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14th Natural Analogue Working Group Workshop (NAWG14),
Olkiluoto 9-11 June 2015

W. Russell Alexander, Heini Reijonen and Timo Ruskeeniemi (eds)

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14th Natural Analogue Working Group Workshop (NAWG14), Olkiluoto 9–11 June 2015 Abstract Book

Edited by

W. Russell Alexander, Heini Reijonen and Timo Ruskeeniemi

Unless otherwise indicated, the figures have been prepared by the authors of the abstracts.

Front cover: Limestone cap on bentonite, Kato Moni quarry, Cyprus.
Bentonite will be extensively used to isolate waste containers in deep
geological repositories for radioactive wastes.
Photo: W. R. Alexander, Bedrock Geosciences, 5105 Auenstein, Switzerland.

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CONTENTS

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PREFACE

In an attempt to define natural analogue studies more clearly, and to orient them towards individual processes for which good analogues can be found, the founding fathers of NAWG (Natural Analogue Working Group) listed a set of guidelines for selecting natural analogues for investigation. The need for well-characterised, process-oriented natural analogue studies is reaffirmed in this report and thus these guidelines are repeated here:

1) The process involved should be clear-cut. Other processes that may have been involved in the natural system should be identifiable and amenable to quantitative assessment as well, so that their effects can be subtracted from the observations.

2) The chemical analogy should be good. It is not always possible to study the behaviour of a mineral system, chemical element or isotope identical to that of interest in the repository. But as long as the limitations of this are clearly stated and fully understood by the data producers and users, this is acceptable.

3) The magnitude of the various physico-chemical parameters involved (e.g. pressure, temperature, pH, Eh, concentration, etc.) should be determinable, preferably by independent means, and should not differ greatly from those envisaged in a repository.

4) The boundaries of the system should be identifiable (whether it is open or closed, and consequently how much material has been involved in the process being studied).

5) The timescale of the process must be measurable, since this factor is of the greatest significance for a natural analogue.

The quantitative philosophy outlined above proved to be the impetus for greater interest in natural analogues. This resulted in an international symposium in 1984 and the formation of the NAWG, which was originally sponsored by the Commission of the European Communities (CEC, now EC). Since its inception, these themes have been addressed at 14 international NAWG workshops and the discussions from the latest one, held in Rauma, Finland in June 2015, are presented here.

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Chairman of the Organizing Committee

Keywords (GeoRef Thesaurus, AGI): radioactive waste, underground disposal, bedrock, safety, natural analogs, applications, risk assessment, communication, symposia

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INTRODUCTION TO THE NAWG-14 WORKSHOP

by

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ASSESSING REPOSITORY PERFORMANCE AT LONG TIMES

A major challenge in the development of a safety case (SC) for a deep geological repository is dealing with the long period of time over which the wastes remain hazardous. Over such a period, a wide range of events and processes act 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, 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 of the wastes, even at times beyond when quantitative safety assessments can be supported. NEA (2009) 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 SC is that, although some experiments can be carried out 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 geological and/or anthropogenic 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.

To complement the quantitative estimates of doses, especially in the period beyond a few tens of thousands of years, additional complementary safety indicators have been proposed (IAEA 2003) 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 (2003) 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 of the radionuclides remaining in the engineered barriers or released to the geosphere decreases due to radioactive decay below the corresponding values for relevant natural materials, such as the original uranium ore or the excavated host rock. Both these areas will be briefly explored here.

INTRODUCTION TO NATURAL ANALOGUES – WHAT ARE THEY AND WHAT DO THEY BRING TO THE SAFETY CASE?

The main arguments employed here relate to natural analogues of the repository systems or processes. As noted in Alexander et al. (2015), argumentation by use of analogy is well established in many fields including philosophy, biology, linguistics and law (Petit 1992), and most earth scientists are familiar with this approach and will have used it at some point in their career. For example, in the oil industry, accessible (surface) analogues of the geological conditions expected in physically inaccessible deep oil and gas reservoirs are often studied.

For the specific case of radioactive waste disposal, the main inaccessible features are:

- the very long time it will take for long-lived waste to decay to safe levels – how can anyone know how the materials that are used to contain the wastes will behave over thousands to millions of years?
- the large spatial scales, which cannot be directly addressed in a laboratory – how can the migration of radionuclides through several hundred metres of host rock from the repository to the earth's surface be studied and modelled?
- the heterogeneity and structural complexity of the geological environment that will host the repository – how can this ever be approached in a laboratory or modelled on a computer?

Hence, the study of natural (predominantly geological) systems has been termed natural analogue research within the radioactive waste disposal community and the term "natural analogue" (NA) has developed a particular meaning associated with providing supporting arguments for a repository safety case (see, for example, Chapman et al. 1984, Côme & Chapman 1986, Miller et al. 1994, 2000, Posiva 2013a, Alexander et al. 2014, for discussion). As noted above, the key factors here are the heterogeneity and complexity of natural systems and, in particular, the very large dimensions and long timescales over which safety must be assured.

Due to the long timescales of concern, the basis of most safety cases is a quantitative evaluation that is based on complex mathematical models and their general lack of transparency only adds to the mistrust of many stakeholders. How then can people be convinced that it is possible to assess the performance (and thus ensure the safety) of a repository over the long timescales of interest? One way is to address the robustness of the safety assessment models, by clearly indicating the form and extent of model testing carried out within the repository safety assessment. Not only can this show that the individual component parts of the complex structure that constitutes most safety assessment models have been checked, but also that the 'mathematical black boxes' (cf. Alexander et al. 2003) constitute an acceptable representation of the repository system.

As noted by Alexander et al. (1998), part of the problem undoubtedly lies in the unusual nature of radioactive waste disposal: in most major engineering projects, such as bridge construction or aerospace engineering, the designs are tested against a range of laboratory experiments backed up by expert judgement based on experience with the same or similar systems. Here, repository design deviates from standard engineering practice in that only a few repositories currently exist and testing their compliance with design limits will be impossible due to the timescales involved. In addition, peoples' anxiety about most things radioactive means that they require some greater form of 'proof' that a repository is safe than they are willing to accept for other engineered systems (see discussion in West et al. 2002, West & McKinley 2007). This being the case, significant additional effort has been expended within the radioactive waste disposal community to make it clear that the SA models can adequately predict the long-term behaviour of a repository.

WHAT ARE THE ADVANTAGES OF NATURAL ANALOGUES?

