induced breakdown detection
 PCS Photon correlation spectroscopy
 RBS Rutherford backscattering spectroscopy
 SEM Scanning electron microscopy
 SPC Single particle counting
 STEM Scanning-tunnelling electron microscopy
 TEM Transmission electron microscopy
 TR-LFS Time-resolved laser fluorescence spectroscopy
 XRD X-ray diffraction

Notes:

- i. Vilks, P., Bachinski, D.B. & Vandergraaf, T.T. 1991. *The role of particulates in radionuclide transport*, in Advanced Nuclear Engineering Research - Global Environment and Nuclear Energy, Proceedings of Third International Symposium, Mito City, Japan, p. 394-401.
- ii. Ledin, A., Düker, A., Karlsson, S. & Allard, B. 1995. *Measurements of colloid concentrations in the fracture zone, Äspö Hard Rock Laboratory, Sweden*. SKB Technical Report TR-95-17
- iii. Vuorinen, U. 2005. *Characteristics of natural colloids in two groundwater samples from the Äspö HRL tunnel*. Appendix 5 in Laaksoharju, M. & Wold, S. *The colloid investigations conducted at the Äspö Hard Rock Laboratory during 2000–2004*. SKB Technical Report TR-05-20.
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- v. Bergelin, A., Nilsson, K., Lindquist, A. & Wacker, P. 2008. *Oskarshamn site investigation. Complete chemical characterisation in KLX13A Results from two investigated borehole sections: 432.0–439.2 m, 499.5–506.7 m*. SKB Report P-07-149.
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- x. Vilks, P., Cramer, J.J., Bachinski, D.B., Doern, D.C. & Miller, H.G. 1993. *Studies of colloids and suspended particles. Cigar Lake Uranium Deposit, Saskatchewan, Canada*. Appl. Geochem. Vol. 8, p. 605-616.
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- xii. Degueldre, C. 1997. *Groundwater colloid properties and their potential influence on radionuclide transport*. Sci. Basis for Nucl. Waste Man. XX, edited by Gray, W.J. & Triay, I.R. Mat. Res. Soc. Symp. Proc. Vol. 465, p. 835-846.
- xiii. Pedersen, K. (ed.). 1996. *Bacteria, colloids and organic carbon in groundwater at the Bangombé site in the Oklo area*. SKB Technical Report TR-96-01.
- xiv. Hauser, W., Geckeis, H., Götz, R., Noseck, U. & Laciok, A. 2007. *Colloid detection in natural ground water from Ruprechtov by laser-induced breakdown detection*. 2nd Annual Workshop Proc. of the Integrated Project 'Fundamental Processes of Radionuclide Migration' 6th EC FP IP FUNMIG, November 21-23, 2006, Stockholm. SKB Technical Report TR-07-05, p. 367-74.
- xv. Noseck, I., Brasser, T., Suksi, J., Havlova, V., Hercik, M., Deneke, M.A. & Förster, H-J. 2008. *Identification of U enrichment scenarios by multi-method characterisation of immobile U phases*. Phys. Chem. Earth. Vol. 33, p. 969-977.
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10 ANALOGUES FOR POTENTIAL FUTURE CONDITIONS IN THE SURFACE ENVIRONMENT

The analogues discussed so far in this report have been used to support our current understanding of processes, to provide evidence of long-term stability of the materials in the EBS and the host rock, including deriving data, or to demonstrate for non-technical audiences the safety functions of the KBS-3 barriers and the concepts on which long-term safety is based. Analogues used in the surface environment are somewhat different as they stand as representatives of credible lines of future evolution at the site. Postglacial land uplift (approximately 6 mm/a; Eronen et al. 1995) profoundly affects future ecosystems of Olkiluoto and its surroundings. For instance, new lakes and mires will develop in the sea area now surrounding Olkiluoto and their properties can be understood based on the properties of the present lakes and mires in the Reference Area, which is an area approximately 100x200 km² large that encompasses the Olkiluoto site within it (the *Biosphere Description* report, sections 1.4 and 2.3; Haapanen et al. 2010, 2011).

As discussed in Chapter 2, the regulatory requirements (STUK's Guide YVL D.5, draft 4) for the time window to be assessed by quantitative dose calculations are for a period that shall extend "at a minimum over several millennia" after the closure of the repository and "during which the radiation exposure of humans can be assessed with sufficient reliability". The biosphere assessment in general has to be based on similar types of climate, human habits, nutritional needs and metabolism as currently exist but must take account of reasonably predictable changes in the environment such as land uplift and the consequent emergence of new land areas.

The overall aims of the biosphere assessment in the safety case are to describe the present and identify credible future conditions at, and the prevailing processes in, the surface environment of the Olkiluoto site using knowledge of relevant past conditions, to model the transport and fate of radionuclides released from the repository through the geosphere to the surface environment, and to calculate doses and dose rates to humans and other biota (*Biosphere Assessment*).

For focusing the studies to the past 10 000 years of development, respective to the assessment time window to the future, on the areas geologically, geomorphologically, climatically and regarding the present vegetation zones, a Reference Area (Figure 10-1) has been delineated (Haapanen et al. 2010, 2011). The knowledge basis on the past development of the Olkiluoto site and the Reference area is projected into future development of the site: the description of the past succession lines forms the basis for assessment models regarding the site development, and the description of the ecosystems and biotopes at present feeds into conceptual element storage and flux calculations, which in turn feed into the radionuclide transport models and dose assessment models. With the development of the Olkiluoto site from a coastal into an inland site due to the land uplift, lakes and peat bogs will form that lack from the site at present, and considerably larger areas preferable for agriculture will emerge from the present sea bottom. Especially these ecosystem types are in focus in the characterisation and use of the Reference Area as an analogue. Also, Eurajoki and Lapijoki Rivers are in focus as their mouths are at present close to the site and they are projected to continue their course past the site during the coming several millennia. However, also other river

catchments in the Reference Area are used, too, to ensure the representativeness of the input data to the assessment (the *Biosphere Data Basis* report).

Lakes

New lakes are formed from coastal bays and the shallow offshore sea areas which become isolated from the sea by post-glacial land uplift. The bays progress through a sequence of different stages: they start with an opening or openings to the sea, which are slowly cut off and the development into a lake or a mire starts.

Four lake systems were chosen for further analyses based on their size, depth, time of isolation, characteristics of the catchment area, European Union Water Framework Directive (WFD) type and whether they represent a drainage/headwater/closed lake type (Haapanen et al. 2010):

- A lake chain (Poosjärvi, Kivijärvi and Lampinjärvi)
- A small lake (Valkjärvi or Lutanjärvi)
- An overgrown lake (Koskeljärvi and Suomenperänjärvi).

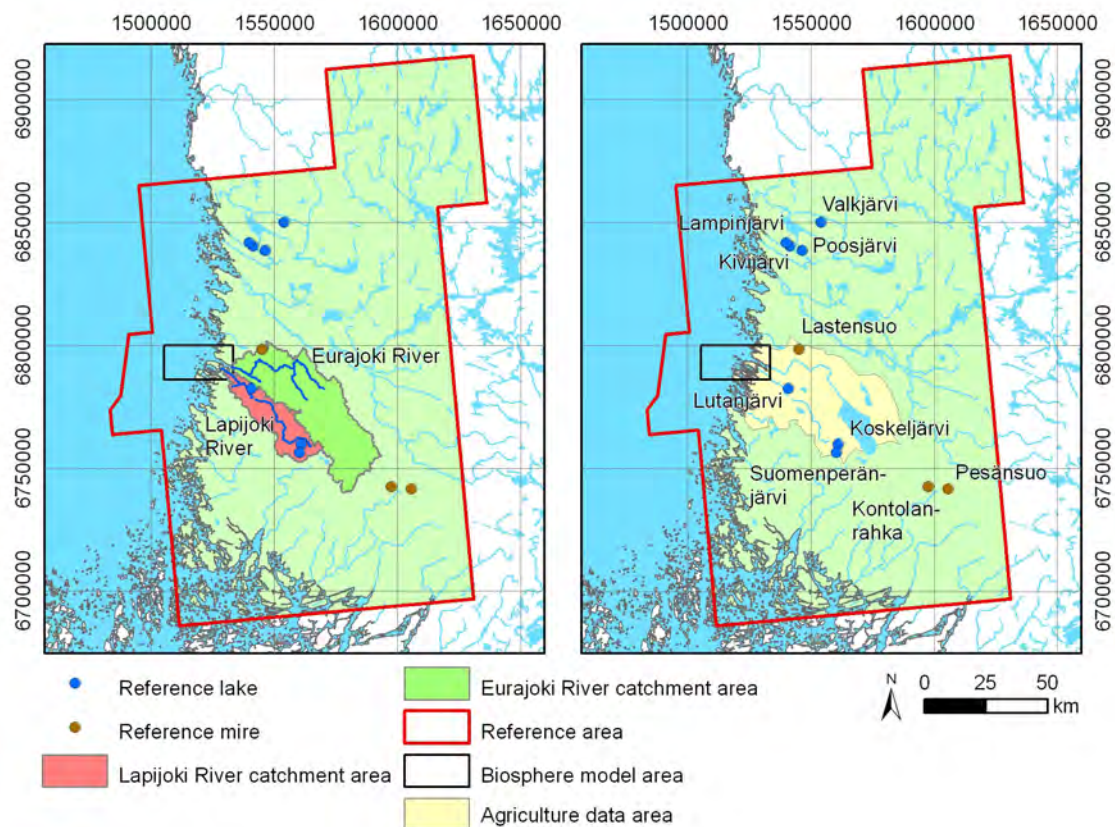


Figure 10-1. Schematic presentation of the Reference Area, the selected reference lakes and mires and the catchments of Eurajoki and Lapijoki Rivers (left), the main area of the agricultural data (right) (*Biosphere Description*, Figure 1-10).

Each represents a specific type of a lake ecosystem also projected for the future, but as the modelling of the long-term development is subject to uncertainties in data and assumptions the assessment model input data representing the ensemble of the selected lakes is considered more appropriate 'analogue' to the ensemble of the projected future lakes than specific pairs.

Mires

A mire is an ecosystem based on peat. The climate, local topography and hydrology are key factors of peat development and spread. Primary mire formation is a process in which the fresh soil surface is occupied directly by mire vegetation after emergence from water. Requirements include poorly permeable land with flat topography or small depressions. Secondary mire formation occurs mainly in places where topography directs enough runoff to create waterlogged conditions. On poorly permeable lands, surface waters may fill depressions, after which wet-loving plants invade. On more permeable ground, the high ground water level causes moist conditions and low oxygen concentration, after which mire vegetation can invade and further accelerate the process. The decomposition of organic material is slowed down, and the development of peat may begin. White mosses (*Sphagnum* spp.) contribute especially significantly to mire development. They absorb and store water efficiently and decrease the diffusion of oxygen to the ground as they (being rootless) do not need it. Sedges (*Carex* spp.), in turn, are peat producers in conditions too moist for white mosses: they have open stems that transport air to roots. Secondary mires can also form from open water by a process known as terrestrialisation. (*Biosphere Description*, section 2.1.5).

Haapanen et al. (2010) describe the mire succession that takes place at the Finnish coast during the present land uplift. The process is variable according to the topography and other factors but it is notable that, as the uplift is continuing, there is a sequence from younger mires at the coast to the older ones at higher elevations. From a large number of individual mires with different characteristics in the Reference Area, in order to represent a range of mires that may develop around the Olkiluoto site with continued land uplift, the following were selected for on-going studies based on their type and availability of pre-existing data to support the focused characterisation (Haapanen et al. 2010):

- A very young mire (Olkiluodonjärvi)
- A young mire (Lastensuo)
- An old and large mire (Kontolanrahka)
- An old and small mire (Pesänsuo).

As with lakes, the ensemble of the mires represent the anticipated mire development than a single mire in its present state and in what can be inferred on its past.

11 DISPOSAL OF SPENT NUCLEAR FUEL IN THE CONTEXT OF OTHER SOURCES OF RADIOACTIVITY

In Chapter 3, naturally-occurring radioactive materials, such as the original U ore mined to provide the uranium that was enriched to form nuclear fuel, were used for comparison with the spent nuclear fuel to illustrate how the activity and radiotoxicity of spent nuclear fuel in the repository evolves over time. The comparisons implicitly assumed that the spent nuclear fuel remained in situ in the repository so that, for example, actinide decay chains remain in secular equilibrium. It is possible that not all the radionuclides from the spent nuclear fuel will remain in the repository but, with one or more canister failures, there will be releases into the geosphere and eventually to the surface environment. Thus it is useful to extend the sort of comparisons made in Chapter 3 to the potential releases from the repository.

This section discusses first how complementary indicators will be applied as support to the dose and activity flux quantities derived in the safety assessment in order to assess compliance with the regulatory radiation protection constraints. The purpose of complementary indicators is to either highlight the performance of a certain component of the disposal system or to represent an alternative line of reasoning. It should be noted that the results from applying these complementary indicators in the safety case is not addressed in this report; these are presented together with the results from the main safety assessment (i.e., in *Assessment of Radionuclide Release Scenarios for the Repository System* and *Biosphere Assessment*).

Thus, this section provides information that can be used for comparison with results from safety assessment. The purpose of such a comparison is not primarily to demonstrate the level of safety provided by the spent nuclear fuel disposal facility itself, but to put the potential radiological impact of releases into perspective, by comparing to the current radiation environment and to other present-day industrial activities giving rise to radiation exposure.

The information provides some background for these alternative indicators for disposal systems and also for, less well appreciated, sources of radioactive waste so that the calculated assessment results can be considered in the broader context of the range of naturally-occurring radionuclides in the environment.

11.1 Complementary indicators

A primary requirement of a radioactive waste disposal system is that “*individuals, society and the environment are protected from harmful effects of ionising radiation, now and in the future, in such a way that the needs and aspirations of the present generation are met without compromising the ability of future generations to meet their needs and aspirations*” (ART 1(ii), IAEA 1997). For the post-closure phase, this can be partly assessed by calculating radiation doses or risks arising from hypothetical repository releases, for example, to a hypothetical human population living in the locality. However, as noted in Chapter 1, it is widely recognised that this method is limited by the assumptions that must be made about the local environment (climate, vegetation) and the life style and practices of the population (IAEA 2003a). For a relatively short period after closure, conditions at the time of closure may be

extrapolated but, at longer times, beyond a few hundreds of years, these assumptions become increasingly untenable. Consequently, other indicators of the level of safety a disposal facility provides have been sought to complement the calculated doses, that are often the primary parameters used when assessing compliance with regulatory criteria. There is no internationally accepted terminology for, or definitions of, different types of indicators. For example, IAEA (2003a) talks about ‘complementary safety indicators’ that can provide alternative comparisons for the repository releases that are expected to be more robust over the long time spans of interest. In the outcome of the PAMINA project (Becker et al. 2009), complementary safety indicators are discussed in the context of indicators complementary to the dose and risk safety indicators, such as radiotoxicity concentration in biosphere water, radiotoxicity flux from the geosphere and the power density¹⁹ in groundwater. Furthermore, PAMINA defines a performance indicator as a quantity, calculable by means of appropriate models, that provides a measure for the performance of a system component, several components or the whole system (Becker et al. 2009). This can for example be activity inventories or concentrations in different compartments of the disposal system. In the present safety case, Posiva is using the term *safety indicator* for the quantities derived in the safety assessment in order to assess compliance with the regulatory radiation protection constraints specifically annual effective doses to most exposed and other people, absorbed dose rates to other biota and activity fluxes from the geosphere (see Section 2.2). All other quantities derived are denoted *complementary indicators*, regardless of whether they are employed to support the level of safety of the disposal system or as a measure of performance of an individual system component.

Comparison of the radionuclide releases from the repository with concentrations or fluxes of naturally-occurring radionuclides allows the significance of the repository releases to be assessed without needing to consider exposure of a specific human population. Natural systems can be used to provide reference values based on stable conditions over long time scales. Moreover, different natural systems could be used to provide such reference values, or a range of values, for alternative environmental conditions (e.g. different climate conditions) without having to make assumptions about possible corresponding changes to human activities. Tables 11-1 and 11-2 show some examples of comparisons that could be made between spent nuclear fuel and radionuclides released from the repository and natural materials.

In addition to considering radionuclides in an undisturbed natural system, the consequences of anthropogenic activities, such as ore mining, mineral processing, fossil fuel extraction and combustion, on natural radionuclide distributions and fluxes can be considered in order to assess how significant repository releases are with respect to these disturbed systems.

¹⁹ Power density is the term used to denote the amount of energy stored in a material or system (per unit volume). For groundwater, which does not store energy in the manner of a fuel such as oil, it is a function of the hydraulic head of the groundwater system as this determines the potential and kinetic energy of the water mass.

Table 11-1. *Natural materials that could be used to make comparisons of radiotoxicity with spent nuclear fuel or with radionuclides released from spent nuclear fuel.*

Radiotoxicity related to the repository	Natural material
Spent nuclear fuel (see also Figure 3-3)	The uranium which was mined to produce the nuclear fuel
	Uranium ores of various grades, e.g. from Palmottu U ore deposit
	Olkiluoto host rock
Radionuclides that have migrated into the geosphere	Olkiluoto host rock
Radionuclides accumulated in the surface environment	Natural soil at Olkiluoto

Table 11-2. *Possible comparisons of radionuclide fluxes originating from the repository with natural radionuclide fluxes (after Table A3.10, Nagra 2002).*

Flux related to the repository	Natural flux
Flux of radionuclides from the repository into the geosphere	Flux of natural radionuclides over the repository area due to erosion
	Flux of natural radionuclides from a weathering U orebody e.g. Palmottu
Flux of radionuclides from the geosphere into the surface environment	Flux of natural radionuclides dissolved in the groundwater within the biosphere aquifer
	Flux of natural radionuclides at the Olkiluoto site prior to repository construction
	Flux of natural radionuclides dissolved in water within typical rivers
	Flux of natural radionuclides due to erosion

A further type of comparison which seeks to provide a wider context for the impact of the disposal system uses naturally-occurring radionuclides that are released during industrial activities. The background to the use of natural radionuclides as comparators is discussed in more detail in Section 11.2.

The following sections discuss the current understanding of natural radionuclide fluxes, and the consequences of disturbing naturally-occurring radioactive materials through industrial processes.

The examples discussed are used to illustrate some of the concerns and uncertainties in estimating fluxes of radionuclides whether in undisturbed natural systems or anthropogenically altered ones. In particular, for the naturally-occurring radionuclides in undisturbed systems, understanding and evaluating the measured concentrations and estimated fluxes in the context of knowledge of the whole hydrogeological system is a

key element. If such parameters are to be used as adjuncts to a safety case, it is essential that their limitations are known.

11.2 Natural radionuclide concentrations and fluxes within the geosphere and to the biosphere

Naturally-occurring radionuclides – the primordial radio-isotopes of potassium, rubidium, thorium and uranium and their progeny are considered here²⁰ – are known to be distributed throughout the earth's rocks, sediments, surface and ocean waters as well as taken up by biota – including humans – so that all surface life is exposed to radiation both externally and internally. The combination of radiation from terrestrial radionuclides with cosmic radiation makes up the natural background radiation. The dose arising from natural background radiation is discussed further in *Biosphere Assessment* but the key point here is that human life has developed against a background of natural radiation exposure. The current level of natural background radiation is, with exceptions usually linked to unusual geological situations, not a significant factor in determining human health (e.g. UNSCEAR 2008) although it cannot be excluded as a consideration, but any releases from a radioactive waste repository should be at a level which does not significantly worsen that situation. The worldwide average annual effective dose from cosmic radiation is a small part of the total natural background radiation: 0.38 mSv out of a total (worldwide average) annual natural background effective dose of 2.4 mSv (UNSCEAR 2008). Cosmic radiation is not further discussed here since the repository has no comparable effect and only terrestrial radionuclides are considered in the following discussion. For comparison the average annual dose from background radiation in Finland is about 3.15 mSv, of which 2 mSv is from inhouse radon, 0.45 mSv from external radiation from ground, 0.3 mSv from cosmic radiation and 0.4 mSv from natural radioactivity in human body (Mustonen 2011). In addition to this, there are annual background doses arising also from non-natural sources, i.e. 0.5 mSv from röntgen examinations, 0.03 mSv from radioisopes used in medicine and 0.02 mSv from the Chernobyl fallout. This totals in 3.7 mSv per year for average effective dose (Mustonen 2011).

Terrestrial radionuclides are lost from crustal rocks over time by the processes of physical and chemical weathering at the Earth's surface by erosion, in the sub-surface by interaction between rocks and groundwater, and in the oceans by interaction with seawater (e.g. Faure 1991). At the same time, they can be removed from the dissolved phase in river water, seawater or groundwater by processes such as mineral formation, adsorption on solids or organic materials, or uptake by biological processes, as part of

²⁰ There are also cosmogenic isotopes formed by the interaction of cosmic radiation with nuclei in the atmosphere. However, with the exception of C-14, H-3, Na-22 and Be-7, which are isotopes of elements with a metabolic function in the body, the cosmogenic isotopes are not significant contributors to overall radiation doses (UNSCEAR 2008). They are mainly considered to be of minor consequence to the discussion here because the combination of their low abundances and, particularly, short half lives ($t_{1/2}$), i.e. – $t_{1/2}$ of H-3 is 12.3 a; Na-22 2.6 a; Be-7 0.15 a, means that they will not be analogues for radionuclides arising from a spent fuel repository. C-14 is different, having a longer half life of 5.7 ka, so potentially useful for comparisons over some thousands of years. However, the environmental concentrations of C-14 are well known as it has an isotopic abundance of about 1 ppt in atmospheric carbon dioxide and living plant material, and this abundance falls due to decay once it is isolated from exchange with atmospheric CO₂. Thus C-14 is not considered further here.

the geochemical cycle. In addition, transport of sediment between land and sea creates a further mass movement of radionuclides. Thus, there is a constant movement or flux of these radionuclides between different states or 'compartments' in the geosphere and surface environment (Figure 11-1). The pathways between compartments represent different processes such as groundwater leaching, fluvial transport or subaerial erosion. In some compartments, radionuclides are available to plants and animals and thus move into the food chain and ultimately into humans.

Miller et al. (1997) considered some characteristics of pathways for natural fluxes which might make them suitable for use in such comparisons:

- The pathway passes through the same material and across the same boundaries as repository-derived radionuclides;
- The pathway has a large natural flux;
- The pathway has material passing along it in a form that is readily taken up by the human body;
- The pathway operates on a long time scale.

It is unlikely that natural pathways will fulfill all four criteria. As Miller et al. (1997) note, the most important repository-relevant pathway, via groundwater transport, is likely to represent a relatively small natural flux because of the hydrogeological criteria used to site a deep geological disposal facility in the first place. However, the radiological risk associated with the groundwater flux may be significant because of the potential for radionuclides in groundwater to be taken up into the food chain and humans via drinking water, irrigation of farmland etc. (Miller et al. 1997).

Since the first IAEA publication considering complementary safety indicators was published (IAEA 1994), there has been a number of further studies considering how natural radionuclide concentrations and fluxes might be used (IAEA 2003a and references therein) reflecting increasing regulatory interest in this area. These publications have recognised that the determination of natural fluxes is not necessarily a straightforward exercise. There are two basic quantities required:

- the concentrations of the radionuclides in the various compartments such as rock, soil, groundwater, and
- the rates of the relevant geological and hydrological processes such as erosion, sedimentation, groundwater flow and discharge, river flow etc.
- The concentrations of elements and radionuclides in surface and groundwaters are relatively well known for many specific locations, for different climatic zones or geological conditions and as regional and global averages or ranges, having long been of interest to geochemists understanding the geochemical cycle (e.g. Livingston 1963). The case is similar for rocks (e.g. Faure 1991 and references therein) and soils (Foth 1984).

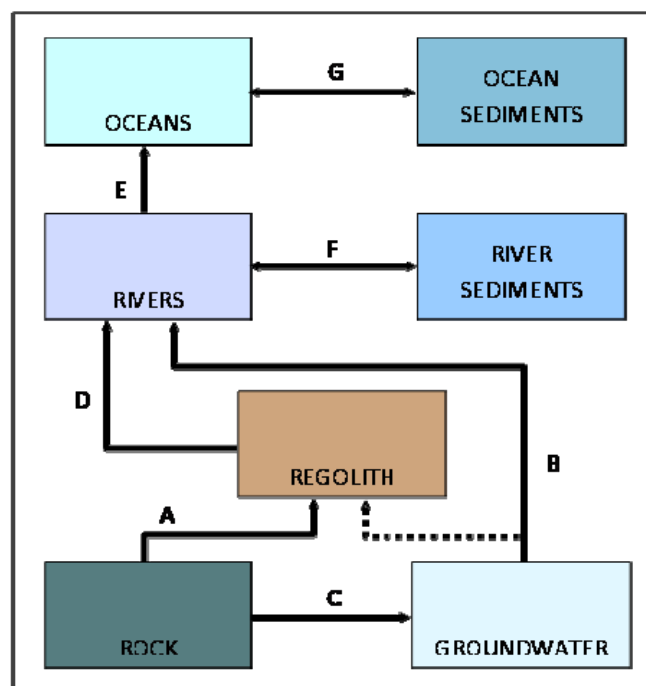


Figure 11-1. An example of a ‘compartment-pathway’ diagram. This example shows some of the fluxes (pathways) that may occur in the geosphere and the surface environment. The fluxes are: A, erosion of bedrock; B, discharge of groundwaters; C, leaching of bedrock by ground waters; D, surface transport of regolith to rivers; E, river transport to the oceans; F, transfer between river sediments and river transported material; and G, sedimentation in the oceans. Under changed climatic conditions, such as a period of glaciation, the pathways will also change as different processes affect the fluxes (after Miller et al. 1997).

The rates of geological processes are rather more difficult to determine as many cannot be directly measured but must be inferred from other data. Furthermore, such data are scarcer than the basic geochemical data, often subject to greater uncertainty and usually restricted to limited localities (IAEA 2003a).

Further, there are the questions about what scale is appropriate for a comparison – should the data for natural radionuclide fluxes be from the repository site area alone, from the surrounding region or from an even wider area of investigation? The question of scale is especially important when considering pathways involving groundwater as groundwater movement is usually very heterogeneous at smaller scales, depending strongly on the nature and frequency of water-conducting pathways in the subsurface.

Other processes too are by no means uniform and are influenced by local geological, hydrological, topological and climatic conditions. Thus the smaller the scale of the comparison, the more specific is that comparison to the repository site. Ultimately, it could be possible to compare natural and repository-derived fluxes between the same geosphere and biosphere compartments under the same conditions, if the appropriate data are available.

IAEA (2003a) does however suggest that there can be some advantages in using large-scale natural fluxes from a variety of geological and climatic conditions for comparison with repository-derived fluxes, rather than site-scale alone, as it helps to avoid misleading conclusions being drawn if the site is in some respect geochemically anomalous. The situation at Olkiluoto, discussed below, is a case in point as the deep groundwater U concentrations are particularly low and do not correlate with the U content of the host rock. This could have implications for the apparent performance of a spent nuclear fuel repository if the natural U flux in groundwater was compared with that from the spent nuclear fuel without the understanding that the Olkiluoto site has some anomalous characteristics when compared with other sites (Pitkänen et al. 2003). However, the larger the scale of the natural system used for comparison, the greater the possibility of anthropogenic influences affecting the data. For example, the use of data for river discharge rate and the dissolved and suspended load in order to determine an integrated, long-term regional flux for a whole drainage basin is vulnerable to human activities in the drainage basin. Activities such as deforestation, changes in agricultural practices, mining or industrial discharge could all affect the river water composition and suspended load. The construction of dams is an obvious example of anthropogenic influences on the discharge rate. IAEA (2003a) notes that “...*It is believed that most current river flow data are affected by human activity and may not be easy to interpret as indicators of long term natural processes.*” The same report suggests that the long-term, global (geological) flux of uranium from bedrock to ocean sediments determined by Fyfe (1979) could be considered as the simplest large-scale reference for comparison with radionuclide releases from repository systems (IAEA 2003a).

The question of data availability for application of complementary indicators using natural radionuclide fluxes has been addressed in the recent IAEA-instigated Coordinated Research Project (CRP), the objectives of which were to compile appropriate data on natural elemental concentrations and fluxes between subsurface and surface compartments on a worldwide basis. The data summarised in the report (IAEA 2005) were obtained in the countries of the nine participant organisations. The Finnish Geological Survey was one such participant and the results of the project within Finland have been published in greater detail as Finnish Geological Survey reports (Hellmuth et al. 2003; Tarvainen et al. 2003; Kaija et al. 2003; Pitkänen et al. 2003).

The CRP identified a number of naturally-occurring target substances (elements, radionuclides and specific chemical compounds) that were considered relevant to repository safety. At the outset of the project, it was envisaged that all the data collected for these target substances would be integrated into a single global database. However, due to differences in analytical techniques, statistical methodology, range of substances measured, sampling areas and sampling densities between (and within) countries, which made much of the data incompatible, this plan was shelved in favour of reporting the data as separate databases by country. In view of the conclusions reported in IAEA (2003a) that natural concentrations and environmental fluxes could best be used as complementary indicators at site and local area scale, the lack of a global database was not seen as a significant shortcoming.

In the following discussion of natural radionuclide fluxes, the results of the Finnish Geological Survey studies are the main focus, because of their specific relevance to this report. Of particular interest are the studies of geochemical fluxes at the four Finnish

investigation sites (Pitkänen et al. 2003) and at the Palmottu U deposit (Kaija et al. 2003). However, the results from other national studies reported in the CRP (IAEA 2003a) are used for comparison where appropriate, as are the results of the earlier SKI study of naturally-occurring radionuclide and elemental fluxes in the vicinity of the Äspö laboratory (Miller et al. 1997).

11.2.1 Groundwater fluxes

The natural radionuclide flux of most obvious relevance to repository radionuclide releases is that of dissolved species in groundwater. Naturally-occurring radionuclides in the geosphere can dissolve in groundwater during its transit through the bedrock from recharge to discharge locations in response to the infiltrating water composition (e.g. redox, pH, salinity). The groundwater composition, including content of these radionuclides, can be measured directly from samples taken from discharging water at the surface, from water in shallow dug wells or deeper drilled boreholes.

It is difficult to measure groundwater flux directly and it is usually estimated on the basis of borehole and well data and modelling of the hydrogeological system at various spatial scales (e.g. regional, local and site). However, for the calculation of U fluxes at the Palmottu uranium deposit, described in Kaija et al. (2003), groundwater flow was estimated simply by using reasonable values for hydraulic gradient and conductivity averaged over the whole rock volume containing the deposit and applying Darcy's Law.

For the estimation of natural U fluxes at four potential repository sites in Finland – prior to the selection of Olkiluoto as the preferred site – Pitkänen et al. (2003) used information from the site-scale hydrogeological modelling, used in the TILA-99 safety assessment (Vieno & Nordman 1999), which was supported by the site characterisation studies. The groundwater flow through a reference plane with the approximate dimensions of the repository (1 km x 1 km) at repository depth (around 500 m b.g.l.) was calculated. Since there was considerable variation in groundwater flux as well as in U content of the groundwater with depth, groundwater U fluxes were also calculated for reference planes 100 m above and 100 m below the target repository depth at each site.

Before discussing the results of this study, it is worth noting the summary of the four sites given in Pitkänen et al. (2003):

“The geology of the sites covers virtually the entire formation history of Finnish Precambrian bedrock, and although the rock types are all felsic the geochemistry differs significantly. Romuvaara represents Archean basement gneisses with low K and U contents. Oxidising conditions have characterised the formation of Proterozoic granitoids at Kivetty and Hästholmen, and particularly anorogenic rapakivi granite at Hästholmen is enriched with incompatible elements such as K, Rb, U and Th. Migmatitic mica gneisses at Olkiluoto represent a very reducing environment with graphite and sulphides. Hydrogeologically the sites differ significantly in relation to the Baltic Sea evolution since the last glaciation. The inland sites, Romuvaara and Kivetty, have been above the highest shoreline since the retreat of the ice, whereas the coastal sites have been below sea level. Hydraulic gradient is also higher inland than on shore.”

The groundwater flow rates and the estimated natural U fluxes at the reference planes for the four repository sites, at the time of repository closure (assumed to be 100 years AP), are shown in Tables 11-3 and 11-4, respectively (data from Pitkänen et al. 2003). Note that the groundwater flux in each case is the sum of the downward and the upward fluxes calculated in the hydrogeological modelling, taking the significant fracture zones into account. In all four sites, the flow direction is predominantly downwards in the sparsely fractured rock but, in the fracture zones, the flow can be upwards or downwards depending on the hydraulic connections and properties of the fracture zone. The variability in intersection of transmissive fracture zones with the reference planes is one reason for the large differences in groundwater flux between sites and between depths shown in Table 11-3. This results in larger calculated U fluxes at Kivetty, as compared to the other sites, although the low U fluxes shown for Olkiluoto are also influenced by the particularly low measured U content in the deep groundwater at that site (Pitkänen et al. 2003).

It is worth noting that the site investigations carried out since the report of Pitkänen et al. (2003) and the consequent greater understanding of site hydrogeology have resulted in much lower values for groundwater fluxes at the Olkiluoto site. Based on data given in Löfman & Karvonen (2012), the total groundwater flux after repository closure across a 1 km² plane at -475 m, located in the same position as that defined by Pitkänen et al. (2003), is less than 50 m³/a (varies depending on how properties are assigned to the hydrogeological zones and the sparsely fractured rock; see Löfman & Karvonen (2012) for details).

The same pattern noted by Pitkänen et al. (2003) is also evident in the newer Olkiluoto data of predominantly downward fluxes in the sparsely fractured rock and a mix of upwards and downwards flow in the hydrogeological zones. Olkiluoto groundwater flux data are given in Table 11-5, calculated for 5000 AD (Löfman & Karvonen 2012), about the point in time when radionuclide releases could be expected from a canister with an initial defect in a less favourably sited deposition hole (the reference case – see *Assessment of Radionuclide Release Scenarios for the Repository System* for details). Table 11-5 also shows the natural uranium and Ra-226 fluxes around repository depth, using the groundwater concentrations for these radionuclides from Pitkänen et al. (2003).

Table 11-3. Summary of the total groundwater fluxes (downwards + upwards) [m³/a] through the 1 km² reference planes located at potential repository depths (+/- 100 m) at the four Finnish investigation sites (from Pitkänen et al. 2003).

Repository site	Total groundwater flux [m ³ /a]		
Depth	~ 400 m	~ 500 m	~ 600 m
Romuvaara	2043	290	143
Kivetty	7050	8692	3754
Hästholmen	1270	792	535
Olkiluoto*	1678	730	311

* The current total groundwater flux at Olkiluoto at the depths indicated in the table is much less than that calculated by Pitkänen et al. (2003); see text for more details, and Table 11-5.

Table 11-4. Summary of the estimated U fluxes [$\text{kg}/(\text{km}^2 \cdot \text{a})$] through the reference planes located at potential repository depths (± 100 m) at the four Finnish investigation sites (from Pitkänen et al. 2003).

Repository site	Olkiluoto* (coastal site)	Hästholmen (coastal site)	Romuvaara (inland site)	Kivetty (inland site)
Upper plane	375 m	375 m	380 m	390 m
U flux	$0.35 \cdot 10^{-3}$	$3.6 \cdot 10^{-3}$	$3.0 \cdot 10^{-3}$	$32 \cdot 10^{-3}$
Middle plane	475 m	475 m	480 m	490 m
U flux	$0.028 \cdot 10^{-3}$	$1.9 \cdot 10^{-3}$	$0.32 \cdot 10^{-3}$	$30 \cdot 10^{-3}$
Lower plane	575 m	575 m	580 m	590 m
U flux	$0.0093 \cdot 10^{-3}$	$0.55 \cdot 10^{-3}$	$0.16 \cdot 10^{-3}$	$11 \cdot 10^{-3}$

In addition to deep groundwater fluxes, Pitkänen et al. (2003) also investigated groundwater's near-surface (at depths between 15 m and 50 m) discharge rates for the four investigation sites. Reference planes (1 km x 1 km) for groundwater discharge were defined for each site on the basis of the site-specific groundwater flow models such that simulated flow paths from the potential repository depth intersected the discharge reference plane.

The groundwater fluxes for the repository depth reference planes and the discharge planes for the four investigation sites are summarised in Figure 11-2. It is clear from comparison of the data in Figure 11-2 that the groundwater fluxes at the discharge planes are much larger, by one to two orders of magnitude, than at the repository reference planes. This illustrates the dominance of shallow groundwater circulation in determining the discharge water composition. This is even more apparent when the more recent calculations for Olkiluoto fluxes are included (Figure 11-2; no discharge data are shown for the recent modelling results). Pitkänen et al. (2003) note that it is reasonable to assume that the contribution of deep groundwaters to the discharged U flux is very small, based on geochemical and isotopic evidence, especially for the coastal sites (Olkiluoto and Hästholmen).

The concentrations of the natural radioactive elements in groundwater vary significantly between the sites (Figure 11-3). In groundwater at shallower depths, oxidising conditions can lead to high U solubilities (e.g. Hästholmen data in Figure 11-3). These concentrations diminish with depth (Figure 11-3) generally reflecting increasingly reducing conditions away from the surface.

Table 11-5. Groundwater fluxes for Olkiluoto [$\text{m}^3/(\text{km}^2 \cdot \text{a})$] at 5000 AD, based on recent hydrogeological modelling (from Löfman & Karvonen 2012), and corresponding U fluxes [$\text{kg}/(\text{km}^2 \cdot \text{a})$] and Ra-226 fluxes [$\text{Bq}/(\text{km}^2 \cdot \text{a})$] through the reference planes located at depths of 375 m, 475 m and 575 m (U and Ra groundwater concentrations at the three depths from Pitkänen et al. 2003 Tables 7-11, 7-12 and 7-13 in Appendix 7).²¹

Repository site	Upper plane 375 m	Middle plane 475 m	Lower plane 575 m
	Total groundwater flux (upwards, downwards) $\text{m}^3/(\text{km}^2 \cdot \text{a})$		
Semi-homogeneous properties ¹	74.2 (30.1, 44.1)	42.6 (17.3, 25.3)	21.3 (8.3, 13.0)
Heterogeneous properties ²	128.9 (46.9, 82.0)	66.3 (21.2, 45.1)	29.6 (8.0, 21.6)
	U flux [$\text{kg}/(\text{km}^2 \cdot \text{a})$] across the planes		
Concentration U	0.208 $\mu\text{g}/\text{L}$	0.038 $\mu\text{g}/\text{L}$	0.03 $\mu\text{g}/\text{L}$
Semi-homogeneous properties ¹	$0.15 \cdot 10^{-3}$	$16.2 \cdot 10^{-6}$	$6.4 \cdot 10^{-6}$
Heterogeneous properties ²	$0.27 \cdot 10^{-3}$	$25.2 \cdot 10^{-6}$	$8.90 \cdot 10^{-6}$
	Ra flux [$\text{Bq}/(\text{km}^2 \cdot \text{a})$] across the planes		
Concentration Ra	5.4 Bq/L	4.6 Bq/L	3.7 Bq/L
Semi-homogeneous properties ¹	$4.01 \cdot 10^5$	$1.96 \cdot 10^5$	$7.88 \cdot 10^4$
Heterogeneous properties ²	$6.96 \cdot 10^5$	$3.05 \cdot 10^5$	$1.10 \cdot 10^5$

Notes (for further explanation, see Löfman & Karvonen 2012):

- 1: Semi-homogeneous properties refers to a model of the hydraulic properties of the geosphere. In this model there is constant transmissivity for most of the hydrogeological zones and the sparsely fractured rock (SFR) between HZs is divided by depth into layers with different hydraulic properties;
- 2: Heterogeneous properties refers to the alternative model of hydraulic properties in which HZs have heterogeneous transmissivities and the properties of the SFR are scaled based on the Discrete Fracture Network (DFN) model.