Traditionally, SA modellers have placed much weight on laboratory data for the construction and testing of their safety assessment models and, with only a few exceptions (e.g. Posiva 2013b), have not integrated in their SA reports data from either NAs or *in situ* experiments in URLs (Underground Rock Laboratories) – see discussion in Alexander et al. (1998). The over-dependence on laboratory data is understandable in that the information is produced under well understood, fully controlled conditions and thus the modellers feel they can place a high degree of confidence in the results obtained. Unfortunately, the full complexity of a repository cannot be re-created in a laboratory and it is necessary to address processes that are influenced by natural heterogeneities, which include large degrees of uncertainty and which operate over very long timescales. In this case, it is necessary to supplement laboratory data with information from *in situ* URL experiments and NAs. The potential evolution of geological repositories 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 (along with archaeological and anthropogenic) analogues - systems that have similar properties to components of repositories, have a unique role to play.

In its basic form, an NA study can be any form of investigation of any relevant natural system, as long as it provides quantitative or qualitative information that can be used to support (and build confidence in) geological disposal. This may mean that a study provides data that are directly applicable to the safety case or, alternatively, it may provide illustrations of concepts or processes that can demonstrate safety (Reijonen et al. 2015). Each repository design will require unique information to assist in building and presenting the SC, but NA studies have historically tended to focus on only a narrow range of natural systems. They can thus be categorised into a few broad groups that are representative of some major components of a repository system or feature of its evolution, namely:

- natural geological and geochemical systems
- archaeological systems
- sites of anthropogenic contamination

It should be noted, however, that this focus is currently changing: Reijonen et al. (2015), for example, present an example of a more broad-based approach to the use of NA in supporting the safety case, whereas Baik et al. (2015) and Wolf & Noseck (2015) look to define specific forms of NA support for repositories in crystalline and evaporite host rocks, respectively.

It is beyond the scope of this publication to provide an overview of the vast range of natural analogues that have been studied; for this, the reader is invited to examine publications focussed on just that, such as Miller et al. (2000, 2006), CSN (2004), Degnan et al. (2005) and Brasser et al. (2008).

NAWG – THE NATURAL ANALOGUE WORKING GROUP

In an attempt to define natural analogue studies more clearly, and to orient them towards individual processes for which good analogues can be found, Chapman et al. (1984) listed a set of guidelines for selecting natural analogues for investigation. The need for well-characterised, process-oriented natural analogue studies is reaffirmed in this report and thus these guidelines are repeated here:

1) The process involved should be clear-cut. Other processes that may have been involved in the geochemical system should be identifiable and amenable to quantitative assessment as well, so that their effects can be subtracted.
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3) The magnitude of the various physico-chemical parameters involved (e.g. pressure, temperature, pH, Eh, concentration, etc.) should be determinable, preferably by independent means, and should not differ greatly from those envisaged in a repository.
4) The boundaries of the system should be identifiable (whether it is open or closed, and consequently how much material has been involved in the process being studied).
5) The timescale of the process must be measurable, since this factor is of the greatest significance for a natural analogue.

The quantitative philosophy outlined by Chapman et al. (1984) proved to be the impetus for a greater interest in natural analogues. This resulted in an international symposium (Smellie, 1984) and the formation of the Natural Analogue Working Group (NAWG), which was originally sponsored by the Commission of the European Communities (CEC, now EC). Since its inception, these themes have been addressed at 14 international NAWG workshops.

- 14th Workshop: 9–11th June 2015 in Rauma, Finland. Here, the application of NAs to repository SA was discussed
- 13th Workshop: 13–16th May 2013 in Nagoya, Japan. A wide area of NA study was covered, and 9 of the presentations were published in the Swiss Journal of Geosciences (volume 108, 2015)
- 12th Workshop: 11–13th May 2011 in Larnaca, Cyprus, where NAs of engineered barriers were predominantly discussed
- 11th Workshop: Liverpool, UK, 2009, where a short meeting was held at the 12th ICEM (International Conference on Environmental Remediation)
- 10th Workshop: Munich, Germany, 2007, where the workshop examined how current and future studies could be better focused on providing appropriate data for the various end-users of natural analogue data
- 9th Workshop: Aarau, Switzerland, 2002, where the theme was the current international status of natural analogues
- 8th Workshop: Strasbourg, France, 1999, was devoted to the presentation of three major international natural analogue projects, Oklo (II), Palmottu and Pena Blanca (EUR 19118)
- 7th Workshop: Stein am Rhein, Switzerland, 1996, where one of the main themes of the workshop was the application of natural analogues to toxic wastes (EUR 17851 EN)
- 6th Workshop: Santa Fe, USA, 1994, where the intention was to review the "state-of-the-art" of several key issues in near-field and far-field processes and their importance to PA, with the intention to provide a consensus view of the remaining areas requiring further research in natural analogues (EUR 16761)
- 5th Workshop: Toledo, Spain, 1992, was held in association with the final workshop of the Alligator Rivers Analogues Project (EUR 15176)
- 4th Workshop: Pitlochry, Scotland, 1990, was devoted to reviewing 5 years of NA studies and the final conclusions drawn from the Poços de Caldas study (EUR 13014)
- 3rd Workshop: Snowbird, USA, 1988, where the application of natural analogues to repository performance assessment was discussed (EUR 11725)
- 2nd Workshop: Interlaken, Switzerland, 1986, where anthropogenic analogues and the role of colloids, complexes and microbes were reviewed (EUR 10671)
- 1st Workshop: Brussels, Belgium, 1985, where the theme was the interaction between modellers and experimenters (EUR 10315)

NAWG reports are a particularly valuable record of the evolution and application of natural analogue studies. Most of the large and significant analogues have been represented at NAWG meetings. Furthermore, the NAWG reports attempt to develop the perception of natural analogues by giving agreed introductory statements on their development.

ADVANCES MADE SINCE THE FORMATION OF NAWG

- The study of natural analogues has greatly increased understanding of repository-relevant processes and improved the capability to describe and effectively model them.
- Larger, multi-objective analogue studies are a very cost effective way of training site characterisation and SA groups on real, complex systems.
- The application of analogues in broadening public perception of the natural context of waste disposal is under development.
- An increased awareness of the potential for studying natural analogues of chemotoxic waste.
- The significant role of natural analogues in understanding the long-term impacts of CO2 sequestration

NAWG continues to grow (currently 25 members from 19 countries), and the 15th workshop will be held in Prague at the end of May 2017

14TH WORKSHOP OF THE NATURAL ANALOGUE WORKING GROUP, 9−11TH JUNE 2015, RAUMA, FINLAND

A three-day meeting discussing the application of NA to radioactive waste disposal was held at Rauma, next to the Finnish final disposal site for spent nuclear fuel (SF) at Olkiluoto Island, in Eurajoki municipality. The workshop was kindly hosted by Posiva Oy, the implementer of geological SF disposal in Finland, at the visitor centre of Olkiluoto and at Vuojoki Mansion. In addition to the two-day workshop, a field excursion was also organised to the Olkiluoto underground exhibition at the low and intermediate level waste repository next to the Olkiluoto nuclear power plants.