²¹ Ra-226 concentrations are based on four samples taken at Olkiluoto from different groundwater types/depths. These concentrations are relatively high compared with other investigation sites in Finland (e.g. Romuvaara, Kivetty), where at the same depth range Ra-226 concentration is no higher than 0.025 Bq/L. Nonetheless the radium concentration is generally higher in the more saline waters which are typical in coastal areas and in Ca-rich rich groundwaters such as Olkiluoto (Pitkänen et al. 2003, see also e.g. Pöllänen 2003).

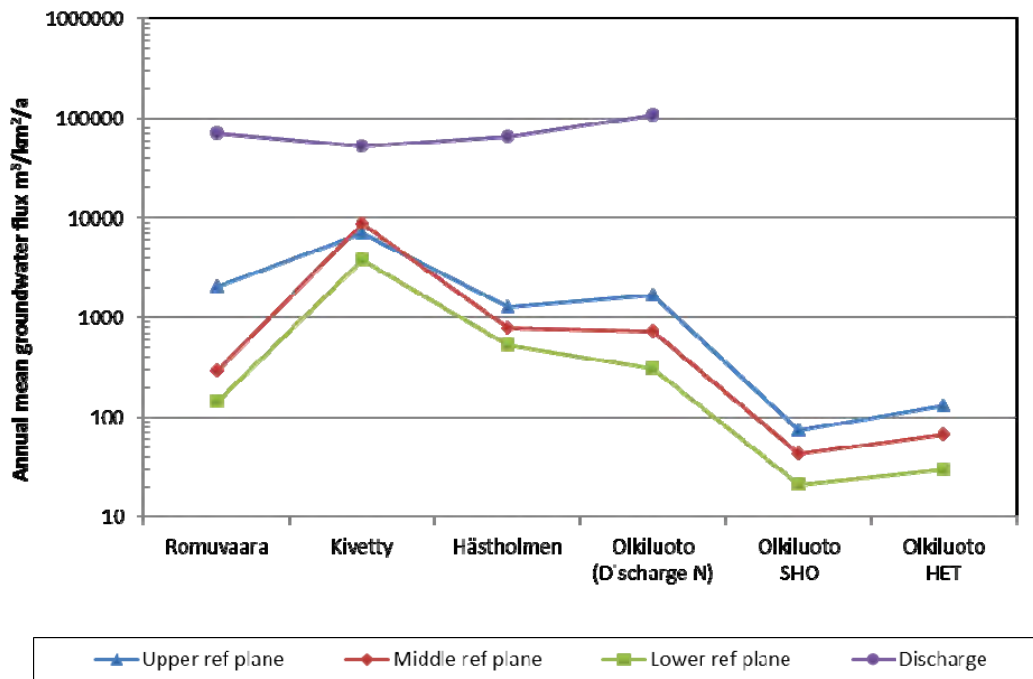


Figure 11-2. Summary of annual mean groundwater fluxes [$\text{m}^3/(\text{km}^2 \cdot \text{a})$] across the repository depth reference planes and the discharge planes at the four Finnish investigation sites at the time of repository closure from Pitkänen et al. (2003) and at Olkiluoto for 5000 AD (SHO and HET) based on current modelling in Löfman & Karvonen (2012). SHO – semi-homogeneous hydraulic properties; HET – heterogeneous hydraulic properties used in the hydrogeological model (see text and notes for Table 11-5).

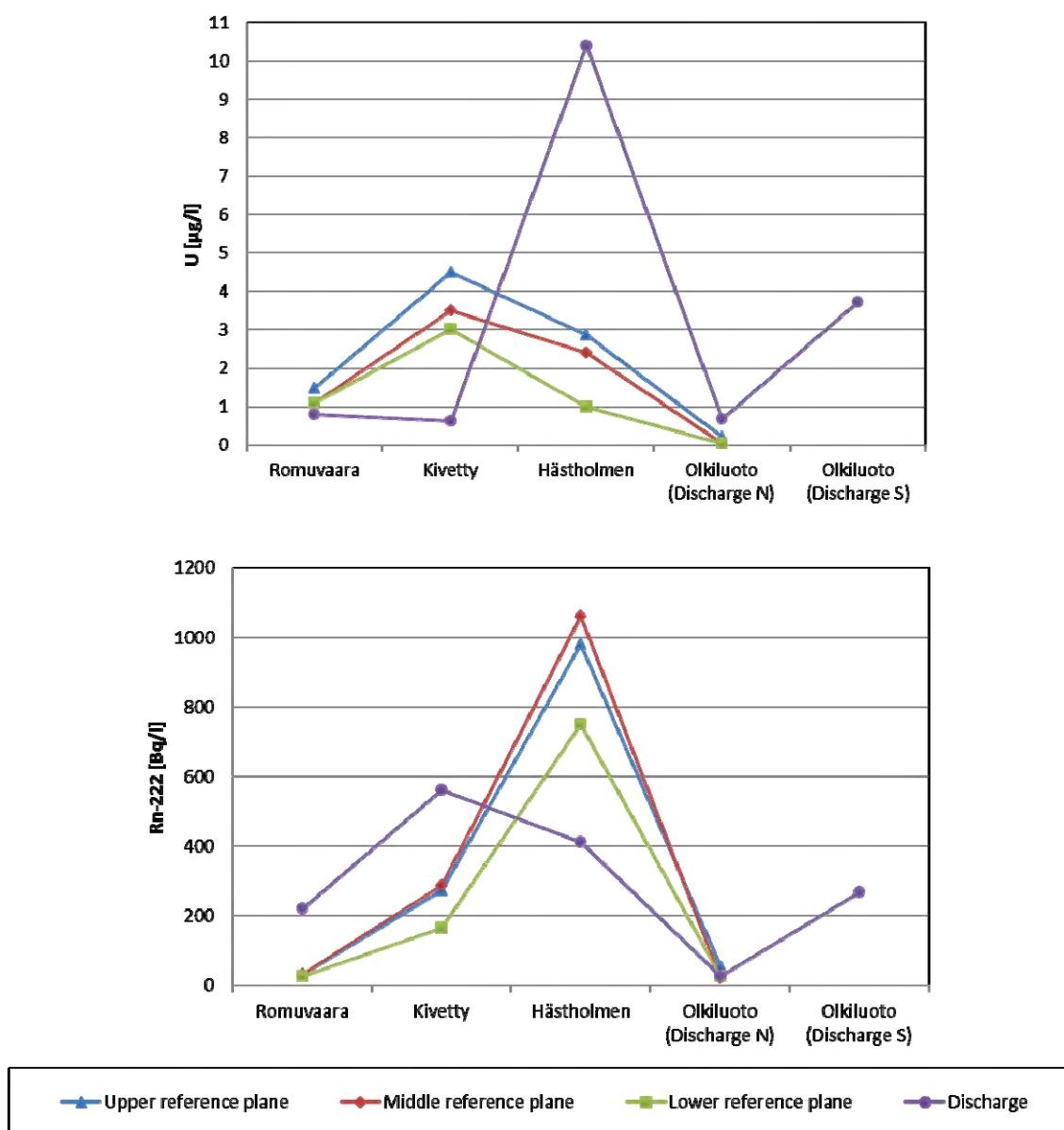


Figure 11-3. (Top) U ($\mu\text{g/L}$) and (Bottom) Rn-222 (Bq/L) concentrations in deep groundwaters (at the depths of the repository reference planes) and discharge waters at the four Finnish investigation sites (from Pitkänen et al. 2003).

Around repository depth, the U groundwater concentration correlates with the rock composition except at Olkiluoto where the U content of the groundwater is particularly low, probably as a result of specific reducing minerals at depth (Pitkänen et al. 2001). But it should be noted that the U measured in the groundwater at depth may have originated elsewhere (i.e. dissolved from rocks along the flow path during transport from the surface). The relatively low U flux at the discharge plane at Kivetty and the discharge water U concentrations, which are lower than the concentrations at depth,

may indicate short residence times and a small degree of rock/water interaction of the discharged water – low pH and alkalinity in the shallow groundwaters support this (Pitkänen et al. 2003). The same may be true also of the Olkiluoto (north) and Romuvaara U discharge concentrations. However, low U in discharge waters could also indicate the lower availability of soluble uranium-bearing minerals along flow paths that have been in existence for long periods.

Miller et al. (1997) calculated natural fluxes of several elements due to groundwater transport for coastal and inland environments based on groundwater composition and groundwater flow data from the investigation area around the Äspö laboratory. In their study, Miller et al. (1997) considered the evolution of the Äspö site through the glacial/interglacial cycles over the next 140,000 years used as the long-term climate scenario in the safety assessment Site 94 (SKI 1994). It was expected that the Äspö site would change from being a coastal site during the current temperate period to an inland site during periglacial conditions in the next glacial period, with a fall in sea level due to development of continental ice sheets. In later periods of glaciation, the site would experience a glacial environment with ice cover. These changes in environmental conditions are likely to have a large influence on the natural radionuclide fluxes at the site as different processes become important, so Miller et al.'s (1997) study is an interesting attempt to anticipate the effects. However, for comparison with the results from Pitkänen et al. (2003) only the estimated natural radionuclide fluxes in groundwater in the coastal and inland environments (as these use the same parameter values) are given in Table 11-6.

Miller et al. (1997) take a single value of $133 \text{ m}^3/(\text{km}^2 \cdot \text{a})$ to represent the groundwater flux in the regional steady state flow field for the crystalline basement rocks around Äspö. This is a significantly smaller groundwater flux than Pitkänen et al. (2003) applied for the CRP studies (Table 11-3) although it is consistent with the fluxes at Olkiluoto according to current knowledge of the site (Löfman & Karvonen 2012). However, the U flux estimated based on this value falls within the range of values for the four Finnish investigation sites, being somewhat higher than for Olkiluoto but similar to the deeper reference plane fluxes estimated for Romuvaara and Hästholmen. The order of magnitude lower flux of Th reflects its lower solubility in the Äspö groundwater compared with U, as it is present in the bedrock at approximately double the concentration of U (11.8 ppm compared with 5.4 ppm). The relative sizes of the fluxes of K, Rb and Ra reflect their abundance in the bedrock directly.

A rather different comparison can be made with the elemental groundwater fluxes calculated for the Carmenellis granite in S.W. England (Norris & Degnan in IAEA 2005, Appendix I). The 290 Ma granite has relatively high contents of U, Th and K (13.4 ppm, 9.8 ppm and 3.9 %, respectively) and an active meteoric water circulation system driven by the high heat flow. Based on an estimated annual groundwater discharge rate of $1.06 \cdot 10^7 \text{ m}^3$ over the 80 km^2 outcrop area (equivalent to $1.33 \cdot 10^5 \text{ m}^3/(\text{km}^2 \cdot \text{a})$), Norris & Degnan (IAEA 2005, Appendix I) calculated the annual elemental fluxes as shown in Table 11-7.

The groundwater flux used for the estimates is much closer to the discharge plane fluxes given in Pitkänen et al. (2003), particularly the Olkiluoto (north) discharge area

(Figure 11-2). However the U concentrations in the discharge waters are very low (Table 11-7) – an order of magnitude lower than those in the Olkiluoto (north) discharge waters and similar to the Olkiluoto deep groundwaters suggesting that there is a significant deep groundwater component in the Carmenellis discharge waters, unlike the Finnish sites where discharge is dominated by shallow groundwaters.

Also shown in Table 11-7 the corresponding activity fluxes show the strong dominance of Rn-222 over the U, K and Rb activities. Norris & Degnan (IAEA 2005, Appendix I) suggest that this is because the U has very low solubility in the reducing groundwaters despite its abundance in the granite. This in turn leads to low groundwater U activities and fluxes even though the groundwater flux itself is large. Rn, however, is easily leached from the rock by the active groundwater/rock interaction and mineral alteration and thus is not in equilibrium with the dissolved U (IAEA 2005).

Table 11-6. Natural fluxes [$\text{kg}/(\text{km}^2 \cdot \text{a})$] due to groundwater transport in the present-day coastal and possible future inland environments assuming a representative deep groundwater flow rate for the crystalline rock of $133 \text{ m}^3/(\text{km}^2 \cdot \text{a})$ (data from Miller et al. 1997).

Element	Flux [$\text{kg}/(\text{km}^2 \cdot \text{a})$]
U	$0.17 \cdot 10^{-3}$
Th	$0.02 \cdot 10^{-3}$
Ra	$2.7 \cdot 10^{-10}$
K	1.3
Rb	$3.9 \cdot 10^{-3}$

Table 11-7. Groundwater concentrations, natural fluxes and activities of U, K, Rb and Rn-222 due to groundwater circulation and discharge from the Carmenellis granite assuming a groundwater flow rate of $1.33 \cdot 10^5 \text{ m}^3/(\text{km}^2 \cdot \text{a})$ (data from IAEA 2005, Appendix I).

Element	Concentration [mg/dm^3]	Flux [$\text{kg}/(\text{km}^2 \cdot \text{a})$]	Activity [$\text{Bq}/(\text{km}^2 \cdot \text{a})$]
U	$8 \cdot 10^{-5}$	0.01	$1.25 \cdot 10^5$ *
K	2.8	2500	$6.1 \cdot 10^7$
Rb	< 0.02	34.9	$3.1 \cdot 10^6$
Rn-222	$24.9 \text{ Bq}/\text{dm}^3$		$3.25 \cdot 10^9$

* The U decay chain activity is $1.2 \cdot 10^6 \text{ Bq}/(\text{km}^2 \cdot \text{a})$

11.2.2 Fluxes due to erosion

Erosion can be divided into two types – mechanical erosion and chemical erosion. With mechanical erosion, the rocks are broken down, perhaps very finely, without necessarily being subject to chemical alteration or dissolution. Chemical weathering on the other hand is the decomposition of the rock by water, enhanced by microbial action in some environments (Berthelin 1988).

Mechanical erosion gives rise to fluxes of material in which the chemical components are not necessarily available for uptake by plants or animals. This has consequences for exposure pathways when fluxes of naturally-occurring radionuclides are considered: fluxes of solid particles due to mechanical erosion are unlikely to result in exposure due to ingestion or inhalation to the same extent as fluxes from chemical weathering.

The two processes of erosion are often coupled but climatic conditions can favour the dominance of one over the other: low temperatures and low precipitation favour mechanical weathering whereas high temperatures and high rainfall favour chemical weathering (Skinner 1987). At the Olkiluoto site, under present climatic conditions, chemical and mechanical erosion are likely to be coupled. The annual mean temperature of around 6.0 °C and rainfall of 540 mm (Table 3 in Haapanen 2010) indicates moderate chemical weathering with frost action according to the classification of Skinner (1987).

Miller et al. (1997) used estimates of weathering rates, both mechanical and chemical, for post-glacial conditions in southern Sweden as a basis for estimating fluxes of radionuclides and heavy metals around the Äspö site. A variety of techniques and sources were used to obtain the total rock erosion rate and chemical erosion rate, and thus mechanical erosion rate by difference. The results indicate a total erosion rate²² of $3.75 \cdot 10^3 \text{ kg}/(\text{km}^2 \cdot \text{a})$, chemical erosion rate of $2.4 \cdot 10^3 \text{ kg}/(\text{km}^2 \cdot \text{a})$ and mechanical erosion rate of $1.35 \cdot 10^3 \text{ kg}/(\text{km}^2 \cdot \text{a})$. As the climatic conditions, geology and topography (as extreme topography tends to increase the contribution of mechanical weathering) in southern Sweden are reasonably similar to SW Finland, this would also suggest that chemical erosion will be somewhat greater than mechanical erosion under present climatic conditions. However, it is not possible to use this chemical erosion rate directly to estimate natural radionuclide or heavy metal fluxes by multiplying by the content of the specific element in the rock because not all constituent minerals in a rock weather at the same rate. In particular, feldspars may weather relatively quickly, releasing K and Rb, whereas U and Th may be mainly associated with more resistant phases such as zircon and monazite, respectively. Thus, elemental fluxes will depend not only on the composition of the weathering rock but on the susceptibility of the host minerals to weathering. This means that additional sources of information must be used.

²² This was based on post-glacial total erosion rates of $1.05\text{--}1.5 \cdot 10^{-6} \text{ m/a}$ assuming a rock density of $2.5 \cdot 10^3 \text{ kg/m}^3$ (Miller et al. 1997). Since total erosion is considered to remove a complete ‘layer’ of the surface, total erosion fluxes for individual elements in the rock can be calculated simply by multiplying the erosion rate by the concentration in the rock. This was the method used in the earlier discussion of natural radionuclide fluxes from erosion for the KBS-3H Safety Studies (Neill et al. 2007).

Assessment of chemical weathering rates in Finland was part of the IAEA Coordinated Research Project (reported in Tarvainen et al. 2003). This study used a combination of data from different drainage systems (free-flowing rivers and river/lake systems) and the extensive database of the geochemical compositions of till to estimate fluxes from chemical weathering for a number of elements, including radionuclides and heavy metals.

Tills are mainly composed of crushed fresh bedrock – the mechanical erosion product of glaciation. In Finland, tills cover about 97 % of the land surface and have a median thickness of 6.7 m although most are around 3–4 m thick (Hellmuth *in* IAEA 2005, Appendix F). The till, especially the lowest layers, is closely representative of the composition of the underlying bedrock as it tends to be unsorted and locally derived. It is young, 10,000–60,000 a, originating from the last ice age so that chemical weathering has had only limited effect, mainly on the surface layer. The bedrock beneath the tills is unweathered as glaciation removed the weathered zone before the tills were deposited. Under conditions since the last ice age, acidic podzolic soils have tended to develop at the surface of the tills beneath a thin humus layer. Poor drainage, low relief and humid conditions have also encouraged the development of peat over a large proportion of the surface (Tarvainen et al. 2003). The main direction of transport is downwards through the soil and infiltration of rainwater leaches elements from the upper soils layers although these elements may be immobilised and remobilised within the soil profile. As noted in the discussion of groundwater fluxes, only a small proportion of the surface water penetrates to greater depths and the shallow groundwaters contain only small proportions of deep groundwater discharges. These factors make it possible to use the chemical processes occurring within the till to derive geochemical fluxes of elements from material which is representative of the underlying unweathered bedrock. Tarvainen et al. (2003) selected a small number of hydrogeologically well-defined catchments where chemical erosion fluxes could be calculated from the stream run-off over the defined areas. Results from this method could then be compared with fluxes calculated from the observations from weathering profiles in tills. Results for the example of U are shown in Table 11-8.

In Table 11-8, the ‘erosion rate from median discharge and median concentration’ is based on measured stream discharge rate and stream chemical analysis data. This in turn is used to normalise the annual ‘median groundwater flux’ for chemical erosion of U over the catchment area. The ‘estimated till weathering flux’ for U is based on analyses of vertical profiles of till²³ but these profile samples were part of a regional mapping

²³ The “estimated till weathering flux” and also the “local till weathering flux” are derived from analyses of vertical soil (till) profiles which measure the depletion of uranium compared with the bedrock value due to chemical weathering over the period since the tills were deposited, which is quite well defined in these areas. In the study (Tarvainen et al. 2003), it was assumed that there was no significant contribution to the groundwater U concentrations from deep groundwater circulation in bedrock (this was investigated and no significant contribution could be identified in the catchments studied), thus there should be a reasonable correlation between the U flux calculated from the spring water concentrations and the till leaching profiles.

Table 11-8. Groundwater chemical erosion rates of U in five selected spring catchments and estimated U chemical erosion rates from till profiles (data from Tarvainen et al. 2003).

Spring	Catchment area [km ²]	Erosion rate from median discharge & median concentration [kg/a]	Median groundwater flux [kg/(km ² ·a)]	Estimated till weathering flux * [kg/(km ² ·a)]	Local till weathering flux** [kg/(km ² ·a)]
Onkalolähde	0.32	0.001	0.0031	0.28	0.0031
Kiilan lähde	0.17	0.074	0.44	0.40	-
Kaikalan lähde	0.13	0.003	0.023	0.28	-
Oinaslähde	0.16	0.017	0.11	0.16	-
Isolähde	0.32	0.003	0.0094	0.15	0.022

* Estimate based on till analysis data from regional mapping – till did not come from the spring catchment but from locations up to 10s of kms away.

** Estimates based on analysis of new till samples collected from the specific catchments.

campaign and may have been taken some tens of kilometres from the study catchments, so they represent only a similar bedrock geology. The ‘local till weathering flux’ is based on till profile samples collected for the CRP from the specific catchments. It is noticeable that the values from the specific till samples correspond much more closely with those calculated from the corresponding catchment stream data than do the regional till sample values.

Values for the chemical erosion fluxes of Th, K and Rb from data in Tarvainen et al. (2003) are summarised in Table 11-9. The measured concentrations of Th in the groundwater were very low (below the detection limit) so the groundwater fluxes shown are hypothetical maximum values. Even so they are significantly lower than the fluxes estimated from the till samples, including the new samples collected within the catchment areas. This suggests that an additional process is removing Th from the groundwaters since the amounts carried in the groundwater do not account for the losses in the till samples. For K which is a major element in the rock-forming minerals of the bedrocks, there is better correspondence between the two fluxes – at least as the order of magnitude.

For Rb, which is expected to follow K in its behaviour although it is present in the bedrocks at trace element levels, the fluxes estimated from the new till samples are also much closer to the groundwater fluxes.

Tarvainen et al. (2003) also report a study of heavy metal chemical weathering in forest soils based on reanalysis of a large number of till samples from southern Finland. The results for U chemical weathering rates are shown in Figure 11-4. It is difficult to compare the results from this study with those discussed above, as the weathering rate was estimated by scaling heavy metal weathering rates to those of base cations (Ca, Mg) which themselves are based on modelling of the till analytical data (see Tarvainen et al.

2003 for a detailed explanation). However, it seems that the chemical fluxes calculated from the median groundwater fluxes (Table 11-9) are of a similar range of magnitude.

Table 11-10 summarises dissolved natural U fluxes either measured or estimated by different methods over different scales from site-specific to regional and in oxidising and reducing conditions.

The examples discussed have been given to illustrate some of the concerns and uncertainties in estimating natural fluxes of radionuclides. In particular, understanding and evaluating the measured concentrations and estimated fluxes in the context of knowledge of the whole hydrogeological system is a key element. If such parameters are to be used as adjuncts to a repository safety case, it is essential that their limitations are known.

Table 11-9. Groundwater chemical erosion fluxes of Th, K and Rb in five selected spring catchments and estimated U chemical erosion rates from till profiles (data recalculated from Tarvainen et al. 2003).

Spring	Median groundwater flux [kg/(km ² ·a)]			Till weathering flux* (new samples**) [kg/(km ² ·a)]		
	Th	K	Rb	Th	K	Rb
Onkalolähde	< 0.0025	95.9	0.19	0.80 (0.019)	217	6.08 (0.30)
Kiilan lähde	< 0.0047	981	0.047	1.54	552	10.7
Käikälän lähde	< 0.0062	568	0.38	0.93	259	5.22
Oinaslähde	< 0.0050	195	0.25	0.73	185	3.76
Isolähde	< 0.0094	260	0.36	0.31 (0.047)	142	3.40 (0.53)

* Estimate based on till analysis data from regional mapping – till did not come from the spring catchment but from locations up to 10s of kms away.

** Estimates based on analysis of new till samples collected from the specific catchments.

Table 11-10. Dissolved natural U fluxes either measured or estimated by different methods over different scales from site-specific to regional and in oxidising and reducing conditions (IAEA 2005, Appendix F).

	U flux [kg/(km ² ·a)]	
	Oxidising zone	Reducing zone
Small catchments, measured	0.003–0.043	
Headwater streams, measured	0.017	
Chemical erosion (till), estimated	0.054	
Palmottu U-mineralisation, estimated	3.2	0.0003
Repository investigation sites, estimated	0.033–0.99	1·10 ⁻⁵ –0.032

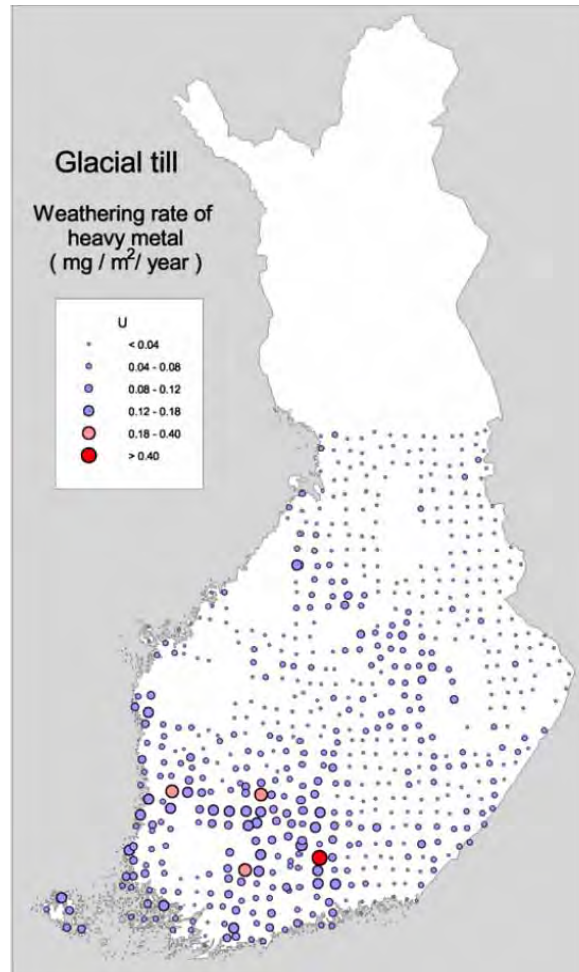


Figure 11-4. Estimated U weathering rates for till soils (from Tarvainen et al. 2003). Note the units of $\text{mg}/\text{m}^2/\text{year}$ are equivalent to $\text{kg}/(\text{km}^2 \cdot \text{a})$ used in the text.

11.2.3 Environmental radioactivity at Olkiluoto

The construction works for the Olkiluoto nuclear power plant started in 1974 and an extensive environmental monitoring programme was begun shortly thereafter. The first surveillance programme for environmental radioactivity was started in April 1977. The purpose was, and is, to clarify the possible exposure to radiation in the environment due to the authorised and potential accidental releases from the nuclear power plant. In the context of disposal of spent nuclear fuel, these data form part of the baseline database and help to build up a firm basis for the environmental surveillance programme of the disposal facility and associated operations. Results from the monitoring programme may be used to make comparisons of the assessment results with existing concentrations of radionuclides in the environment. In this section, selected data regarding environmental radioactivity are presented; all the data are presented in detail in Roivainen (2005).

The main limitation with the results from surveillance programme for environmental radioactivity is that the set of radionuclides detected, such as K-40, Cs-137, Sr-90, are very different from the ones expected from a hypothetical release from the repository.

Thus, a straightforward comparison between present-day environmental concentrations with calculated ones in the safety assessment is generally not possible.

However, since 2003, C-14 has been analysed from 2–3 biological samples once a year. In 2003, hair moss, pine needle and birch leaf samples were analysed, with resulting activity concentrations of 89, 104 and 94 Bq/kg_{dw}, respectively (Roivainen 2005).

Another result that can be used for comparison purposes from the surveillance programme for environmental radioactivity is the concentrations of the main radionuclides in drinking water. Even though the sets of radionuclides differ, these results can be used to derive the effective dose from ingestion of drinking water to compare with the corresponding effective dose as calculated in the safety assessment. The radionuclide activities in the untreated water of Olkiluoto local waterworks (Korvensuo Reservoir, KO) in 2002–2003 are presented in Table 11-11.

11.3 Radionuclides relating to industrial activity

Not all non-nuclear, radionuclide fluxes and exposure are attributable to nature, of course, and anthropogenic activities over the millennia have been responsible for large, if often inadvertent, fluxes of radionuclides and some consequent harm to human populations and the environment. As Baxter (1996) notes “*Man increased his radiation exposure above natural baseline when he began cave-dwelling and mining and working metals and minerals*”.

In this section some of the most radiologically important non-nuclear industrial activities are discussed in terms of their radionuclide fluxes in order to give some perspective on those fluxes that might arise from a spent nuclear fuel repository. The subsequent exposure to the public from these activities is also briefly addressed. The intention is not to make any judgement on the acceptability of releases from nuclear or non-nuclear activities involving radioactive materials, but simply to illustrate through these data and comparisons that all such activities can have effects on humans. In looking at the information given here, it is important to keep in mind that these industrial fluxes are often estimates of uncertain rigour and accuracy, but that they are actual whereas repository releases are calculated and will occur only in the far distant future.

Table 11-11. Ranges of measured activity concentrations (Bq/m³) in the untreated water of Olkiluoto local waterworks (Korvensuo Reservoir, KO) in 2002–2003; data from Roivainen (2005).

Bq/m ³	H-3	Sr-90	K-40	Cs-137
	<4000	7.2–10.4	83–140	4.7–7.8

11.3.1 NORM and TENORM

NORM and TENORM are acronyms for the terms ‘naturally-occurring radioactive material’ and ‘technologically-enhanced naturally-occurring radioactive material’, respectively. The term NORM was originally coined sometime in the 1980s to describe the radium-rich scales (i.e. mineral precipitates) which had been known by the oil industry since the early 1900s to form inside oilfield piping and pumps (Wymer 2008). However, as a term it is not very useful as it is the radionuclides that are naturally-occurring and not the scale, which precipitates from oilfield formation waters during oil production. Furthermore, virtually all materials contain some naturally-occurring radionuclides, as discussed in the previous section. So the term TENORM was coined to acknowledge this distinction between unaltered natural materials, for example uranium ore, and the materials that resulted from industrial processes such as tailings at uranium mines (i.e. waste after processing to remove the main uranium minerals) or the radium-rich oilfield scales. However, this separate category of TENORM is not entirely useful as it implies that the radionuclides are concentrated compared with the natural material and are thus more hazardous, which is not necessarily always the case.

According to Wymer (2008):

“The approach adopted by the IAEA for the purposes of international radiation protection standards is to use only the term NORM, regardless of the origin of the material, while defining ‘radioactive material’ more narrowly as ‘material that is designated in national law or by a regulatory body as being subject to regulatory control because of its radioactivity’. NORM is then a particular form of radioactive material (defined in the narrower sense) where the radioactivity in question is associated with radionuclides of natural origin.”

Thus in its widest sense, NORM is material that has been moved from its original location and form by human activities such as mining and processing. It is worth noting that uranium ore falls within the definition of NORM but is a special case because its mining and processing operations are part of the nuclear fuel cycle and are now subject to stringent regulatory control²⁴. This has led to a situation where the ‘nuclear style’ standards applied to these operations are in some respects inconsistent with the standards now being established for other NORM-related activities (Wymer 2008). With respect to uranium mining wastes, however, in the “Radioactive Waste in Perspective” Report (NEA 2010), the authors state that:

“There are exceptions to most radioactive waste classification schemes for the following materials:

- mining and milling wastes: residues left from mining and extraction of uranium and other raw materials that contain naturally occurring radionuclides;

²⁴ Up to 2003, total world uranium production was ~2 Mt but this has given rise to around 2 billion tonnes of tailings. Current tailings are well managed but many sites of historic mining operations are abandoned and few have been remediated (UNSCEAR 2008).

- environmental contamination: radioactively contaminated environmental media, such as soil and groundwater...”.

Further discussion of these regulatory aspects of NORM is outside the scope of this report except to note that regulation of management and control of industrial NORM is an on-going international problem as documented in IAEA (2006) and IAEA (2008).

It is not widely appreciated that many industrial activities unconnected to the nuclear industry involve materials which contain sufficient quantities of naturally-occurring radionuclides that the by-products potentially create hazards which are not present with the original materials. Some of implications of these industries in terms of fluxes of natural radionuclides and exposure to the public are discussed below. It is difficult to derive fluxes in the same way as discussed for natural processes. This is because the range of materials, processes and activities is so wide, and variable between different parts of the same industries around the world.

11.3.2 Coal power production

Worldwide coal consumption in 2010 was around 7.3 billion tonnes (BP 2011) and coal burning for power production accounts for the major part²⁵ creating a significant source of NORM in the resulting ash. Coal contains very variable amounts of natural radionuclides. IAEA (2003d) gives a breakdown by country (although there are no data for China, one of the world’s largest producers) of the content of major radionuclides in hard coal and lignite. The data for hard coal are summarised in Table 11-12. Lignite and brown coal tend to have lower radionuclide contents. In both hard coal and brown coal/lignite, volatilisation and sorption during combustion of the pulverised coal result in fly-ash²⁶ which is considerably enriched in the radionuclides. Baxter (1993) gives examples of average radionuclide contents for coal and fly-ash (Table 11-13). Gabbard (1993) calculated the annual worldwide releases of U and Th from coal combustion in electricity generation to the ash waste products. However, since 1993, the rapid expansion of coal consumption for power generation means that the current rates are now almost twice as great, at about 16,500 tonnes of thorium and 6600 tonnes of uranium released worldwide per year. Gabbard also noted that the content of U released (which includes over 44 tonnes of U-235) had an energy potential one and a half times greater than that of the coal consumed.

²⁵ The proportion of coal used in electricity generation varies strongly by country – in China, the largest coal producer and consumer, only about 30 % of coal is used for electricity generation, in the U.S. the proportion is about 95 % and in Europe 80 %. Worldwide the average is ~70 % (based on BP 2011).

²⁶ Fly-ash is fine glassy material composed of the mineral matter in coal which is entrained in the smoke during combustion and collected in the smoke stacks by filters. Bottom ash is the coarse mineral material remaining in the furnace. Fly-ash makes up around 75 % of the total ash and is particularly enriched in more volatile metals (e.g. Pb, Po – see Table 11-18) compared with bottom ash. Boiler slag is the melted portion of bottom ash.

Table 11-12. Example ranges of radionuclide activity concentrations in hard coal (Bq/kg) (data summarised from IAEA 2003b).

Bq/kg	K-40	U-238	Ra-226	Pb-210	Po-210	Th-232	Ra-228
	10–785	6–480	8–200	12–500	16–580	4–170	8–64

Table 11-13. Typical natural radionuclide activity concentrations in coal and fly-ash (Bq/kg) (Baxter 1993).

Bq/kg	K-40	U-238	Ra-226	Pb-210	Po-210	Th-232	Ra-228
Coal	50	20	20	20	20	20	20
Fly-ash	265	200	240	930	1700	70	130

This consumption of coal represents a large scale flux of radionuclides from an initial state – in coal, underground – into a state in which the radionuclides potentially impact on the biosphere much more directly. About 2 to 5 % of fly-ash produced is released directly to the atmosphere, depending on the efficiency of the stack filters (UNSCEAR 1993; EPA 2011). This material will mainly be deposited in an area down wind of the power plant (the ‘stack shadow’) according to prevailing weather conditions. UNSCEAR (2000) estimated the releases of radionuclides to the atmosphere for a typical 600 MWe coal-fired power plant (Table 11-14).

The main exposures to the general public from commercial coal burning²⁷ relate to the use of fly-ash in industry (UNSCEAR 1993, Baxter 1996). Figures for the United States give an idea of the scale of the use of fly-ash and other coal combustion products (2009 figures from ACAA 2009):

- Fly-ash: approximately 23 Mt per year is used out of a total fly-ash production of 56 Mt of which around 50 % is used in concrete and in concrete blocks. It is typically substituted for cement in concrete at a rate of about 10–30 %. About 20 % is used as structural fill and in embankments, and a further 15 % in waste stabilisation and solidification processes;
- Bottom ash: 6.4 Mt is used out of 15 Mt mainly for structural fill and embankments, as road base and aggregate, as well as additive for cement. A small quantity is used for snow and ice control (<5 %);
- Boiler slag: 1.6 Mt is used out of 2 Mt predominantly as blasting grits and roof granules.

²⁷ There are few data for coal burning for domestic heating and cooking but UNSCEAR (1993) suggest that worldwide atmospheric releases from domestic coal burning could be of the order of 0.7 TBq of K-40, 0.3 TBq U-238 series and 0.3 TBq Th-232 series (excluding Rn) if the radionuclide concentrations in smoke are the same as in the coal, but up to twenty times these if the smoke has the same concentrations as fly-ash.

Table 11-14. Releases [in GBq/a] of natural radionuclides to the atmosphere from a 600 MWe coal-fired power station, assuming a coal supply of 1.35 Mt per annum (UNSCEAR 2000).

K-40	U-238	Th-228	Ra-226	Rn-222	Pb-210	Po-210
0.27	0.16	0.08	0.11	34	0.4	0.8

EPA (2011) notes “Coal ash is also used to level out uneven terrain or applied as a stable fill for building construction. Typical applications include sites where shopping malls, housing developments, and industrial parks are planned for construction...”.

Production and use of coal combustion products are similar in Europe – about 35 Mt were used from the total production in 2006 of 50 Mt (ECOPA 2006).

The significance of the use of fly-ash in terms of exposure of a general population is assessed in UNSCEAR (2008): a study carried out in the UK (Watson et al. 2005) estimated doses arising to a critical group from use of building materials containing fly-ash as being around 600 $\mu\text{Sv/a}$ from radon inhalation and a further 900 $\mu\text{Sv/a}$ from external radiation. However, Watson et al. (2005) note that as these doses are not necessarily higher than those from other building materials, they are not strictly critical group doses. No other exposures from use of typical coal ash are considered of significance according to UNSCEAR (2008).