The timing of the workshop in Finland was optimal in the sense that a major step in the licencing process of the SF repository had just been achieved; the construction licence application had been submitted to the regulator just two years before, and the safety case for the application finalised. In the safety case for the SF repository, natural analogues were extensively used in the complementary considerations report (Posiva 2013a), which complemented the safety assessment by providing alternative lines of argumentation, and supporting the safety assessments results with relevant examples of long-term processes considered in the safety case.

NAWG 14 was attended by a group of scientists working in wide spectrum of different organisations and companies acting in the radioactive waste management industry. In total, around 40 people attended the workshop, representing a range of countries including Switzerland, Japan, Spain, Czech Republic, Sweden, Canada, UK, Netherlands, Germany, Korea and Finland.

Presentations covered a wide range of natural system processes relevant to the safety case of a wide range of waste types and repository designs. In addition to perhaps more ‘traditional’ engineered barrier system-specific and radionuclide migration-related issues, the emphasis in this workshop was also placed on geosphere processes and how they can be better understood via natural analogue research. These included:

- the Saimaa project in Finland, focusing on deep groundwater chemistry at sites where massive marginal end-moraines occur, in order to examine the

effects of a stagnant ice margin and consequent longer recharge of dilute melt waters in the deep groundwater systems
- the recently completed multinational GAP (Greenland Analogue project), focussing on deep groundwater chemistry at a site where a large continental ice-sheet has affected/ is affecting the groundwater system for even longer periods of time (cf. Saimaa)
- post-glacial faults (also in Finland), improving understanding of crustal adjustment after glacial periods
- fracture mineral investigations, which will focus on understanding buffer montmorillonite reaction to differing groundwaters by looking at the same mineral in the natural systems on site (at Olkiluoto in this case, but the approach is applicable to any host rock)

In addition to the above geosphere processes, novel NAs of materials that will be introduced to repository systems (e.g. bituminous seals) were discussed: often, the long-term behaviour can only be assessed through NA information, especially when conceptualising the systems/processes to be assessed.

Potential still exists to increase knowledge and the conceptual understanding of material mixtures and coupled processes: for example, how does bentonite-sand mixture behave or what is the combined effect of certain processes (e.g. heat and high pH) on bentonite behaviour. In addition, the degree of potential over-conservatism of SC assumptions can be reassessed when new information emerges from natural system investigations. To give an example, high temperatures are inherent in all high-level waste and SF repositories, and it has long been studied in relation to bentonite buffer longevity. While bentonite behaviour is well understood at temperature below 100°C, there is still potential to look at natural systems and ascertain whether it is over-conservative to impose a 100°C or even a 150°C limit on the temperature in the bentonite.

This short introduction does not list all the topics discussed but, when glancing though the agenda, it is apparent that much of relevance to SC can be learned from appropriate NAs and that information from natural (and anthrogeneic and industrial) systems can be utilised in many phases in disposal projects and for various purposes, from scientific input to the SC to stakeholder communication.

We plan to continue these fruitful discussions at NAWG 15, which will be held in Prague in May 2017. In addition to this collection of abstracts, please note that all the presentations are also appended.

Photo: Tae-Jin Park, KAERI.

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CHEMICAL EROSION OF BENTONITE – TRUE OR FALSE?

by

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INTRODUCTION

Bentonites are planned to be used as buffer materials in various types of geological repositories designed to host radioactive waste. Especially in the case of HLW/SF disposal, the longevity of compacted bentonite in the repository environment is highly important due to the very long timescales for the safety assessment (up to My). For the bentonite buffer and backfill materials, one of the main environmental factors affecting their evolution is the hydrological and hydrogeochemical conditions. It has been observed in laboratory tests that smectite, especially montmorillonite, forms colloids and dissolves in very dilute conditions. A scenario for chemical erosion has consequently been discussed at length, particularly in relation to geological repositories located in future glaciated terrains and locations otherwise potentially hosting dilute groundwater conditions (e.g. Posiva 2013a).

The general understanding is that bentonite erosion does not occur when the total charge equivalent of cations in groundwater is higher than 4 mM, which is reflected in the requirements (SKB 2011, Posiva 2012). Regarding the occurrence of chemical erosion within bentonite EBS, two processes need to be considered: 1) infiltration of dilute waters in the bedrock aquifer and related rock–water interaction in the system, and 2) the actual process of erosion of swelling clay in repository conditions, assuming that the former produces sufficiently dilute conditions.

DILUTION PROCESS OF GROUNDWATER

When assessing the infiltration of dilute waters into deep bedrock (~400 m below ground level in the case of Olkiluoto), the paleohydrogeological data are first examined. Site data provide no indication of dilute glacial water infiltration ~300 m or more below the ground surface (Posiva 2013b). In addition to glacial infiltrating waters, long-term meteoric water recharge and consequent dilution additionally need to be considered, since the climate scenarios projected to the future also consider the possibility of extended global warming. In order to understand the processes controlling the dilution, other sites than Olkiluoto can also be considered for increased process understanding. Data are available from several settings in the fractured Finnish bedrock that display differences, for example, in the topographical setting, structural geology, and setting in relation to the melting ice sheet during the last glaciation. Data on groundwater chemistry in deep settings can be obtained at least from the site-selection sites (sites considered for repository) (see McEwen & Äikäs 2000 and references therein), Palmottu (Smellie at al. 2002) and Hyrkkölä (Marcos & Ahonen 1999) in southern Finland, and Outokumpu deep borehole (Kietäväinen et al. 2014). Sites from Sweden are also well studied and could provide even more relevant data (e.g. Forsmark, Oskarshamn, Äspö). Another potential source of information is the GAP project (see e.g. Claesson Liljedahl *in prep*).

NATURAL ANALOGUES FOR SMECTITE STABILITY IN RELATION TO SALINITY VARIATION

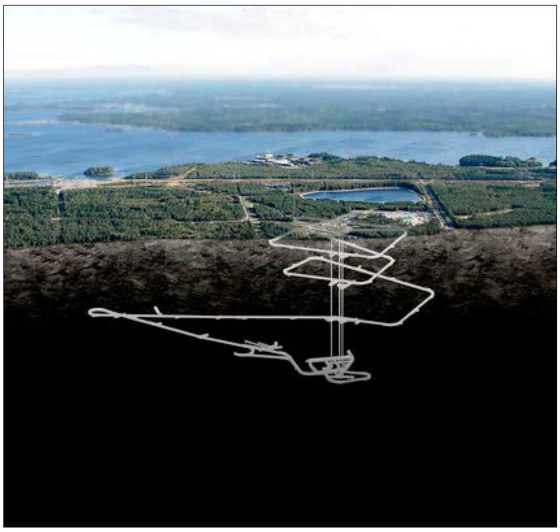
However, the focus of this paper is on the natural analogue potential for studying smectite stability in various salinity conditions, especially those showing dilute compositions. As discussed by Reijonen & Alexander (2015), surficial bentonite deposits and their interaction with meteoric water can be examined, but distinguishing other erosional mechanisms from the effects of meteoric water poses complications. Fortunately, smectite also occurs elsewhere than in bentonites. It is additionally found in a wide spectrum of geological settings within the Fennoscandian shield, for example:

- weathering zones of crystalline basement (e.g. Vartiainen 2005),
- altered ultramafic rocks such as lamprophyres and kimberlites (Suominen 1997, Laine & O'Brien 2008),
- as a fracture filling mineral, e.g. at Olkiluoto, Hyrkkölä, Hästholmen, Kivetty, Romuvaara, and Forsmark (see e.g. Front et al. 1998, Marcos 2002, Gehör et al. 1997a, Gehör et al. 1997b, Kärki et al. 1997, Sandström et al. 2008).