From the figures for use of coal combustion products, it is clear that a significant proportion is not used and must be disposed of, usually in landfill sites from where radon can be released to the atmosphere as well as groundwater potentially leaching radionuclides from the ash. This has been a source of popular concern in several countries. However, in most cases where groundwater contamination due to leakage from coal ash landfills is of concern, the major hazards are associated with arsenic and heavy metals such as lead and cadmium rather than the radioactivity (see for example, EIP 2010). However, the landfills and other disposal sites themselves can create radiation anomalies: a remote sensing survey using airborne gamma spectrometers carried out over a lake near the Danube river in Bulgaria detected two anomalous areas which, on inspection, turned out to be the settlement ponds into which the ash from a 200 MW coal-fired power station was deposited and allowed to settle (Baxter 1993). The water from the ponds drained into the nearby Danube. The local soils were also enriched in K-40, Ra-226, Th-232, Po-210 and Pb-210, and the gamma dose rate at the site was about 70 % higher than surrounding areas, requiring action by the local authorities to prevent radiological consequences to the surrounding population.

A final NORM example with respect to coal which is of particular interest for comparison with radionuclide releases from a deep geological repository relates to mining activities rather than coal consumption. During coal mining operations, large quantities of water are pumped out of the workings. The composition of these waters, where they represent old formation waters, can be significantly enriched in natural radionuclides. The coal mines in southern Poland, in the Upper Silesian Coal Basin, are a good example (Wysocka et al. 2002). The total drainage volume from the 50 mines in

the area is around $2.9 \cdot 10^6 \text{ m}^3/\text{a}$. In the southern and central parts of the basin, the coal measures are overlain by a thick layer of impermeable clay and, in these areas, highly saline, Ra-rich waters with up to 390 kBq/m^3 are found. Activity inflows into individual mines are as high as 78 MBq per day of Ra-226 and 145 MBq per day of Ra-228. The radium-rich waters are pumped into surface settlement ponds in which radium co-precipitates with barium as scales or sorbs onto bottom sediments. These solid materials have extremely enhanced radium concentrations, up to 400 kBq/kg – similar to 3 % uranium ore (Michalik et al. 2002) – thus careful disposal is required when these sites are remediated. Water from the settlement ponds is released into local rivers where the contamination can be detected at long distances from the mining. In some small rivers, the Ra-226 activity levels are such that the river waters actually exceed the Polish limits for liquid radioactive waste (at 0.7 kBq/m^3).

11.3.3 Oil and gas production

Oil-field formation waters, produced during the recovery of oil and gas, are a source of naturally-occurring radionuclides. The formation waters are often highly saline and contain significant radionuclide concentrations. Radium is usually enriched relative to U-238 and Th-232 reflecting the low solubilities of the primordial elements under reducing conditions in the reservoir compared with their progeny Ra-226, Rn-222, Ra-228 and Ra-224, which tend to partition into the formation waters (IAEA 2003c).

Minerals can precipitate from hot saline formation waters as they cool when pumped from the reservoir. Radium is most often concentrated as a co-precipitate with Ba in barite (BaSO_4) although it is also present in carbonates and complex sulphates (IAEA 2003e). The precipitates form scales in the well tubular and in pipelines. The use of seawater to enhance oil recovery can worsen the problem by providing an additional source of sulphate. The scale can, if left in situ, block pipe work so it must be removed from pipes, wellhead, valves, pumps and other equipment such as separators on a regular basis. In addition, minerals can precipitate from the brines in water treatment settling tanks forming a sludge composed of silicates with carbonate and sulphate minerals (IAEA 2003e). Table 11-15 shows ranges of radionuclide activity in oil, gas and by-product NORMs from Jonkers et al. (1997).

Table 11-15. Activities of radionuclides in oil, gas and by-products (Jonkers et al. 1997).

Radio-nuclide	Crude oil Bq/kg	Natural gas Bq/m ³	Produced water Bq/L	Hard scale Bq/kg	Sludge Bq/kg
U-238	0.0001–10	-	0.0003–0.1	1–500	5–10
Ra-226	0.1–40	-	0.002–1200	100–15·10 ⁶	50–0.8·10 ⁶
Po-210	0–10	0.002–0.08	-	20–1500	4–0.16·10 ⁶
Pb-210	-	0.005–0.02	0.05–190	20–75,000	100–1.3·10 ⁶
Rn-222	-	5–0.2·10 ⁶	-	-	-
Th-232	0.03–2	-	0.0003–0.001	1–2	2–10
Ra-228	-	-	0.3–180	50–2.8·10 ⁶	500–50,000
Ra-224	-	-	0.5–40	-	-

A further, small volume, NORM results when radon gas, entrained with oil and water and separated along with the gas fraction, decays to form very thin films containing Pb-210 and Po-210 inside gas pipe work and in liquefied gas plants (Gesell 1975).

It is difficult to estimate fluxes for these NORMs since few global statistics have been published for the volumes of produced waters, scales or sludges. Khatib & Verbeek (2003) give an estimate for 1999 worldwide annual produced water volume of 77 billion barrels ($\sim 12 \cdot 10^9 \text{ m}^3$). In that year, global oil production was 26 billion barrels of oil and 2333 billion m^3 of gas. Clark & Veil (2009) question the basis for this figure and derive a figure of 21 billion barrels of produced water for 2007 from the U.S. onshore and offshore oil and gas production of 1.75 billion barrels of oil and 540 billion m^3 of gas. The U.S. EPA 'Oil and gas production wastes' information sheet²⁸ states that formation waters are produced at a rate of around 10 barrels of water per barrel of oil which is pretty much in agreement with Clark & Veil (2009). However, that 10 to 1 ratio is very dependent on the age and state of the reservoir being produced. The U.S. oil fields tend to be in later stages of production thus produce relatively more water than younger fields. Khatib & Verbeek (2003) use a lower ratio of three barrels of water per barrel of oil as a worldwide average.

If the Khatib & Verbeek (2003) figure is accepted and the 2010 worldwide oil production of around 30 billion barrels and gas production of 3200 billion m^3 used to scale up the 1999 water volumes, the result is around 90 billion barrels ($\sim 14.3 \cdot 10^9 \text{ m}^3$) of produced water in 2010. At the upper limit of the range of U and Th in the produced water (8 ppb U and 0.24 ppb Th), this volume of water represents an annual flux of, at maximum, 1400 GBq U-238 and 14 GBq Th-232, or up to about 114 tonnes of U and 3.5 tonnes of Th over the entire worldwide industry.

Even if the fluxes of radionuclides in the solid by-products were several orders of magnitude greater than this (e.g. 1 Mt of hard scale with 0.5 Bq/g U gives a total U flux of around 40 tonnes per annum), it is clear that these fluxes are fairly minor in comparison to those associated with coal combustion. However, it should be noted that the fate of oil industry NORM is currently relatively unregulated since activity levels and radionuclide concentrations tend to fall below those at which the NORM would be covered by radioactive waste regulation.

Produced water from onshore fields is often injected back into the formation but offshore it is more likely to be discharged into the sea. Likewise, scale from decontamination of equipment off-shore has been discharged directly to the sea. Heling & van der Steen (1994) estimated the total Ra-226 activity discharged into the Dutch Continental Shelf area from oil and gas production in that area as roughly 100 GBq/a.

Environmental regulation has tended to be concerned with hydrocarbon contamination of the water, and heavy metals, in particular mercury, lead and zinc, in scale solids and sludges rather than radioactivity (e.g. IAEA 2003e; Clark & Veil 2009). In a recent case

²⁸ <http://www.epa.gov/radiation/tenorm/oilandgas.html>, accessed 6/07/2011.

in Scotland, however, discharge of scale slurry via a pipeline at Aberdeen harbour has been stopped and the producer must store the material (ca. 40 tonnes per year) for disposal, possibly as very low-level radioactive waste (SEPA v. Scotoil 2008). In the United States, scale produced by onshore operations has caused concern for land contamination (UNSCEAR 2008) and the health implications of disposing of the solid NORM in non-hazardous waste landfills has been investigated (Smith et al. 2003).

11.3.4 Phosphate industry

Many mining and mineral processing activities give rise to NORM. IAEA (2003d) lists mineral resources from aluminium to zirconium, giving associated sources of radionuclides and ranges of activity. In most cases the radionuclides initially in the ore or in associated gangue minerals end up in the tailings, slags and other waste products of processing. From this large range of activities, however, the mineral industry that is believed to be the most radiologically significant is the mining, milling, processing and disposal of phosphates. The products of phosphate processing are very widely distributed – fertilisers, detergents and acids used in other processes. Environmental exposures occur both from use of these products and from the waste products, particularly phosphogypsum²⁹, which are used in building materials. UNSCEAR (1993) estimate a per capita average annual effective dose of ~10 µSv but doses to critical groups are likely to far exceed this.

The phosphate rock typically contains around 1500 Bq/kg of uranium and radium but some sedimentary ores contain up to 20 kBq/kg of U (UNSCEAR 2008). In general, phosphate rock of sedimentary origin is richer in uranium-series nuclides whereas igneous-derived phosphates such as those from Kola (Russian Federation) or Siilinjärvi and Sokli^{30,31} in Northern Finland contain lower U and higher Th, and the total activity is also usually lower (UNSCEAR 2008). In 2010, crude phosphate rock production was about 200 Mt (Heffer & Prud'homme 2010). The main product was fertilisers – 36.4 Mt were produced, predominantly in China, United States, India, Russian Federation, Tunisia and Morocco (Heffer & Prud'homme 2010).

The as-mined phosphate rock is treated to concentrate the phosphate ore but this does not reduce the activity of the ore. More than 90 % of phosphate ore is treated with acid to produce phosphoric acid with phosphogypsum as the by-product. A 'rule of thumb' indicates that each tonne of P₂O₅ production results in 4 to 5 tonnes of phosphogypsum (IAEA 2003d and references therein). About 80 % of the Ra-226, 30 % of Th-232 and 14 % of U-238 remain in the phosphogypsum while the fertiliser produced contains

²⁹ This is gypsum precipitated as a by-product when apatite or phosphorite, the main phosphate-containing minerals, are treated with sulphuric acid to produce phosphoric acid. Radionuclides, particularly Ra-226, co-precipitate with the gypsum.

³⁰ Solatie et al. (2010) give values of 8.5 kBq/kg for Th-232 and 2.2 kBq/kg for U-238 in the niobium ore (high grade Nb-Ta-P ores) from Sokli which was analysed as part of a baseline study by STUK.

³¹ Siilinjärvi is the largest phosphate ore mine in Europe, producing around 10 Mt apatite per annum which is then used to manufacture around 0.5 Mt phosphate fertiliser and 0.3 Mt phosphoric acid (company data available from www.yara.com). Solatie et al. (2010) give values of 8.5 kBq/kg for Th-232 and 2.2 kBq/kg for U-238 in the niobium ore (high grade Nb-Ta-P ores) from Sokli which was analysed as part of a baseline study by STUK.

about 70 % of the U and Th and thus is enriched by about 150 % compared to the phosphate ore (IAEA 2003d and references therein).

Elemental phosphorus is produced by a thermal process in which the phosphate rock is melted with sand, iron oxide and coal. The residual slag contains almost all of the U, Th and Ra and some of the Pb. The rest of the Pb is filtered from the gases leaving the furnace but Po is mainly lost in the off-gases (IAEA 2003d). U-238 concentrations in the slag are usually around 1100 Bq/kg and Ra-226 concentrations around 1300–1500 Bq/kg. The slags are used in road building as hardcore, as concrete aggregate and in plaster wallboards.

In the recent past in Europe, the waste from phosphate processing has been discharged to rivers and the sea in large amounts. UNSCEAR (2008) give some examples:

- prior to 1990 France discharged about 3 Mt of phosphogypsum into Baie de la Seine;
- in the U.K. the annual discharge of Po-210 exceeded 0.5 TBq in the period 1980–1983;
- two phosphoric acid plants in the Netherlands were responsible for release of about 0.6–0.8 TBq of Ra-226 per year (UNSCEAR 2000);
- about 10 Mt of phosphogypsum³² was produced in the European Union in 1993 of which 25 % was discharged to the sea and 60 % stored on land (the remainder was used in construction and building materials).

More recently, Europe has tended to import phosphoric acid directly, rather than phosphate ore, so that production of phosphogypsum has declined, although the problem has largely just been shifted to the ore-producing countries such as Morocco and Tunisia (UNSCEAR 2008).

The environmental radiological impacts of fertiliser production have been investigated and summarised in an IFA/UNEP/UNIDO report (IFA 1998). In general, the very limited solubility of gypsum in phosphogypsum storage heaps means that the radionuclides are retained in the mineral phase (Burnett & Elzerman 2002; Silva et al. 2002), the fluxes of Ra to the groundwater are low and consumption of contaminated groundwater supplies does not give rise to significant health impacts (Burnett 1998). The surface of the heaps also tends to harden as the gypsum dries so that dust generation is also low. The major concern is intrusion into the heaps when high exposures can result from radon and external radiation. Similarly, where phosphogypsum discharge into rivers or coastal waters has resulted in subsequent land contamination, exposure to radon and external radiation of populations living close by is also of concern (IFA 1998).

A significant flux of phosphate-derived radionuclides is via fertiliser use. Studies have been carried out investigating the radiological impacts of use of phosphate fertiliser or

³² IAEA (2003b) gives values for radionuclide activities (in Bq/kg) in phosphogypsum produced in Europe as: Ra-226 15–1700; U-238 500; Pb-210 1300; Po-210 900; Th-232 10. These are at the higher end of typical ranges worldwide.

phosphogypsum on agricultural land. Table 11-16 gives typical activity levels in different phosphate fertilisers and Table 11-17 shows averages, weighted by production volume, over several different types of phosphate fertiliser produced by five factories in Finland. Mustonen (1985) analysed samples of NPK fertilisers used for agriculture, forestry and domestic gardens in Finland³³ for U-238, U-235, Ra-226, and Th-232. From the average activity levels, radionuclide fluxes were estimated based on an annual phosphate application rate of 30 kg/ha: it was estimated that the annual contribution of U-238 in the fertilisers was about 0.25 % of the total U naturally-occurring in the upper 10 cm layer of the soil. The applied phosphate eventually mixes with greater depths of soil (probably a 25-cm layer), so the annual U contribution would be less than this.

Table 11-16. Activities of radionuclides (Bq/kg) in phosphate products (IAEA 2003d and references therein).

Product	U-238	Ra-226	Th-232
Phosphoric acid	1200–1500	300	-
Normal superphosphate	520–1100	110 ⁽¹⁾ –960	15–44
Triple superphosphate	800–2160	230 ⁽¹⁾ –800	44–48
Mono-ammonium phosphate	2000	20	63
Diammonium phosphate	2300	210	<15
Dicalcium phosphate	-	740	<37
PK ⁽²⁾ fertilisers	410	370	<15
NP ⁽³⁾ fertilisers	920	310	<30
NPK ⁽⁴⁾ fertilisers	440–470	210–270	<15

Notes:

- 1: Low values for Russian Federation (igneous-derived phosphates)
- 2: Phosphate-potash
- 3: Nitrogen-phosphate
- 4: Nitrogen-phosphate-potash

³³ It should be noted that the levels of naturally-occurring radionuclides in fertilisers will vary depending on the feedstock (phosphate ore or phosphoric acid) used in their manufacture. As commercial fertiliser producers are likely to vary their source of feedstock from year to year, according to desired quality, price, availability and transport costs, it is also likely that there will be corresponding changes to the levels of radionuclides in the products. In countries which have a domestic phosphate supply (such as Finland) these variations will probably be smaller than in countries which source all their phosphate from a variety of imports. Thus the values given, e.g. in Table 11-16, represent specific examples from a point in time rather than a fixed state.

Table 11-17. Mean activities of radionuclides (Bq/kg P) in compound fertilisers from various factories in Finland (Mustonen 1985) and the annual flux arising from an application of 30 kg/ha P of the weighted mean composition.

Factory	U-238	U-235	Ra-226	Ra-228	Th-228
I	2900	160	830	87	210
II	980	42	290	59	140
III	2400	130	770	95	220
IV	7400	420	1500	62	200
V	4100	230	2000	100	160
Weighted mean	3800	210	1100	78	190
Flux assuming 30 kg/ha P applied [MBq/(km ² ·a)]					
Application	11.4	0.63	3.3	0.23	0.57

Mortvedt & Beaton (1995) give two further examples which are very much consistent with these results: More (1977, cited in Mortvedt & Beaton 1995) estimated that phosphate fertiliser use would cause an annual increase of about 0.04 % of the total Ra concentration in tilled agricultural soils in Sweden. Rothbaum et al. (1979, referenced in Mortvedt & Beaton 1995) reported that most of the U applied in superphosphate or triple superphosphate in long-term field experiments at Rothamsted, England, and also in New Zealand, remained in the upper 23-cm layer of the soil.

Long-term effects have also been studied. For example, triple superphosphate made from Florida phosphate rock applied at a rate of 30 kg/ha phosphate per annum over 50 years did not change U, Th and Ra concentrations in vegetation compared with untreated plots (IAEA 2003d). However, if the U content is still considered problematic, it is possible to extract the uranium from the fertiliser. Currently these products command a premium price but it has been suggested that phosphate processing represents an under-utilised source of U for nuclear power (Ragheb & Khasawneh 2010).

Phosphogypsum is also used as a fertiliser and soil conditioner – it is the primary use in the United States, accounting for 1—2 % of production, since its use in construction and building materials was banned in 1992 due to concerns about the radon emissions (IAEA 2003d). A field study examined the effects on soil and vegetation of large applications of phosphogypsum – at 10,000 kg/ha and 20,000 kg/ha phosphogypsum compared to the regulatory limited rate of 400 kg/ha – by monitoring over a period of 5 years (Rechcigl et al. 2001). In some respects the results were inconclusive due to the low level of measured effects. However, modelling assessment of the impacts of limited phosphogypsum application of 400 kg/ha (see Table 11-18 for representative fluxes) to farmland over a 100 year period suggests that the increase in soil Ra concentrations would be within the normal variation (<20 % increase).

Table 11-18. Ranges of activities of radionuclides (Bq/kg) in phosphogypsum from Florida (from data in IAEA 2003d) and the flux [GBq/(km²·a)] arising from use of North Florida phosphogypsum as a fertiliser/soil conditioner assuming an application rate of 400 kg/ha per year.

Source	U-238	Ra-226	Pb-210	Po-210	Th-232
North Florida	270–599	22–451	348–551	355–566	-
South Florida	507–1358	41–366	577–1853	437–1765	11
N. Florida mid	430	235	450	460	-
Flux assuming 400kg/ha North Florida mid applied annually [MBq/(km ² ·a)]					
Application	17.2	9.4	18.0	18.4	-

With conservative assumptions about human consumption behaviour, the forage-beef-human pathway suggested additional annual individual doses of <1–2 % of background. However, the major radiological impacts were for Rn inhalation, external exposure (assuming dwellings built over the treated land) and groundwater consumption. In total, the phosphogypsum impacts over all pathways gave an additional lifetime risk³⁴ of $6.6 \cdot 10^{-4}$ (for a lifetime of 75.2 years).

11.3.5 Doses arising from NORM activities

The discussion in the preceding sections has focused on the fluxes and concentrations of naturally-occurring radionuclides in various industrial processes but it would clearly be useful to look at the level of doses to exposed members of the public.

The discussion on the coal power production in Section 11.3.2 makes clear the scale of the industry and the large flux of radionuclides it induces. But this activity is spread around the world, in developed and less developed countries so the question arises of how significant is this flux for individual humans – can the average effective dose be estimated, or the effective dose to particular populations which might be particularly exposed by proximity to power stations or sites where the coal slag or ash is dumped?