Of these, Olkiluoto provides a "self-analogue" and the potential to study smectite longevity on site in open and closed fractures. Figure 1 presents observations of montmorillonite from shallow (<100 m depth) ONKALO samples. At Olkiluoto, the current groundwater is generally saline enough (regarding the requirements, see above), and there is relatively large variation vertically from fresh surface waters to saline waters at depth (see e.g. Posiva 2013b).

In addition, Kivetty and Hyrkkölä sites provide naturally more dilute groundwater conditions that can be used as a comparison for the Olkiluoto data. A detailed study on the mode of occurrence of these smectites is needed on order to have sufficiently detailed data for use in the safety case. The age of smectites and potential alteration are also currently unknown. In Figure 2, smectite from the Hyrkkölä locality is illustrated. Here, smectite occurs in an open fracture location at about 80 m depth. The current groundwater ionic strength is about 1.5 mM.

At the Kivetty site in central Finland, montmorillonite has been reported to occur throughout the depth range in the deep drill holes from 40 to 1000 m depth (m.b.s.l.) (Fig. 3). It was also observed here that the hydraulically conductive fractures host most of the clay minerals that occur in fracture gouges (kaolinite, montmorillonite, illite) (Gehör et al. 1997a). Groundwaters at the Kivetty site are



		Fraction		
		<2 µm	2-20 µm	20-63 µm
Sample PL70				
Montmorillonite		**x**		**x**
Sample PL81				
Montmorillonite		**x**	**x**	
Sample PL87				
Montmorillonite		**x**	**x**	**x**
Sample PL295				
Montmorillonite			**x**	
Sample PL957				
Montmorillonite		**x**		
Sample PL960				
Montmorillonite		**x**	**x**	

Fig. 1. The ONKALO facility at Olkiluoto and montmorillonite observations (by Gehör 2007) from the fracture gouges from samples taken at relatively shallow depths (<100 m below the ground surface). The depth of the facilities shown in the ONKALO picture is about 450 m the below ground surface.

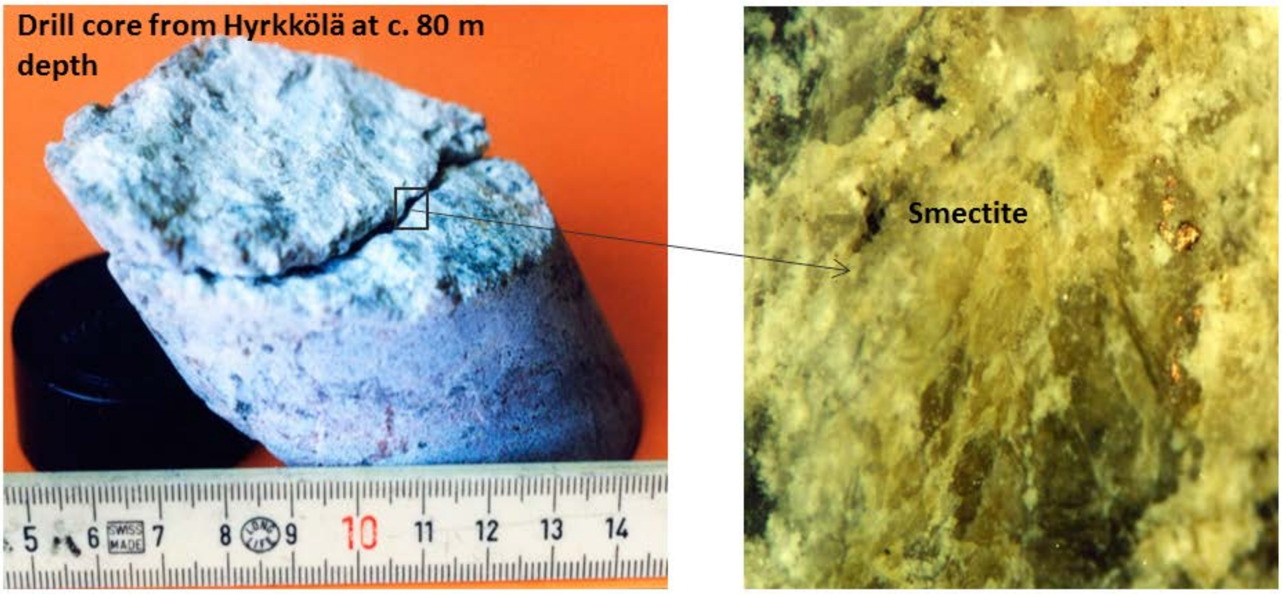


Fig. 2. Hyrkkölä smectite occurrence at ~-80 m depth (Marcos 2002).

fresh (TDS < 1 g/L; Pitkänen et al. 1998), with reported minimum and maximum TDS values of 133–240 mg/L. Considering the requirement limit for bentonite buffer (>4 mM), Kivetty groundwaters show a lower ionic strength than the requirement. Based on the minimum and maximum values for the main cations given in McEwen & Äikäs (2000), the range of ionic strength is 0.8–3.6 mM.

CONCLUSIONS

As an answer to the question raised in the title, it is noted that bentonite erosion as a process exists, but whether such conditions are encountered in nature and how the smectite will behave is not yet completely clear. This is because:

- at dilute groundwater sites, smectite is seemingly stable, and this would imply that the stability of smectite is better in nature than in experiments conducted in the laboratory;
- the reason for the apparent difference has not been studied in detail (regarding natural smectites).

This difference is most likely due to differences in mineralogy, especially the EC of smectite and groundwater chemistry vs. laboratory groundwater simulant compositions. Since montmorillonite has been observed in the XRDs from the fracture minerals, there is a good possibility of having smectites that are directly analogous to EBS bentonite (which usually mostly consists of montmorillonite) from among the natural occurrences mentioned in this paper. In addition, the potential exists for enlarging the scope to other smectites, e.g. saponite.