UNSCEAR (2000) give the estimated releases of radionuclides to the atmosphere for a typical 600 MWe coal-fired power plant (Table 11-14) and the concomitant maximum effective dose rate from all exposure pathways to the most exposed population living in the vicinity of the plant to be 12 µSv/a. This was based on the study of Leenhouts et al. (1996) in the Netherlands and was almost an order of magnitude higher than previous UNSCEAR estimates (UNSCEAR 1993). However, this gives an idea of the scale of

³⁴ *Lifetime risk* as used here is the cumulative probability of fatal radiation-induced cancer in an individual arising as a result of the annual dose from all the exposure pathways multiplied by the life expectancy of 75.2 years. Thus exposure is simply assumed to be constant over a whole lifetime. Further, *additional lifetime risk* means that this is the extra risk arising to the population exposed to the phosphogypsum-derived radiation compared to a population, similar in other respects of background radiation, eating habits and lifestyle etc. but not exposed to this source of radiation. The estimation of risk rising from exposure, especially at low doses and where dose varies over time, is still somewhat uncertain, as discussed e.g. by Muirhead & Preston (2001) who were responsible for the cancer risk estimates in the UNSCEAR (2000) report.

exposure for the people most affected by the plant itself but studies from the UK (Penfold et al. 1998, Smith et al. 2001a) have looked at the detailed breakdown of exposures of different groups and by different parts of the power generation cycle as shown in Table 11-19.

This breakdown shows that the main hazard apparently does not arise during the coal burning but from exposure to the ash when it is used in construction materials. However, a large proportion of the dose from these materials arises from other components – cement, plaster (gypsum), aggregate in concrete etc. and, in fact, the doses are not greatly higher than for other building materials (Smith et al. 2001a, Watson et al. 2005).

The UK Health Protection Agency published a review of exposure of the UK population to radiation arising from industrial processes (Watson et al. 2005) which includes a comparison of various non-nuclear industries. Some of their data are shown in Table 11-20, including effective doses arising from the use of gas, both for power production and for cooking. It is worth noting that the effective dose to workers in commercial kitchen may be up to 500 $\mu\text{Sv/a}$ arising from cooking with natural gas.

Table 11-19. Individual dose arising from various types of exposure to coal-fired power production (from Smith et al. 2001a).

Exposure and pathways	Exposed individual	Dose rate ($\mu\text{Sv/a}$)
PP atmospheric releases ¹ inhalation, ingestion & direct irradiation	Hypothetical critical group ²	1.5
PP atmospheric release ¹ inhalation, ingestion & direct irradiation	Typical individual living in the vicinity ³	0.1
Ash pile exposure ⁴ inhalation, ingestion & direct irradiation	Hypothetical critical group ²	1.3
Ash pile exposure ⁴ inhalation, ingestion & direct irradiation	Typical individual living in the vicinity ³	0.1
Rn inhalation from building materials in the home	Home occupant	893 (362) ⁵

Notes:

- 1: From the UK coal-fired power plant emitting the largest quantity of ash to the atmosphere
- 2: For atmospheric release this was a farmer 500 m living and working from the plant, for the ash pile it was individuals undertaking activities which maximised their exposure.
- 3: 5 km downwind of the plant
- 4: From the UK's largest ash pile
- 5: In brackets, the proportion arising from the coal ash

Also of interest is the dose calculated to arise from discharges of NORM-contaminated production waters from oil and gas production in the North Sea. However, at 10 $\mu\text{Sv/a}$ to individuals of the critical group, this is much smaller than has been estimated for disposal of phosphate waste at sea or in rivers. This has led to detectable increases in Po-210 concentrations in marine organisms commonly used as seafoods (McDonald et al. 1996, Timmermans & Van der Steen 1996). In the case of contamination in the Irish Sea from a phosphate processing plant in Whitehaven, Cumbria, McDonald et al. (1996) estimated that critical group annual doses can be in the order of mSv. UNSCEAR (1993) quotes the study by Köster et al. (1985) that examined the impact of the discharge to the river Rhine of 2 million tonnes per year of phosphogypsum by the Netherlands fertiliser industry. Until the practice was stopped, it was estimated to result in a maximum individual annual effective dose of 150 μSv (Köster et al. 1985). In Spain, discharge of 0.4 million tonnes per annum of phosphogypsum into a river estuary is estimated to give rise to an individual annual effective dose of 60 μSv mainly from consumption of fish and shellfish (Cancio et al. 1989).

Leenhouts et al. (1996) also include some data for the phosphate industry, estimating that elemental phosphorus production gives rise to a maximum annual effective dose of around 130 μSv for external irradiation (the dominant exposure pathway in their analysis) to the critical group, phosphoric acid production gives rise to annual doses from exposure via air dispersion pathways of around 2000 μSv (but this is noted as a 'rather uncertain value') compared with 8 μSv by external radiation. The maximum effective annual dose for fertiliser production is split between external irradiation (20 μSv) and water dispersion pathways (15 μSv) (Leenhouts et al. 1996). This was for the critical group of the general public.

Table 11-20. Radiation doses to critical groups and average individual due to releases of naturally-occurring radioactive materials in industry, typical of a recent year (Watson et al. 2005).

Industry	Discharge route	Pathway	Annual Critical Group Dose (μSv)	Annual Average Dose (μSv)
Coal-fired power stations	Atmospheric release via stack ¹	All	1.5	0.1
	Building materials made from power station ash	Inhalation of radon	600 ⁽²⁾	
	Building materials made from power station ash	External	900 ⁽²⁾	
Oil and gas extraction	Authorised discharges to sea and NORM-contaminated scales	Ingestion of seafood and external exposure to fishing gear	< 30	
	Discharges to sea of NORM-contaminated produced water		10	
Gas-fired power station	Atmospheric releases via stack ¹	All	0.75	0.032
Use of natural gas	Cooking with natural gas ³	Inhalation and external	< 20 to 500	< 10

Notes:

- 1 The critical group are individuals living in close proximity to the power station. Average doses are to members of the public living in the locality of power stations.
- 2 As this dose is not necessarily higher than from other building materials it is not strictly a critical group dose.
- 3 Critical group dose to workers in a commercial kitchen, average dose is to average dose to the population from cooking with gas.

11.3.6 Conclusions

Table 11-21 provides a summary of radionuclide fluxes to the atmosphere and to water (sea, river or groundwater) associated with the three NORM-producing industries discussed. The data from UNSCEAR (2000) are put in context by adding an indication of the approximate proportion of world production represented by the ‘typical installations’. Solid wastes that are stored or disposed of, e.g. phosphogypsum heaps, coal ash in landfills, are not included.

This overview of the NORM arising from large-scale industrial activities indicates that the fluxes of naturally-occurring radionuclides they create are great enough to give rise to significant radiation exposure in some circumstances. These exposure situations usually occur when the NORM is concentrated as in coal ash, dumped without appropriate care or is present in waste materials that are used in situation which enhance exposure – use of phosphogypsum in building materials, for instance. Workers in these

industries are also exposed to a far greater degree than even the critical groups of the general public.

These fluxes represent very much a ‘worst case’ comparison for fluxes arising from a spent nuclear fuel repository. In some respects, it is rather surprising that the phosphate industry, which is relatively little known by the general public, turns out to be a greater potential risk to health than environmental radiological effects of oil and gas extraction or coal burning for power generation, both of which have been the subject of negative environmentalist campaigns, albeit due to concerns in recent years about anthropogenic global warming rather than radioactivity.

These industries are not the only ones to give rise to NORM – processes as diverse as drinking water treatment, ceramics production, cement production, titanium pigment production and geothermal power generation are all implicated to various degrees, depending on the origin and composition of raw materials. But the cases discussed serve to highlight the scale of fluxes of NORM both worldwide and on a more local basis.

Table 11-21. Release of radionuclides from typical installations of mineral processing industries (GBq/a) (data from UNSCEAR 2000).

			Release to atmosphere (GBq/a)						
Industry	Throughput ⁽¹⁾ kt/a	% world annual ⁽²⁾	U-238	Th-232	Ra-226	Rn-222	Pb-210	Po-210	K-40
Elemental phosphorus	570	<10	-	-	-	563	66	490	-
Phosphoric acid	700	<0.5	0.07	0.002	0.09	820	0.08	0.14	0.008
Phosphate fertiliser	375	~1	-	-	-	221	0.044	0.034	-
Coal-fired PP ⁽³⁾	1350	0.04	0.16	0.08	0.11	43	0.4	0.8	0.27
Oil extraction	3500	<0.1	-	-	-	540	-	-	-
Gas extraction	72,000 ⁽⁴⁾	~2	-	-	-	500	-	-	-
			Release to water (GBq/a)						
Elemental phosphorus	570	<10	-	-	-	-	24	166	-
Phosphoric acid	700	<0.5	336	8	737	-	654	997	79
Phosphate fertiliser	375	~1	-	-	-	-	0.054	0.057	-
Coal-fired PP ⁽³⁾	1350	0.04	-	-	-	-	-	-	-
Oil extraction	3500	<0.1	-	217	174	174	174	174	-
Gas extraction	72,000 ⁽⁴⁾	~2	-	2.7	32	32	32	32	-

Notes:

- 1: Annual throughput of feedstock (e.g. phosphate ore concentrate, phosphoric acid [for fertiliser], coal) or production (oil, gas)
- 2: Estimate of the proportion of the global annual production for the industry
- 3: 600 MWe power plant
- 4: Gas production by volume (10^6 m³/a)

12 THE EVOLUTION OF THE REPOSITORY SYSTEM BEYOND A MILLION YEARS IN THE FUTURE

12.1 Evolution of the site beyond 1 Ma

Performance Assessment describes the expected lines of evolution of the repository system for the assessment period of 1 Ma.

The far future evolution extending beyond 1 Ma years can be evaluated qualitatively by looking at the geological history of the Olkiluoto site (see Table 12-1 for a summary of the geological evolution) and extrapolating the knowledge of the past taking into account the known geological processes in action today. Although not all events can be predicted on such a long timescale, the main processes affecting the site can be assessed conceptually.

Plate tectonics will start to change the geographical situation and geological setting eventually but the timescales are tens to hundreds of millions of years for significant change. Olkiluoto is situated away from the plate margins and will be located away from the active regions also in the far future (50 Ma and beyond) according to the plate tectonic forecasts based on knowledge of the current regime as well as on the reconstructed global plate tectonic history (see e.g. Scotese 2001).

In the quest to build reasonable conjectures about the geological future beyond 1 Ma, the main focus is on the Quaternary events (the last 2 Ma) and, for the larger context, to the period of the Tertiary (66 Ma–2 Ma) in which the geographical position of Finland was similar to the present.

During the first million years AP, glacial and interglacial periods will most likely form the basis for the geological processes affecting the site (see *Performance Assessment* and *Formulation of Radionuclide Release Scenarios*). Glacial erosion and related mass redistribution will be expected to continue at low rates which can be predicted from the past glacial cycles (see Section 7.5). An alternative possibility is that the climate will change towards warmer conditions, which will lead to even less erosion than is to be expected during glacial/interglacial periods as the topography of the site is already flat.

The repository closure structures will be affected by the same processes as the bedrock. In some cases the erosion may affect the uppermost plugs and backfilled locations where they are composed of materials more prone to erosional forces than the bedrock itself. In addition to erosion, particularly in case of global warming, sedimentary conditions are also a possibility. As can be seen in Table 12-1, for hundreds of millions of years a variety of conditions have prevailed. On the larger timescale, for example, the fact that the Lappajärvi impact crater formed 73 Ma ago and the crater is still observable after subsequent geological processes, including 2 Ma years of repeated Quaternary glaciations, does tell a story of fairly stable conditions and slow surface processes. It is also worth noting that much older crater remnants have been discovered in Finland (see Table 7-2).

How changing loads from burial and erosion in the far future will affect the deformation properties of the site is, of course, uncertain and it cannot be assumed that the pattern

and properties of the brittle geological structures will remain the same in spite of the stable tectonic setting. However, these structures originally formed over one billion years ago (see Table 12-1) and have stayed relatively stable during the last tens of millions of years at least. On this timescale, reactivation seems to occur only in already existing brittle deformation structures giving support to the idea that no great changes will occur to the present brittle structures at the site in the next several million years.

12.2 Evolution of the engineered barriers

The long-term stability of the barriers relies on the material selection for the expected conditions. The long-term behaviour of the repository components has been evaluated in earlier chapters at length, from perspective of the support given by natural and archaeological analogues to the results of the safety and performance assessments made of those components. However, the analysis of the performance becomes increasingly uncertain after 1 Ma. Much has been written about the problems of handling the increasing uncertainties inherent in consideration of processes over very long times – although usually only up to about 1 million years is meant by long times in this context, e.g. NEA (2009a) states *“The time frames covered by modelling in recent safety assessments range from 10,000 years to one hundred million years, although a million years seems to be emerging as a commonly accepted time frame in recent safety assessments.”* So any statements concerning even longer times are almost inevitably going to be based on observations from natural systems – an extension of the arguments that have been set out in the preceding chapters.

It is not expected that all the canisters will fail at the same time, but that failures will occur gradually, distributed over a long period of time. As discussed in Smith et al. (2007b) (see also Crawford & Wilmot 1998), for the majority of canisters, the most likely eventual failure mechanism is the slow corrosion of the copper overpack. The time span over which this might occur can be illustrated by the observations from natural copper deposits such as the Littleham mudstone (Section 8.2.2). Here thin sheets of native copper within a naturally-compacted mudstone with far from the favourable properties of the EBS bentonite buffer have survived for at least 170 Ma. These observations support the possibility that, if copper corrosion is the main failure mechanism, then failures will be distributed over many millions of years as long as the buffer retains its hydraulic barrier function.

One of the arguments for using bentonite is that it is a natural material, known to persist in a range of geochemical environments (Section 8.3). This argument can be extended to support the contention that the bentonite barrier will continue to perform its functions for much longer periods than considered in the safety assessment. It is possible that over thousands of years, some bentonite will be degraded by contact with the leachate from cement-based low-pH grouts used in fractures in proximity to the deposition tunnel or from concrete in the deposition tunnel low-pH concrete plugs. Perhaps, over longer periods, some bentonite will be chemically eroded in a few localised disposal positions. These processes will affect the integrity of the buffer around some waste containers, but that they will occur universally and potentially affect all the canisters seems unlikely even over very long periods, especially considering the actually relatively limited amounts of cement in the repository and the care with which disposal positions are chosen.

Table 12-1. *Timescale for Olkiluoto geological evolution and selected regional events.*

Event/process	Age (Ma)	Reference
Olkiluoto bedrock formation		
D2 deformed tonalitic gneisses at Olkiluoto	~1851–1863	Mänttari et al. 2006, 2007a
Igneous Lähteenmäki tonalite ca. 16 km SE of Olkiluoto	1863±7	Suominen et al. 1997
Olkiluoto pegmatite granite	1860–1820	Mänttari et al. 2006
Formation of SE dipping thrust faults (Fennian orogeny)	~1800	Aaltonen et al. 2010a
Laitila rapakivi batholith	~1580	
Diabases at Olkiluoto	~1560	Mertanen 2007
NE-SW strike-slip faults	1560–1270	Aaltonen et al. 2010a
Possible fault reactivation	1385±27	Mänttari et al. 2007b
Possible fault reactivation	1373±27	Mänttari et al. 2007b
Possible fault reactivation	1225±24	Mänttari et al. 2007b
Later regional events		
Satakunta sandstone deposition	~1400	
Possible fault reactivation	912±18	Mänttari et al. 2007b
Lake Sääksjärvi impact structure (consists of 180 m thick suevite breccia and impact melt breccia above deformed Proterozoic mica gneiss (Pihlaja & Kujala 2000).	560	Papunen 1973, Müller et al. 1990, Elo et al. 1992, Pihlaja & Kujala 2000
Possible fault reactivation	550±11	Mänttari et al. 2007b
Sedimentation of Palaeozoic Mesozoic sedimentary cover	540–70	Paulamäki & Kuivamäki 2006
Faulting disturbing both proterozoic basement and palaeozoic sedimentary cover	Late palaeozoic	Paulamäki & Kuivamäki 2006
Lappajärvi meteorite impact (Palaeozoic-Mesozoic sedimentary cover still existed)	73	Mänttari & Koivisto 2001
Erosion of the Palaeozoic-Mesozoic sedimentary cover		Paulamäki & Kuivamäki 2006
Tectonic uplift of western Scandinavia in connection to opening of the North Atlantic Ocean	~65–5	Riis 1996, Stuevold & Eldholm 1996
Uplift in the Bothnian Sea area ~500 m	~65–5	Van Balens & Heeremans 1998
Weathering of Olkiluoto bedrock is assumed to be a results of processes taking tens of millions of years (few to 56 m)	10s of Mas	Aaltonen et al. 2010a
Quaternary period begins (repeated glaciations)	2.6	
Olkiluoto emerged from the Weichselian ice cover	0.011	Posiva 2009
Olkiluoto emerged from the Baltic Sea	0.003	Eronen & Lehtinen 1996
present	0	
Operational phase	0.0001	
Biosphere assessment period	0.01	
Temperate period	0.01–0.05	
Assessment period	1	
Intracratonic location remains	50 and beyond	Scotese 2001

The conclusion of the natural and anthropogenic analogue studies described in Chapter 8 seems to be that there are not really any processes which will universally degrade the buffer properties and that the long-term durability of the key component of the bentonite – the montmorillonite – is vulnerable only to chemical alteration by substances supplied in groundwater, most likely illitisation. The rate of this process is uncertain and the process itself not well understood (e.g. Karnland & Birgersson 2006) but with respect to the performance of the buffer over periods greater than 1 Ma probably the most important observations come from natural bentonites in a wide range of environments. These observations support the conclusion of longevity over geological timescales of this material, in the absence of a ready supply of potassium, even at temperatures higher than the repository ambient temperature.

So there is the possibility that the copper canisters will remain more or less intact – even if technically failed i.e. radionuclides are able to escape – and still surrounded by a hydraulic barrier to some extent over periods much greater than one million years. However, after tens to hundreds of millions of years, when the repository horizon may be much closer to the surface, the further evolution of the repository materials is uncertain and any comments are necessarily speculative. It is possible, but unlikely, that the fuel, radionuclides and repository construction materials will eventually become widely dispersed in the geological environment. It is more likely that at least some of the materials, including the spent nuclear fuel, will remain largely in situ. For example, the copper may be partly replaced under reducing conditions by a suite of copper sulphides, which have very low solubility and are not likely to become dispersed until erosion brings the repository horizon to the surface, and the fuel matrix may experience only limited dissolution over time due to its relative geochemical stability. Thus, in some respects, after very long times the repository materials may tend to resemble a heterogeneous uranium ore body, perhaps analogous to granite- or sediment-hosted Cu-U deposits. The consequences of possible exhumation of the repository are difficult to assess, given the extreme length of time before this could occur. However, the processes involved are likely to be similar to exhumation of small uranium deposits where the local climatic and topographic conditions primarily determine the rate at which the ore body is dispersed.

13 SUMMARY

13.1 Support for the robustness of the KBS-3 method

The safe disposal of spent nuclear fuel aims to ensure that it is isolated from the biosphere and potential impact on humans and the environment is minimised for very long periods. Geological disposal has been identified as the way in which these aims can most effectively be achieved (Chapter 4), and the KBS-3 method (Chapter 5) is a way of implementing geological disposal for the specific spent nuclear fuel and geological conditions found in Finland.

According to Neall et al. (2007), “A disposal concept based on a system of passive barriers which provide multiple safety functions through processes and properties that are well understood and that is not unduly affected by the residual uncertainties can be said to be robust”.

The three key objectives are isolation, containment, and retardation for the radionuclides (cf. Figure 5-2). Isolation – in terms of removing the spent nuclear fuel from the sphere of human influence (and interest) – is predominantly a function of the repository site itself (and the depth of the repository, see e.g. Figure 5-2) and support for the choice of Olkiluoto as the site is summarised in the following section. But containment and retardation are the functions that are required to be performed for hundreds of thousands of years by the engineered components of the KBS-3 repository in conjunction with the repository host rock and the geological environment.