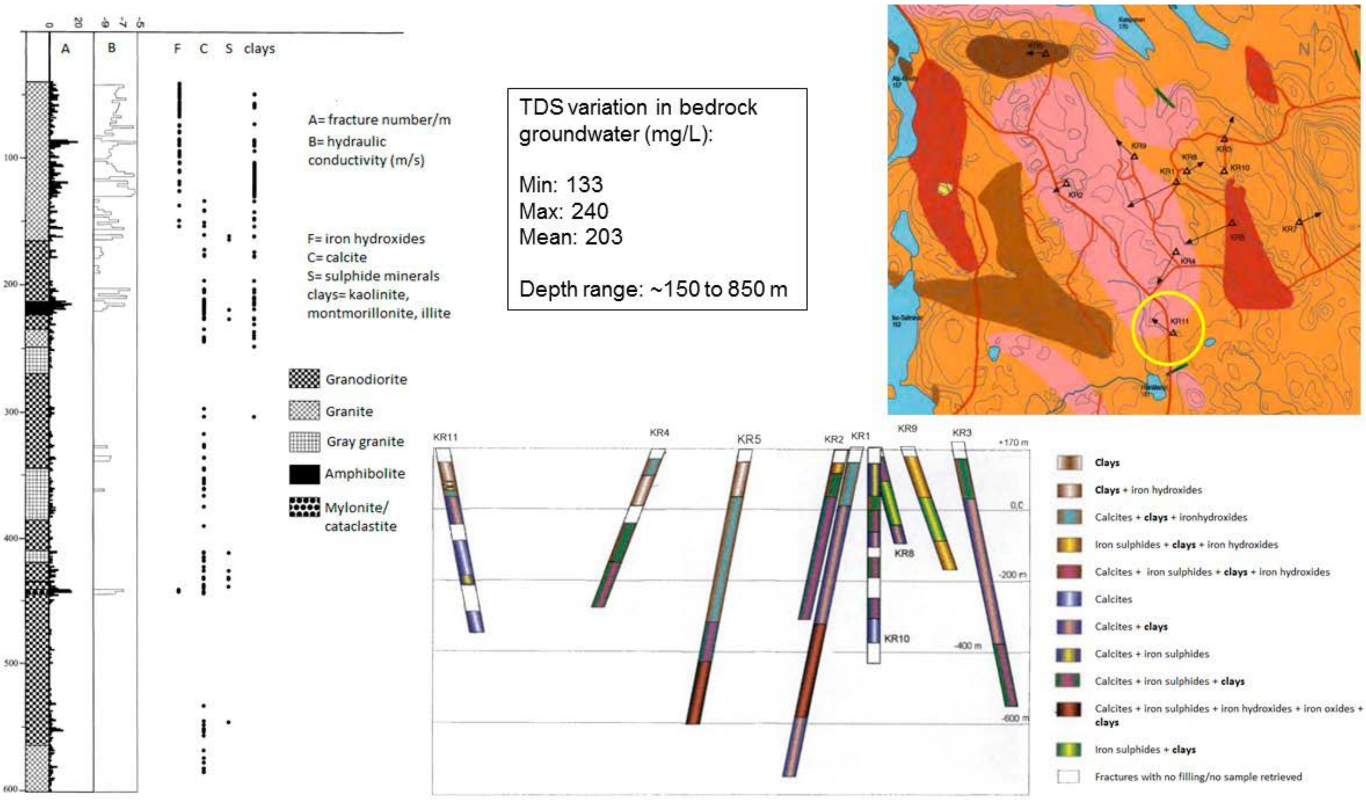


Fig. 3. Fracture mineral study results for Kivetty drillhole (KI-KR11) showing clay observations in relation to hydraulically conductive features of the site (left). On the right, the locations of Kivetty drillholes and the clay minerals observed are shown (Gehör et al. 1997a). TDS from groundwater samples is given based on the site characterization data of McEwen & Äikäs (2000).

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MINIMAL ALTERATION OF MONTMORILLONITE FOLLOWING LONG-TERM REACTION IN NATURAL ALKALI SOLUTIONS: IMPLICATIONS FOR GEOLOGICAL DISPOSAL OF RADIOACTIVE WASTE

by

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Bentonite is one of the more safety-critical components of the engineered barrier system in the disposal concepts developed for many types of radioactive waste. Bentonite is utilised because of its favourable properties, which include plasticity, swelling capacity, colloid filtration, low hydraulic conductivity, high retardation of key radionuclides and stability in geological environments of relevance to waste disposal. However, bentonite is unstable under the highly alkaline conditions induced by Ordinary Portland Cement (OPC: initial porewater pH>13) and this has driven interest in using low alkali cements (initial porewater pH9-11) as an alternative to OPC. To build a robust safety case for a repository for radioactive wastes, it is important to have supporting natural analogue data (see Alexander et al., 2015 for discussion) to confirm understanding of the likely long-term performance of bentonite in these lower alkali conditions.

Recently, therefore, there have been extensive efforts to better understand the interactions of alkaline fluids with bentonite (e.g. Savage et al. 2010, Sidborn et al. 2014), coupled with studies aimed at reducing the potential risk by development (cf. NDA 2010) and testing (e.g. Vuorinen et al. 2005) of low alkali cement formulations which typically have leachates with a pH of 9-11. Attempts to examine potential reaction of bentonite at these lower pH values have been complicated by the inherently slow kinetics of such reactions (e.g. Heikola et al. 2013). Clearly, this is an area where studying natural systems could play a valuable role – bridging the disparity in realism in temporal and spatial scales between laboratory studies and the systems represented in repository performance assessment (see discussion in Alexander et al. 1998, 2014, 2015, Miller et al. 2000). Indeed, in this case, the particularly slow kinetics of bentonite reaction in low alkali cement porewaters suggests natural system studies would appear to be the only viable method of assessing bentonite reaction within a timescale which would allow input to current repository safety cases.

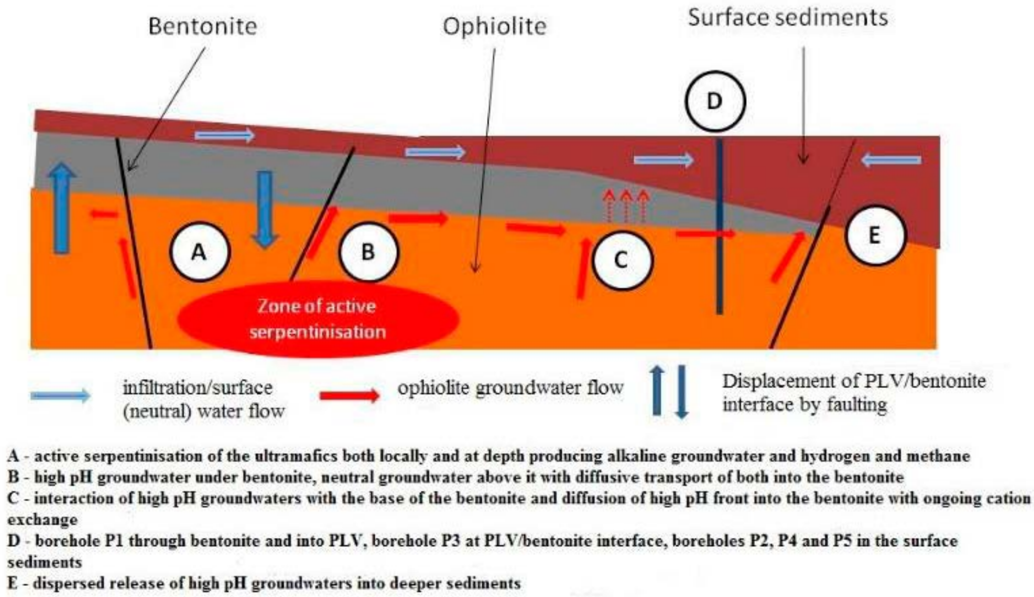

Fig. 1. Conceptual model of groundwater flow in the Troodos ophiolite. The ophiolite- derived, natural alkali (high pH) groundwaters are in contact with the base of the bentonite, while surficial (neutral pH) groundwaters flow across the top (Alexander & Milodowski, 2015).

In Cyprus, the presence of natural bentonite in association with natural alkaline groundwater (Fig. 1) permits the zones of potential bentonite/alkaline water reaction to be studied as an analogy of the potential reaction between low alkali cement leachates and the bentonite buffer in the repository. Here, the results (Milodowski et al. 2015) indicate that a cation diffusion front has moved some metres into the bentonite, whereas the bentonite reaction front is restricted to a few millimetres into the clay. This reaction front shows minimal volumetric reaction of the bentonite, with the production of a palygorskite secondary phase following the reaction of the primary smectites over time periods of 10^5-10^6 a.

The physical and temporal similarities between the natural bentonite/ophiolite groundwater environment studied in Cyprus and that expected for processed bentonite in a repository exposed to low alkali cement leachates argues strongly for limited reaction in the repository environment. That Mg-silicate reaction phases have not been considered in previous waste disposal studies is simply a reflection of the limited range of repository EBS designs and host rock types examined to date. In any case, the precise nature of the secondary phase may well prove less important to the long-term performance of the repository than the limited extent of alteration of the bentonite, something which points to an EBS that is robust enough to survive for the timescales of concern to the repository safety assessment.

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EFFECTS OF A LONGER DRY PERIOD ON BENTONITE COMPOSITION AND PERFORMANCE

by

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RATIONALE

In PA exercises of KBS-3 repositories, it is assumed that the bentonite barrier will be saturated a few years after closure of the repository.

This is definitively true for the average behaviour of the clay barriers, but there is a possibility that some portions of the repository with lower flow fields will remain unsaturated for longer periods.

In this event, the unsaturated bentonite buffer will be exposed to a heat transient for longer periods under dry conditions.

Under these conditions, it is not known to which extent the heat transient will impose mineralogical changes that will alter the hydraulic conditions of the buffer and to which extent these changes are reversible once the buffer is resaturated.

In this context, it would be very interesting to investigate these processes in the temperature and time regimes expected in the repository. These conditions can only be attained in natural formations of bentonite, which have been exposed to heat transients.

In the late 1990s, Amphos 21 (at that time Enviros) was deeply involved in the Natural Analogue programme of Enresa. Among the sites investigated were the bentonite deposits of Cabo de Gata (Almeria) in southeastst Spain, where the clay deposits have originated in a post-volcanic environment. At that site, there are a number of locations where bentonite has been exposed to a temperature transient around 120°C, and therefore there is a possibility to study the mineral alterations that had occurred and more importantly the reversibility of these alterations once the bentonite became saturated.

BENTONITE DEPOSITS FROM CABO DE GATA (SE SPAIN)

Cabo de Gata is a volcanic region, whose activity was related to the extensional tectonic phases in the Alboran Sea during the Serravallian-Lower Tortonian (14-10 Ma) and the upper Tortonian (9-7.5 Ma) periods. Volcanic rocks outcrop in a narrow band following a NE-SW trend from Cabo de Gata to the south of Murcia. Volcanic activity started with explosive phases that produced the formation of pyroclastic rocks and the formation of large calderas (sometimes over 5 km in diameter), followed by the extrusion of lava flows and subvolcanic dome intrusions. Bentonites were formed by the alteration of pyroclastic rock, but only in the areas where these rocks are crosscut by fractures. This seems to indicate that fluids flowing through these fractures drove the alteration process.

Carbonate rocks with abundant marine fossils frequently intercalate the different volcanic deposits, which, together with the presence of hydromagmatic volcanic facies, indicates that much of the volcanic activity took place in shallow or coastal submarine conditions. However, the presence of Upper Tortonian - Lower Messinian palaeosoils in the northern part of Cabo de Gata confirms that, at least during this period, the volcanic rocks were emerged.

More than 30 bentonite outcrops have been described in the Cabo de Gata area. All of them have a similar composition. The relative homogeneity of the bentonites in this area depends not only on the type of rock undergoing alteration, but also on the chemical composition and temperature of the hydrothermal solution.

The bentonite deposit of Morrón de Mateo is located in the Central Sector of the Cabo de Gata volcanic field, in the relatively depressed area of Los Escullos. The rocks of the area (pyroclastic rocks, limestones and bentonite) were intruded by a sub-volcanic dome, which caused a contact metamorphism (mineralogical changes due to the development of a high temperature aureole) in these rocks. This effect is clearly identified in the limestones sandwiched between volcanic tuffs partially transformed into bentonites (Fig. 1). This outcrop has been studied by ENRESA in the frame of their natural analogue programme (Arcos et al. 2001, Pérez del Villar et al. 2005). The main conclusions of these studies indicate that these bentonites were affected by a thermal event associated with the volcanic intrusion and some changes were induced to the bentonite.

We will discuss the potentiality of a more comprehensive investigation of this site in order to qualify the changes induced in bentonite in a long thermal period and the potential reversibility of the alterations once saturated conditions are restored.