The KBS-3 method uses a few simple, common materials, with which we are familiar, for the engineered barriers: copper and iron for the canister, swelling clay for the buffer and backfill (the host rock is also a barrier but is not considered ‘engineered’ as the canister and buffer/backfill). The purpose of this choice is to reduce both the number of components whose properties need to be understood and also the number of potential interactions between those components. Demonstration of safety means that the long-term stability of the engineered barriers must be robustly assured and by choosing these materials which are long lasting (occurrences of them have been identified that have lasted over geological timescales) and with which there is already long experience, this requirement is simplified. Of course, the very long timescales of interest – much longer than historical experience – means that much more than empirical experience is required and understanding of the processes involved, for example, in copper corrosion or bentonite swelling and extrusion, must be based on a combination of experimental evidence and the study of natural analogues. Furthermore, the models used for the interpretation of natural analogues and to make the safety assessment calculations are based on the application of fundamental laws of nature, such as mass and energy balances and the laws of thermodynamics, which provide a robust basis for the results.

As detailed in Chapter 8, our experience with natural analogues for both materials and processes is providing a large degree of confidence in our understanding of the disposal system and how it will evolve over hundreds of thousands of years. In Appendix C an overview of the available natural analogue data in support of the Olkiluoto safety case (TURVA-2012) is given in table form. For each engineered barrier and key process,

there is increasing analogue evidence to support the models and parameters. For example, taking the copper canister;

- although there is no ‘perfect’ analogue, there is strong evidence from a range of natural occurrences of native copper for very low corrosion rates of Cu for millions of years under the groundwater and redox conditions similar to, or less favourable than, those at Olkiluoto;
- archaeological artefacts, while representing much more variable and often more severe conditions, suggest low localised copper corrosion rates that are not likely to be significant in determining canister longevity compared with general corrosion.

Taken together, this supports the assertion that the copper canister can provide complete containment for at least hundreds of thousands of years if the surrounding environment maintains the favourable chemical conditions and protects the canister from rock movements. This means that the bentonite buffer needs to maintain its low permeability and plasticity, and prevent microbial activity (which could cause sulphate reduction).

So considering the bentonite,

- On the basis of observations from natural systems, there is good evidence to indicate that temperature-induced changes occur in bentonite at temperatures above about 150–200 °C, but that significant changes are also dependent on a supply of potassium, which will be limited in the buffer (due to favourable groundwater composition and limited amount of potassium introduced associated with foreign materials to the system). This supports the assertion of stability of the bentonite buffer during the repository thermal period if the maximum buffer temperature is around 90–100 °C;
- Several analogue (and many experimental) studies have examined the chemical stability of bentonite under various conditions, but interaction with cement leachate seems to be the only potential detrimental consideration. The use of high pH cement, either as a component of concrete or cement-based grouts can be avoided by design, substituted by other materials (silica sol grout, for example) or replaced (low pH concrete) so as to ensure the minimum possible interaction. Mass balance calculations also suggest that with these reasonable precautions, the amount of unavoidable cement in the repository is unlikely to cause any significant detriment (e.g. Alexander & Neall 2007);
- There are several excellent illustrations of the long-term performance of bentonite as a hydraulic barrier in preserving wood and human cadavers, which also suggest that microbial activity was significantly reduced (this aspect of microbial activity has also been addressed by microbiological studies).

Natural analogue studies as well as many experimental studies suggest that, with suitable repository design and implementation, there are no significant threats to the integrity and performance of the bentonite buffer. Infiltration of large volumes of fresh water to repository depth after periods of glaciation has raised some concerns due to the potential for loss of bentonite colloids in water-conducting fractures, resulting in mass and density loss. However, this seems unlikely and, if it were to occur, would not affect many canister positions, since efforts are made to avoid such fractures intersecting deposition holes. The provision in the repository layout of 20 % extra deposition hole

positions will allow any positions that are considered to be significantly at risk from fractures identified during excavation to be excluded during operations, thus minimising the number of deposition holes that could be affected.

This brings up another aspect supporting the robustness of the KBS-3 method: the procedures put in place for ensuring quality throughout the design, engineering and repository implementation stages leading to final completion. These processes constitute the key components of the safety concept – as seen in Figure 5-2, the base of the pedestal for the pillars of safety is ‘Robust system design’ and the right hand pillar is ‘Proven technical quality of the EBS’.

The definition of robustness also includes a mention of uncertainties. Addressing uncertainties is part of the procedures for the implementation of the KBS-3 concept: for example, one of the strengths of KBS-3 is its simplicity of components and materials, and foreign materials – essentially anything which is not part of the EBS – introduce uncertainty as they increase the number of chemical components and the number of possible interactions between them. Thus tracking (and minimising) all foreign materials offers an additional support to the robustness of the resulting disposal system.

13.2 Support for the suitability of geological disposal at the Olkiluoto site

For deep geological disposal, a wide range of concepts has been studied and many potential host rocks have been identified, including crystalline basement, salt, basalt, tuff and a range of argillaceous sediments. The relative emphasis in the safety concept on the EBS and the geological barriers varies between national programmes. These differences reflect obvious features, such as the particular host rock, or are a consequence of the national regulatory requirements.

In Finland, the range of geology available has limited choices of host rock type as there are no evaporites, tuffs, or argillaceous sedimentary sequences of sufficient extent to provide suitable sites. Thus, fractured crystalline basement necessarily became the host rock and a detailed understanding of the favourable characteristics of a site is needed in order to select an appropriate specific site. Such rocks, however, have also been considered as suitable for locating a deep geological repository in many other countries, including several in which alternative rock formations (salt, basalt, tuff and a range of argillaceous sediments) are available e.g. in Canada (see e.g. AECL 1994, p. 3-4 and Witherspoon & Bodvarsson 2006), Switzerland (Nagra 1993), the USA (Rechard et al. 2011) and Japan (JNC 2005 and NUMO 2004).

The important safety functions provided by the specific host rock and geosphere at a site are:

- physical isolation due to depth, which minimises the risks of future perturbations (e.g. due to glaciation) and human intrusion;
- mechanical protection of the EBS;
- favourable geochemical conditions (key characteristics being redox, pH and total salinity) and sufficiently low groundwater flow rates; and

- an additional barrier providing retardation of radionuclides, if they are released, due to uptake on minerals and diffusion into the rock matrix.

In addition to geological stability of the site, which given the quiescence of the Fennoscandian Shield area over the recent millions of years is a characteristic common to most of Finland, the key parameters influencing the performance of the geosphere are the amounts and composition of groundwater (the influx) and the nature of the paths through which it flows (geometry and rock-water interaction).

The groundwater flux is defined by the hydraulic conductivity and the hydraulic gradient. Advective flow can be so small that solute transport is dominated by the process of molecular diffusion. Such a diffusion-dominated system leads to high performance of both the EBS and the geological barrier. If the flux is significant, the overall performance of the geological barrier depends on the properties of the flow paths in which advection occurs and also means that a disposal concept must be chosen which is not too sensitive to this parameter, for example, by using hydraulic barriers such as the bentonite buffer.

The performance of both the engineered barriers and the geosphere as a retardation barrier for radionuclides is improved considerably when conditions are chemically reducing. This is because corrosion processes are generally slower under such conditions and many key radionuclides are both less soluble and more highly sorbed.

Total salinity can play a significant role in defining the effectiveness of the barrier system. Most information supporting the design and performance of the EBS and radionuclide retention in the geosphere is defined for conditions of low to medium salinity. However, the presence of old, very dense brines can provide evidence of geological stability as these brines indicate groundwater movement has been very low and exchange with lower salinity groundwaters in overlying rocks has been limited over very long periods of time. Thus they contribute strong evidence for the favourable performance of this barrier.

The pH of groundwater is a less sensitive parameter and is unlikely to have much direct impact on performance of the total system unless it lies at the very high (>10, say) or very low (<4) end of the range. It is therefore advantageous if the host rock has the capacity to buffer any pH excursions caused by either perturbations to the rock itself or directly by the repository.

So from a consideration of these and other factors, Olkiluoto was chosen from the four potential candidate sites – Hästholmen, Kivetty and Romuvaara being the others – identified in the site selection process.

The overall favourability of the Olkiluoto site has been discussed in detail in *Site Description* (Chapter 11) and in Chapter 7 in this report. The site characteristics are well known, and it was noted previously by Neall et al. (2007) that the Olkiluoto site was admirably suited as a repository site because “*Olkiluoto site characterisation activities have been on-going for over 20 years and the current state of knowledge about the site can be summarised by noting that surface conditions, geology, rock mechanical properties and the status of in situ stress are well understood although some*

uncertainties remain. It has been found that the rate of groundwater flow at the planned repository depth is low and geochemical conditions are favourable to the engineered barrier system with reducing conditions, low levels of sulphide and moderate salinity of about 10–20 g/L. Furthermore, the number of major, fast water transport pathways via the fracture zones is low in the repository area and their characteristics are known,...

and all this is still perfectly correct. However, if one looks at the bigger picture, there are other reasons for accepting the suitability of Olkiluoto:

- crystalline rock has been investigated as a potential repository (including L/ILW repositories) host rock in numerous countries, including Argentina, Canada, China, France, Japan, Norway, Sweden, Switzerland, the UK and the USA;
- L/ILW repositories already exist in this rock type in Norway (Himdalen), Sweden (Forsmark) and Finland (Loviisa and Olkiluoto);
- coastal sites such as Olkiluoto have also been assessed in Japan, Korea, Sweden and the UK and two coastal repositories are already in place – Gyeongju in Korea and Rokkasho in Japan.

Evidence that a site like Olkiluoto is appropriate as a repository host rock can be seen by the presence of numerous ore bodies throughout the crystalline Fennoscandian Shield. Their very existence indicates the fundamental barrier properties of fractured crystalline rocks where the geochemical environment of the host rock is appropriate for radionuclide retention – as it most certainly is at Olkiluoto. Even at a disturbed site such as nearby Palmottu (see Figure 7-2), penetration of oxidising glacial meltwaters into the site was buffered by the host rock after only a hundred metres depth (see Figure 7-3), indicating the suitability of the crystalline host rock for a repository.

Like Palmottu, the Olkiluoto site also shows indications of the presence of glacial meltwaters at depth, but not at repository depth. And also as in the case of Palmottu, meltwaters appear to have been mixed with deeper, more mineralised groundwaters or to have been buffered by rock-water interactions so preventing dilute meltwaters reaching the repository horizon.

Although data are still sparse (e.g. Pedersen 2008, Pedersen et al. 2010), microbial populations are generally low at the Olkiluoto site, in line with values for most deep crystalline groundwaters (e.g. Smith et al. 2001b, Fukuda et al. 2010). Additional work in the ONKALO facility will allow this feature to be evaluated more thoroughly in future.

To all the information above it must be added that the use of complementary indicators, for which background is given in Chapter 11, will aid in putting the results of the safety analysis presented in *Assessment of Radionuclide Release Scenarios for the Repository System* and *Biosphere Assessment* in a broader perspective to show, as has been done previously (see Neall et al. 2007), that the radiation originating from a spent nuclear fuel repository remains in most cases much below natural background radiation or that caused by non-nuclear industries.

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APPENDIX A. RADIOTOXICITY

Geological repositories provide safety by isolating the spent nuclear fuel from humans and their environment and containing the radionuclides associated with the spent nuclear fuel. The possibility of some eventual release of radioactivity cannot, however, be completely excluded. The harmful stochastic effects on human health due to exposure to any such releases are quantitatively related to the annual effective dose to an adult individual. For convenience, the term “annual effective dose”, or simply “dose”, is used in this report.

Not all radionuclides are equal when it comes to impact on the human body so simply comparing the activity – in disintegrations per second or Becquerels – of two radionuclides does not give a good comparison of their relative harm. Radiological toxicity or “radiotoxicity” is a concept which has been used to make comparisons of different radionuclides more meaningful in terms of the potential damage they can do. However, it is worth noting that there is no universally agreed definition of radiotoxicity but NEA (2004b) suggests that *“it can, for example, be loosely defined as the hypothetical dose resulting from ingestion of the radioactive material”*. They further suggest the radiotoxicity can provide a useful indicator in two particular roles in safety assessment:

- *“At times when geological stability can be assured, it can usefully illustrate the rationale behind the strategy of isolating and containing the waste while radioactive decay reduces the associated hazard. In spite of the limits of predictability [...], the methods of geological sciences are generally adequate to show, for a suitably chosen site, that the waste will be isolated in a repository for a sufficient time for its radiological toxicity to decrease substantially.*
- *“At later times when the geological stability of the site can no longer be assumed and less weight is attached to the results of dose and risk calculations, radiological toxicity gives an indication of the residual hazard posed by the waste and provides a safety indicator that can complement dose and risk if accepted reference values or yardsticks for comparison can be derived.” (NEA 2004b)*

The radiological hazard associated with a given amount of radioactive material (whether contained in the repository or released to its surroundings or, indeed arising from a natural source) is also sometimes expressed in terms of a “radiotoxicity index” by comparing the dose arising from ingesting a quantity of a radionuclide to a regulatory limit or constraint (Nagra 2002, Hedin 1997).

In Eq. A-1, the radiotoxicity index or RTI(t) [-] is defined as the hypothetical dose at time t resulting from ingestion of the activity $A_j(t)$ [Bq] of radionuclide j, divided by 0.1 mSv (derived from the Finnish regulatory dose constraint for the most exposed person):

$$RTI(t) = \frac{\sum_j A_j(t) D_j}{10^{-4}} \quad (\text{Eq. A-1})$$

where D_j [Sv/Bq] is the dose coefficient for ingestion (see Appendix B for a table of these coefficients).

The RTI allows a direct comparison of the radiological hazard through hypothetical ingestion associated with radioactive waste, or radionuclides released from spent nuclear fuel, with that associated with different natural materials.

For comparisons of radionuclide releases associated with natural processes such as weathering with releases of radionuclides from the repository, an alternative indicator is the “radiotoxicity flux”, or RTF. This considers the RTI of a flux of radionuclides across a given interface, which could be an area of the ground surface undergoing weathering or a hypothetical plane in the subsurface across which groundwater is considered to flow. The RTF [RTI/a] is defined by replacing the activity $A_j(t)$ in Eq. A-1 by the annual activity flux $F_j(t)$ across that interface, see Eq. A-2.

$$RTF(t) = \frac{\sum_j F_j(t) D_j}{10^{-4}} \quad (\text{Eq. A-2})$$

The RTF can be used for direct comparison of the radiological hazards of activity fluxes from the repository with those from natural activity fluxes (Table 11-2).

Comparison of natural radioactive materials – by activity, RTI or RTF – with the spent nuclear fuel or radionuclides released from the repository forms the basis of complementary safety indicators which, as noted by NEA (2004b) can provide an indication of the relative hazards at long times when more conventional measures such as dose no longer apply.

It should be noted that these measures of hazard relate mainly to ingestion (or, by a trivial modification, to inhalation). The repository protects against the hazard associated with external exposure, by shielding and by its deep underground location. Hazard due to external exposure is, however, relevant in the case of human intrusion scenarios (exposure to the intruder) and also in groundwater scenarios where radionuclides accumulate in soils and sediments.

REFERENCES TO APPENDIX A

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APPENDIX B. CALCULATIONS AND DATA FOR CHAPTER 3

The information and data used to generate the radiotoxicity indices and radiotoxicity index fluxes of spent nuclear fuel and natural materials shown in figures in Chapter 3 are presented in this appendix.

Radiotoxicity index of spent nuclear fuel

The description of the radionuclide composition and activity of 1 tonne of Finnish spent OL3 (EPR in some figures) fuel (60 MWd/kgU, 4.2 % enrichment), used as the reference in Chapter 3, is given in Anttila (2005) at eight different times up to 1 million years after unloading from the reactors. The nuclide activity data are reproduced in Table B-1. The dose coefficients for the radionuclides in the spent nuclear fuel are listed in Table B-2 based on ICRP (1996), and Avila & Bergström (2006).

The RTI for the spent OL3 fuel at the eight times calculated by Anttila (2005) are given in Table B-3. The RTI values for the full inventory in Fig. 3-1 were obtained by multiplying the RTI for 1 tonne of reference OL3 spent nuclear fuel by 9000 to account for the total amount of spent nuclear fuel planned for disposal in the repository (*Description of the Disposal System*) – the differences in RTI of different spent nuclear fuel types / burn-ups is not taken into account.

Radiotoxicity index of natural materials

The natural materials that are used in the radiotoxicity comparisons in Chapter 3 are natural uranium, uranium ores and the Olkiluoto tonalite, which is a component of the TGG (tonalitic-granodioritic-granitic gneisses) and is taken as a ‘representative host rock type’ for its radionuclide content. The radionuclides of interest in these natural materials are U-235, U-238, Th-232 and K-40. The dose coefficients used for these radionuclides in natural materials is given in Table B-4. Note that the dose coefficients for Th-232, U-235 and U-238 include radioactive decay products (the progeny) as the parent nuclides are assumed to be in equilibrium with the progeny for solid materials.

Table B-5 shows the radionuclide content of the natural Cigar Lake (8 % U) and Palmottu (1600 ppm U) uranium ores and the Olkiluoto tonalitic and granitic rocks along with the activity levels and the resultant RTI per m³ of ore or rock. The details of the U and Th content of the Olkiluoto rocks and the Palmottu ore are from Pitkänen et al. (2003).

The RTIs for the natural materials shown in Fig. 3-3 were based on the assumption that the deposition tunnels of the KBS-3V repository were filled with Cigar Lake (8 % U) or Palmottu (1600 ppm) uranium ores. The volume used for this calculation was $7.2 \cdot 10^5$ m³ (based on the excavation volumes given in Karvonen (2011), scaled for 9000 tU). This was also the figure used for the volume of Olkiluoto tonalite removed during excavation of the same deposition tunnels. The total area of the repository was taken as 2.7 km² and, with a depth of 420 m, this gives a volume of ~1.15 km³ above the repository foot print.

Figure 3-4 shows the RTI of the 8 tonnes of natural uranium from which the original fuel was derived. The details of this calculation are shown in Table B-6.

Table B-1. Radionuclide activity (GBq/tU) of 1 tonne spent nuclear fuel (Finnish OL3-type 60 MWd/kgU, 4.2 % enrichment) at eight times (in years) after unloading from the reactor (after Anttila 2005).