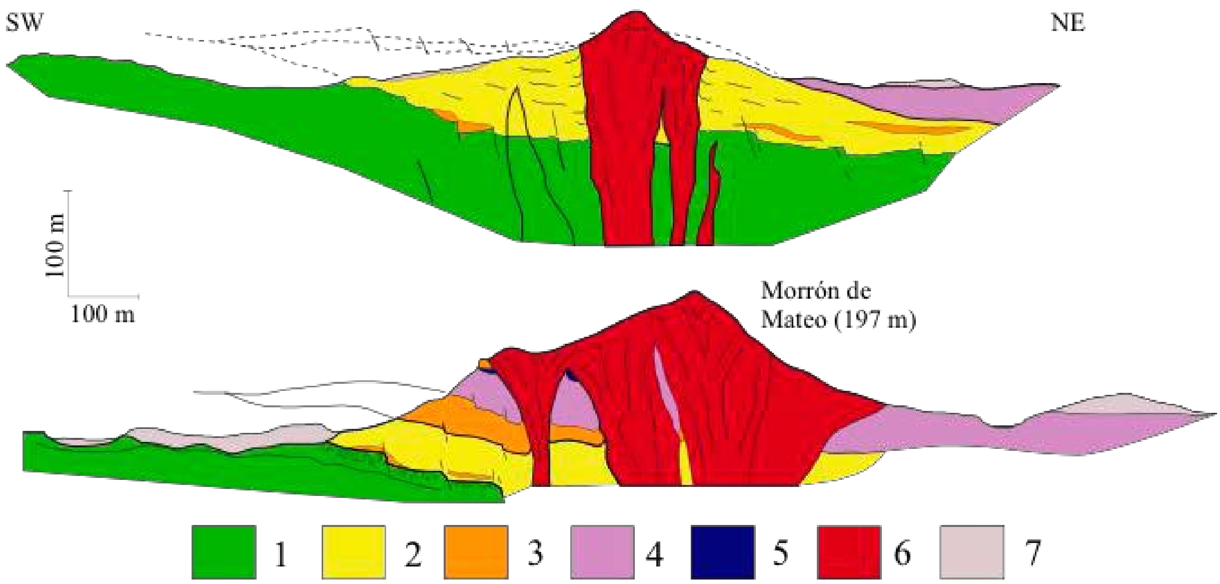

Figure 1. Geologic cross-sections of the Morrón de Mateo. 1. Andesites; 2. Layered tuffs; 3. Limestones; 4. Grey tuffs; 5. White tuffs; 6. Morrón de Mateo dacite dome; 7. Alluvial. Grey and white tuffs were locally transformed in bentonite prior to the Morrón de Mateo intrusion.

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PRELIMINARY STUDY ON KOREAN BENTONITE TO PROVIDE THE BASIS FOR USE OF NATURAL ANALOGUES IN SUPPORTING SAFETY CASES FOR RADWASTE DISPOSAL

by

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To validate the performance of buffer materials for high-level radioactive waste disposal in Korea, understanding of the properties of domestic buffer materials is a prerequisite. Bentonites are to be used as buffer materials in a geological repository designed to host high-level radioactive wastes. Under the repository conditions, the long-term behavior of bentonites in the engineered barrier system is of crucial importance in safety assessment on a geological timescale as well as in safety cases. An approach based on a natural analogue study can provide insights into the longevity of the bentonites. As a preliminary study on Korean bentonite natural analogues, we have investigated the status of domestic bentonite studies (Noh 2002a, Noh 2002b). Furthermore, the fundamental properties of the bentonite around the Gampo area in Kyungju, which is in the southeastern part of South Korea, have been examined. Figure 1 shows the sampling sites around the Gampo area and the XRD results from the bentonite samples.

The mineralogical compositions of the Gampo bentonite samples from four different mines were analyzed and compared. The major components include smectite, quartz, feldspar and mica. Bentonite or the samples with a higher bentonite content mainly show smectite. The surrounding rocks are mostly composed of quartz and/or feldspar. Some bentonites (e.g., G57-1, G57-2) show a higher feldspar content, while samples with a low bentonite content contain higher amounts of mica. SEM results match well with those from XRD. The samples with a higher bentonite content show the characteristics of smectite, i.e. a layered structure. We note that significant amounts of Ca and Fe were observed in the samples with a higher bentonite content. This is a characteristic of Korean bentonite, which shows a predominance of Ca-bentonite in the ore type instead of Na-bentonite. The chemical compositions of the Gampo bentonite samples were investigated using XRF. Briefly, the samples with a higher bentonite content have higher concentrations of Mg, Ca, and H_2O and a lower content of K and Na. The Gampo bentonite samples possess slightly different properties. The cation