Time (a)	0	5	30	100	1000	10,000	100,000	1,000,000
Light elements								
H-3	1.07E+01	8.08E+00	1.98E+00	3.87E-02	4.10E-24	0.00E+00	0.00E+00	0.00E+00
C-14	3.13E+01	3.13E+01	3.12E+01	3.10E+01	2.78E+01	9.35E+00	1.74E-04	0.00E+00
Cl-36	1.19E+00	1.19E+00	1.19E+00	1.19E+00	1.19E+00	1.17E+00	9.48E-01	1.19E-01
Co-60	1.17E+05	6.08E+04	2.27E+03	2.27E-01	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Ni-63	2.69E+04	2.60E+04	2.18E+04	1.35E+04	2.64E+01	2.26E-26	0.00E+00	0.00E+00
Zr-93	8.84E+00	8.84E+00	8.84E+00	8.84E+00	8.84E+00	8.80E+00	8.45E+00	5.62E+00
Nb-93m	1.75E+04	1.42E+04	4.85E+03	2.65E+02	2.35E+01	1.13E+01	8.45E+00	5.62E+00
Nb-94	4.15E+02	4.15E+02	4.15E+02	4.14E+02	4.01E+02	2.95E+02	1.37E+01	6.14E-13
Mo-93	2.17E+01	2.17E+01	2.16E+01	2.13E+01	1.78E+01	2.99E+00	5.38E-08	0.00E+00
Total	2.24E+07	3.37E+05	3.00E+04	1.44E+04	7.02E+02	5.08E+02	1.10E+02	1.15E+01
Actinides								
Pb-209	0.00E+00	0.00E+00	0.00E+00	7.58E-05	7.71E-03	9.89E-01	2.34E+01	5.79E+01
Pb-210	0.00E+00	0.00E+00	0.00E+00	6.12E-04	2.15E-01	9.99E+00	7.86E+01	2.33E+01
Pb-212	2.45E-01	1.36E+00	1.82E+00	9.09E-01	1.39E-04	2.66E-05	1.13E-04	1.05E-03
Pb-214	0.00E+00	0.00E+00	0.00E+00	1.16E-03	2.15E-01	1.00E+01	7.86E+01	2.33E+01
Bi-210	0.00E+00	0.00E+00	0.00E+00	6.13E-04	2.15E-01	9.99E+00	7.86E+01	2.33E+01
Bi-212	2.45E-01	1.36E+00	1.82E+00	9.09E-01	1.39E-04	2.66E-05	1.13E-04	1.05E-03
Bi-214	0.00E+00	0.00E+00	0.00E+00	1.16E-03	2.15E-01	1.00E+01	7.86E+01	2.33E+01
Po-210	0.00E+00	0.00E+00	0.00E+00	6.13E-04	2.15E-01	9.99E+00	7.86E+01	2.33E+01
Po-214	0.00E+00	0.00E+00	0.00E+00	1.16E-03	2.15E-01	9.99E+00	7.86E+01	2.33E+01
Po-216	2.45E-01	1.36E+00	1.82E+00	9.09E-01	1.39E-04	2.66E-05	1.13E-04	1.05E-03
Po-218	0.00E+00	0.00E+00	0.00E+00	1.16E-03	2.15E-01	1.00E+01	7.86E+01	2.33E+01
Ra-226	0.00E+00	0.00E+00	0.00E+00	1.16E-03	2.15E-01	1.00E+01	7.86E+01	2.33E+01
Th-229	0.00E+00	0.00E+00	0.00E+00	7.58E-05	7.71E-03	9.89E-01	2.34E+01	5.79E+01
Th-230	0.00E+00	2.75E-03	1.32E-02	6.28E-02	1.23E+00	1.29E+01	7.77E+01	2.32E+01
Th-234	1.13E+01	1.14E+01	1.14E+01	1.14E+01	1.14E+01	1.14E+01	1.14E+01	1.14E+01
Pa-233	2.27E+01	2.26E+01	2.37E+01	2.88E+01	6.36E+01	7.44E+01	7.24E+01	5.41E+01
Pa-234m	1.17E+01	1.14E+01	1.14E+01	1.14E+01	1.14E+01	1.14E+01	1.14E+01	1.14E+01
U-233	0.00E+00	1.35E-03	3.86E-03	1.18E-02	2.08E-01	3.03E+00	2.58E+01	5.74E+01
U-234	3.02E+01	3.48E+01	5.53E+01	9.53E+01	1.49E+02	1.46E+02	1.16E+02	1.96E+01
U-236	1.34E+01	1.34E+01	1.34E+01	1.34E+01	1.42E+01	1.87E+01	2.15E+01	2.10E+01
U-238	1.14E+01	1.14E+01	1.14E+01	1.14E+01	1.14E+01	1.14E+01	1.14E+01	1.14E+01
Np-237	2.22E+01	2.26E+01	2.37E+01	2.88E+01	6.36E+01	7.44E+01	7.24E+01	5.41E+01
Np-239	8.00E+08	3.10E+03	3.09E+03	3.07E+03	2.82E+03	1.21E+03	2.55E-01	1.64E-04
Pu-238	3.12E+05	3.21E+05	2.64E+05	1.52E+05	1.33E+02	6.34E-19	0.00E+00	0.00E+00
Pu-239	1.40E+04	1.42E+04	1.42E+04	1.42E+04	1.39E+04	1.11E+04	8.77E+02	1.64E-04
Pu-240	2.75E+04	2.78E+04	2.87E+04	2.90E+04	2.64E+04	1.02E+04	7.60E-01	4.99E-05
Pu-241	7.45E+06	5.85E+06	1.75E+06	5.95E+04	8.18E+01	3.93E+01	2.55E-02	0.00E+00
Pu-242	1.92E+02	1.92E+02	1.92E+02	1.92E+02	1.92E+02	1.89E+02	1.60E+02	3.01E+01
Am-241	9.68E+03	6.24E+04	1.93E+05	2.24E+05	5.35E+04	4.11E+01	2.55E-02	0.00E+00
Am-242m	6.45E+02	6.30E+02	5.57E+02	3.95E+02	4.73E+00	2.89E-19	0.00E+00	0.00E+00
Am-242	5.64E+06	6.27E+02	5.54E+02	3.93E+02	4.71E+00	2.87E-19	0.00E+00	0.00E+00
Am-243	3.10E+03	3.10E+03	3.09E+03	3.07E+03	2.82E+03	1.21E+03	2.55E-01	1.64E-04

Table B-1 continued. Radionuclide activity (GBq/tU) of 1 tonne spent nuclear fuel (Finnish OL3-type 60 MWd/kgU, 4.2 % enrichment) at eight times (in years) after unloading from the reactor (after Anttila 2005).

Time (a)	0	5	30	100	1000	10,000	100,000	1,000,000
Actinides								
Cm-242	3.89E+06	2.17E+03	4.59E+02	3.25E+02	3.89E+00	2.38E-19	0.00E+00	0.00E+00
Cm-243	2.52E+03	2.23E+03	1.22E+03	2.21E+02	6.89E-08	0.00E+00	0.00E+00	0.00E+00
Cm-244	6.96E+05	5.76E+05	2.21E+05	1.51E+04	1.63E-11	0.00E+00	0.00E+00	0.00E+00
Cm-245	8.86E+01	8.86E+01	8.84E+01	8.79E+01	8.16E+01	3.92E+01	2.54E-02	0.00E+00
Cm-246	2.78E+01	2.77E+01	2.76E+01	2.74E+01	2.40E+01	6.41E+00	1.20E-05	0.00E+00
Fission products								
H-3	3.96E+04	2.99E+04	7.33E+03	1.43E+02	1.52E-20	0.00E+00	0.00E+00	0.00E+00
Se-79	4.62E+00	4.62E+00	4.62E+00	4.62E+00	4.61E+00	4.52E+00	3.65E+00	4.40E-01
Kr-85	5.56E+05	4.02E+05	7.99E+04	8.64E+02	4.58E-23	0.00E+00	0.00E+00	0.00E+00
Sr-90	4.45E+06	3.93E+06	2.12E+06	3.79E+05	8.98E-05	0.00E+00	0.00E+00	0.00E+00
Y-90	4.65E+06	3.93E+06	2.12E+06	3.79E+05	8.98E-05	0.00E+00	0.00E+00	0.00E+00
Zr-93	1.10E+02	1.11E+02	1.11E+02	1.11E+02	1.10E+02	1.10E+02	1.06E+02	7.02E+01
Nb-93m	1.20E+01	3.10E+01	8.33E+01	1.09E+02	1.10E+02	1.10E+02	1.06E+02	7.02E+01
Tc-99	8.21E+02	8.23E+02	8.23E+02	8.23E+02	8.21E+02	7.97E+02	5.93E+02	3.09E+01
Pd-107	9.15E+00	9.15E+00	9.15E+00	9.15E+00	9.15E+00	9.14E+00	9.06E+00	8.23E+00
Cd-113m	3.16E+01	2.51E+01	7.35E+00	2.35E-01	1.43E-20	0.00E+00	0.00E+00	0.00E+00
Sn-126	3.81E+01	3.81E+01	3.81E+01	3.81E+01	3.78E+01	3.55E+01	1.91E+01	3.72E-02
Sb-126	1.98E+04	5.33E+00	5.33E+00	5.33E+00	5.30E+00	4.98E+00	2.67E+00	5.21E-03
Sb-126m	2.35E+04	3.81E+01	3.81E+01	3.81E+01	3.78E+01	3.55E+01	1.91E+01	3.72E-02
I-129	1.87E+00	1.88E+00	1.88E+00	1.88E+00	1.88E+00	1.88E+00	1.87E+00	1.80E+00
Cs-134	1.30E+07	2.41E+06	5.40E+02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Cs-135	3.25E+01	3.26E+01	3.26E+01	3.26E+01	3.26E+01	3.25E+01	3.16E+01	2.41E+01
Cs-137	6.95E+06	6.19E+06	3.48E+06	6.90E+05	6.41E-04	0.00E+00	0.00E+00	0.00E+00
Ba-137m	6.61E+06	5.85E+06	3.28E+06	6.51E+05	6.05E-04	0.00E+00	0.00E+00	0.00E+00
Ba-140	5.46E+07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Pm-147	6.32E+06	1.75E+06	2.36E+03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Sm-151	2.17E+04	2.11E+04	1.74E+04	1.02E+04	9.93E+00	7.60E-30	0.00E+00	0.00E+00
Eu-152	3.65E+02	2.81E+02	7.66E+01	2.01E+00	9.48E-21	0.00E+00	0.00E+00	0.00E+00
Eu-154	5.38E+05	3.59E+05	4.78E+04	1.69E+02	5.06E-30	0.00E+00	0.00E+00	0.00E+00
Eu155	2.16E+05	1.03E+05	2.54E+03	7.99E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Total	5.77E+09	2.82E+07	1.12E+07	2.11E+06	1.18E+03	1.14E+03	8.91E+02	2.06E+02

Table B-2. Ingestion dose coefficients for radionuclides in spent nuclear fuel, applicable to adults older than 17 years (see text for source).

Nuclide	Dose coefficient Sv/Bq Adult >17 a	Nuclide	Dose coefficient Sv/Bq Adult > 17 a
H-3	1.8E-11	Ra-223*	1.0E-07
Be-10	1.1E-09	Ra-224*	7.1E-08
C-14	5.8E-10	Ra-225*	1.2E-07
Cl-36	9.3E-10	Ra-226*	2.8E-07
Ca-41	1.9E-10	Ra-228*	6.9E-07
Co-60	3.4E-09	Ac-225*	2.4E-08
Ni-59	6.3E-11	Ac-227*	1.2E-06
Ni-63	1.5E-10	Th-227*	1.1E-07
Se-79	2.9E-09	Th-228*	1.4E-07
Sr-90*	3.1E-08	Th-229*	6.1E-07
Y-90	2.7E-09	Th-230	2.1E-07
Zr-93*	2.6E-09	Th-231	3.4E-10
Nb-93m	1.5E-09	Th-232	2.3E-07
Nb-94	1.7E-09	Th-234*	3.4E-09
Mo-93*	4.6E-09	Pa-231	7.1E-07
Tc-99	6.4E-10	Pa-233	8.7E-10
Pd-107	3.7E-11	U-233	5.1E-08
Ag-108m	2.3E-09	U-234	4.9E-08
Sn-126*	5.1E-09	U-235*	4.7E-08
Sb-126	2.4E-09	U-236	4.7E-08
I-129	1.1E-07	U-238*	4.8E-08
Cs-134	1.9E-08	Np-237*	1.1E-07
Cs-135	2.0E-09	Np-239	8.0E-10
Cs-137	1.3E-08	Pu-238	2.3E-07
Sm-151	9.8E-11	Pu-239	2.5E-07
Eu-152	1.4E-09	Pu-240	2.5E-07
Eu-154	2.0E-09	Pu-241	4.8E-09
Eu-155	3.2E-10	Pu-242	2.4E-07
Ho-166m	2.0E-09	Am-241	2.0E-07
Pb-210*	1.9E-06	Am-242m*	1.9E-07
Bi-210*	1.2E-06	Am-243*	2.0E-07
Po-210	1.2E-06	Cm-244	1.2E-07
Rn-222	3.5E-09	Cm-245	2.1E-07
-	-	Cm-246	2.1E-07

* include contributions from short-lived progeny.

Table B-3. RTI of 1 tonne of OL3 reference spent nuclear fuel (as Table B-1) at eight times (years) after unloading from the reactor (from Anttila 2005).

Cooling time (a)	RTI Fission Products	RTI Actinides	RTI Activation Products	Overall RTI
1	4.76E+12	8.44E+12	4.30E+09	1.32E+13
5	2.49E+12	1.95E+12	2.33E+09	4.44E+12
30	1.11E+12	1.46E+12	1.91E+08	2.57E+12
100	2.07E+11	9.34E+11	3.21E+07	1.14E+12
1000	1.29E+07	2.14E+11	8.04E+06	2.14E+11
10,000	1.26E+07	5.69E+10	5.44E+06	5.69E+10
100,000	1.03E+07	4.73E+09	4.61E+05	4.74E+09
1,000,000	4.50E+06	1.02E+09	1.47E+05	1.03E+09

Table B-4. Dose coefficients for naturally-occurring radionuclides (ingestion for adults older than 17 years).

Nuclide	Dose coefficient (Sv/Bq)
K-40 ¹	$6.20 \cdot 10^{-9}$
Th-232 ²	$1.06 \cdot 10^{-6}$
U-235 ³	$1.97 \cdot 10^{-6}$
U-238 ⁴	$2.43 \cdot 10^{-6}$

1 Nagra (2002)

2 Dose coefficients for Th-232 + Ra-228 + Th-228 + Ra-224 from Table B-2

3 Dose coefficients for U-235 + Th-231 + Pa-231 + Ac-227 + Th-227 + Ra-223 from Table B-2

4 Dose coefficients for U-238 + Th-234 + Th-230 + Ra-226 + Rn-222 + Po-210 + Pb-210 from Table B-2

Table B-5. Activity and RTI of natural U ores and Olkiluoto tonalite.

Nuclide	kg element per kg rock	Bq precursor per kg rock	RTI per kg rock	RTI per m ³ rock
Cigar Lake U ore 8 % (density 2800 kg/m³)¹				
U-235	$8.0 \cdot 10^{-2}$	$4.5 \cdot 10^4$	$8.8 \cdot 10^2$	$2.5 \cdot 10^6$
U-238	$8.0 \cdot 10^{-2}$	$1.0 \cdot 10^6$	$2.5 \cdot 10^4$	$7.0 \cdot 10^7$
Total			$2.6 \cdot 10^4$	$7.2 \cdot 10^7$
Palmottu ore 1600 ppm (density 2700 kg/m³)²				
U-235	$1.6 \cdot 10^{-3}$	$9.1 \cdot 10^2$	$1.8 \cdot 10^1$	$4.9 \cdot 10^4$
U-238	$1.6 \cdot 10^{-3}$	$2.0 \cdot 10^4$	$5.0 \cdot 10^2$	$1.3 \cdot 10^6$
Total			$5.2 \cdot 10^2$	$1.3 \cdot 10^6$
Olkiluoto tonalite (density 2600 kg/m³)²				
K-40	$2.5 \cdot 10^{-2}$	$7.9 \cdot 10^2$	$4.9 \cdot 10^{-2}$	$1.3 \cdot 10^2$
Th-232	$5.8 \cdot 10^{-6}$	$2.4 \cdot 10^1$	$2.6 \cdot 10^{-1}$	$6.8 \cdot 10^2$
U-235	$1.6 \cdot 10^{-6}$	$9.2 \cdot 10^{-1}$	$1.8 \cdot 10^{-2}$	$4.7 \cdot 10^1$
U-238	$1.6 \cdot 10^{-6}$	$2.0 \cdot 10^1$	$5.0 \cdot 10^{-1}$	$1.3 \cdot 10^3$
Total			$8.3 \cdot 10^{-1}$	$2.2 \cdot 10^3$

1 Nagra (2002)

2 Pitkänen et al. (2003)

Table B-6. Activity and radiotoxicity indices of natural uranium (Nagra 2002).

Nuclide	kg element per kg material	Bq precursor per kg material	RTI per kg material	RTI per 8 tonnes uranium
U-235	1	$5.6 \cdot 10^5$	$1.1 \cdot 10^4$	$8.8 \cdot 10^7$
U-238	1	$1.2 \cdot 10^7$	$3.0 \cdot 10^5$	$2.4 \cdot 10^9$
Total			$3.1 \cdot 10^5$	$2.5 \cdot 10^9$

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APPENDIX C. OVERVIEW OF THE AVAILABLE NATURAL ANALOGUE DATA IN SUPPORT OF THE OLKILUOTO SAFETY CASE

Process	Natural analogue data available?	Of direct relevance? Qualitative or quantitative?
Fuel behaviour - criticality - radiolysis - spent nuclear fuel corrosion	Few relevant studies have been carried out Yes, for several orebodies Yes, numerous studies	Oklo clearly shows that criticality is unlikely Data implies that radiolytic change will be minimal. But not enough information on fluxes of oxidants at natural analogue sites is available to allow a direct comparison with the likely situation in and around a SF canister Dissolution rates cannot be quantified readily from natural analogue data, but abundance of naturally-occurring uraninite may be taken as a general indication of its stability in the geological environment. Extrapolation of this apparent longevity to spent nuclear fuel should be done cautiously, due to many uncertain boundary conditions
Canister corrosion - Cu	Numerous studies available, including studies of localised corrosion of copper	Kronan, quantitative Littlehampton, qualitative Rare pitting studies
Canister corrosion - Fe	Numerous studies available, including a few on pitting	Wide range of studies providing quantitative and qualitative data which support the conservative assumptions in SA
Buffer – thermal alteration/cementation	Wide range of studies available	Generally, the vast majority of the natural analogue data available is of little value as the environments studied have little relevance to repository conditions.
Buffer – canister sinking	No	Not applicable
Buffer – chemical alteration - Saline - Canister interaction – copper - Canister interaction – steel - Cement interaction	Yes, various Yes Not really yes	Not really as no full characterisation of 'reacted' samples have been published Kronan, qualitative No Khushaym Matruk, qualitative Cyprus, quantitative Philippines, qualitative Searles Lake, qualitative

Process	Natural analogue data available?	Of direct relevance? Qualitative or quantitative?
Analogues of foreign materials - Cements – longevity - Cement-host rock interaction - Cement carbonation	Yes (Jordan, archaeological examples etc.) Yes, Jordan for OPC (Ordinary Portland Cement) and other sites for low pH cement Not currently, but sites are available	Jordan, quantitative Archaeological generally qualitative Jordan, quantitative Nothing currently for low pH systems Not applicable
Natural analogues of radionuclide retardation in the geosphere - Retardation in clays - Retardation in fractured crystalline rocks - Matrix diffusion - Redox fronts - Colloids	Yes, numerous worldwide, several associated with URL projects Very few, directly relevant natural analogue studies available Yes, one of the most thoroughly studied aspects of retardation in fractured rocks Yes, numerous worldwide, but very few, directly relevant natural analogue studies available Yes, one of the most thoroughly studied aspects of retardation in fractured rocks	Yes, quantitative data on long-term diffusion processes in clay formations and some on a scale representative of the clay buffer in the EBS Yes, but qualitative Yes, clear long-term, long-distance quantitative information Not applicable to a deep geological repository (EBS or geosphere) but one of the least understood

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ISBN 978-951-652-192-6

POSIVA 2012-11

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December 2012

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ISBN 978-951-652-192-6
ISSN 1239-3096