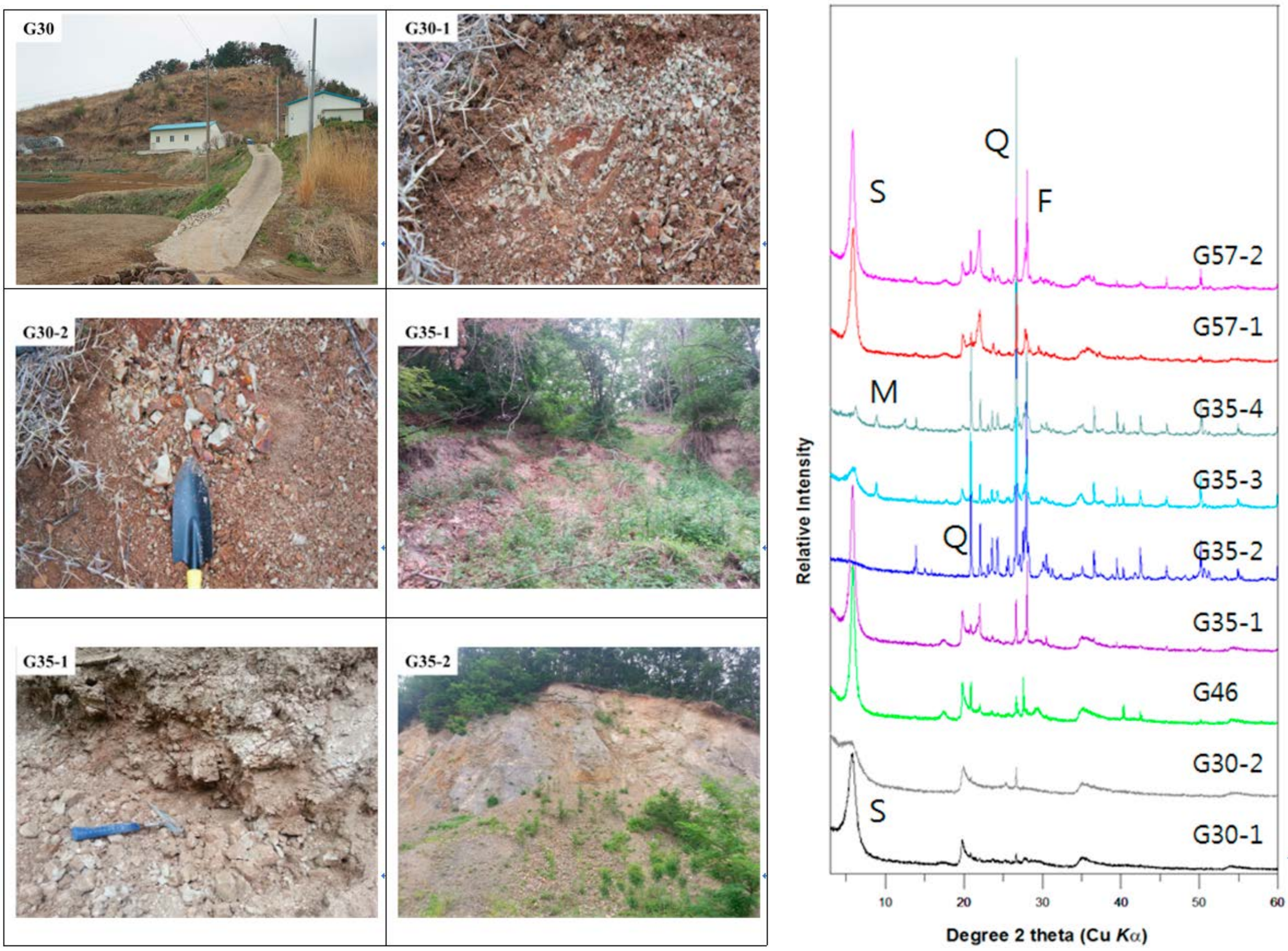

Fig. 1. Bentonite samples around the Gampo area in Kyungju (left) and their XRD results (right).

exchange capacity of the samples increases as a function of the bentonite content. We note that interaction between bentonite and the surrounding rocks was also observed.

A potential natural analogue study site for Korean bentonite was determined to be the newer bentonite mines around the Gampo area, with consideration of domestic bentonite mine development. A natural analogue study on domestic bentonite could be used as a supplementary safety indicator to support disposal safety.

In supporting the Korean safety cases for radioactive waste disposal, summarizing the status of natural analogue studies and evaluating the applicability of existing information relevant to the disposal is of importance. To assess the applicability of information from natural analogues to the overall evaluation of the safety cases, we have collected, classified, and evaluated data on natural analogues relevant to safety cases for the geological disposal of radioactive wastes (Baik et al. 2015). The natural analogue data collected were classified into categories including the site information, components/processes of the disposal system, properties/phenomena, reference, safety case application, application method, and suitability to a safety case. Figure 2 presents a sample of the natural analogues information database developed using an Excel-based spreadsheet. The information is categorized into the geological formation, system, component or processes, properties or phenomena, basic information, natural analogue

NO	Geological formation	NA Site	Nationality	System	Component Process	Properties Phenomena	Basic Information	NA information	Reference	Safety case	Application [1)]	Suitability [2)]
1	Sandstone, Schist	Alligator River (Koongarra U deposit)	Australia	NBS	Elemental retardation	Transport and retardation within fractured crystalline rocks	istribution of U in minera	• The majority of U in samples from the secondary ore body was contained in crystalline iron minerals (42-60%)	Yanase et al., 1991		UI	1
2	Sandstone, Schist	Alligator River (Koongarra U deposit)	Australia	NBS	Elemental retardation	Transport and retardation within fractured crystalline rocks	Diffusivity	• In unaltered quartz chlorite schist, uranium was mainly uraninite (UO2), which occurred in zones, fissure coatings and vein lets. The apparent diffusivity of uranium into chloritic schist from fissures was estimated to be about 1E-18 m^2/s.	Edghill, 1991		ISP, MV	1
3	Granite	Archaen, Lad du Bonnet	Canada	NBS	Matrix diffusion	Depth and volume of interconnected porosity	Diffusion depth	• There exists adjacent to fractures a zone a limited width (a few centimeters) to which the exchange of radionuclides between the water in the fracture and the rock itself may be restricted.	Gascoyne and Cramer, 1987; Ivanovich et al., 1987;		UI	1
4	Sediment	Askola mine	Finland	EBS	Spent nuclear fuel	Radionuclide retardation by secondary alteration products	eralogy and precipitation	• Electron microscopy studies show that precipitated uranium occupies intra-granular fractures in feldspars and quartz. In addition, secondary uranium was found to be distributed within goethite nodules as well as around the margins of iron-containing minerals in the form of silicate and phosphate precipitates. • Equilibrium modeling calculations predict that uranium would be precipitated as uranyl silicates, most likely soddyite and uranophane, in the prevailing chemical conditions beneath Lakeakallio hill. • modeling assumption: currently oxidizing saturated groundwater, weathering - U dissolution - leaching out from matrix - migration	Jokelainen et al., 2010		MV, UI	1
5	Granite	Aspo hard rock lab.	Sweden	NBS	Elemental retardation	Transport and retardation within fractured crystalline rocks	Fracture mineralogy	• Fracture coating phases are clay gouge, calcite and iron oxyhydroxides.	Landstrom & Tullborg, 1995		UI	1
	①	②	③	④	⑤	⑥	⑦	⑧	⑨	⑩	⑪	⑫

① Geological formation
② NA site
③ Nationality
④ System
⑤ Component / Process
⑥ Properties / Phenomena
⑦ Basic information
⑧ NA information
⑨ References
⑩ Safety case
⑪ Application
⑫ Suitability

Fig. 2. Information on natural analogues (NA) collected and classified using a spreadsheet with the categories considered (Baik et al. 2015).

information, natural analogue site, nationality, references, safety case, application, and suitability to the Korean case.

Natural analogue data were evaluated based on their suitability for the Korean safety case. Several experts in radioactive waste disposal participated in this evaluation process. Information on seventy-five natural analogues was categorized as suitability level 1 (SL-1), which is the data important in the Korean safety case and having high applicability. The information derived herein will provide further insights and perspectives in natural analogue studies for radioactive waste disposal in Korea.

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ALKALINE ALTERATION PROCESSES OF BENTONIC SEDIMENT IN THE PHILIPPINES NATURAL ANALOGUE AND PERSPECTIVE ON THE SURVEY OF THE ACTIVE TYPE

by

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INTRODUCTION

Ophiolites have been identified as sources of hyperalkaline groundwaters that can be considered as natural analogues of the leachates produced by cementitious materials in repositories for radioactive waste of various types (e.g. TRU and HLW). Saile mine and Bigbiga in the Philippines are suitable survey sites for studying natural analogues of bentonite–cement interaction, because bentonic sediments exist close to Zambales ophiolite (Fig. 1).

In this study, we have shown the alkaline alteration processes of bentonic sediment using the natural analogues in both of the areas. This research was initiated within a project to develop an integrated Natural Analogue Program in Japan (FY2013~2014), which was funded by the Japanese Ministry of Economy, Trade and Industry (METI).

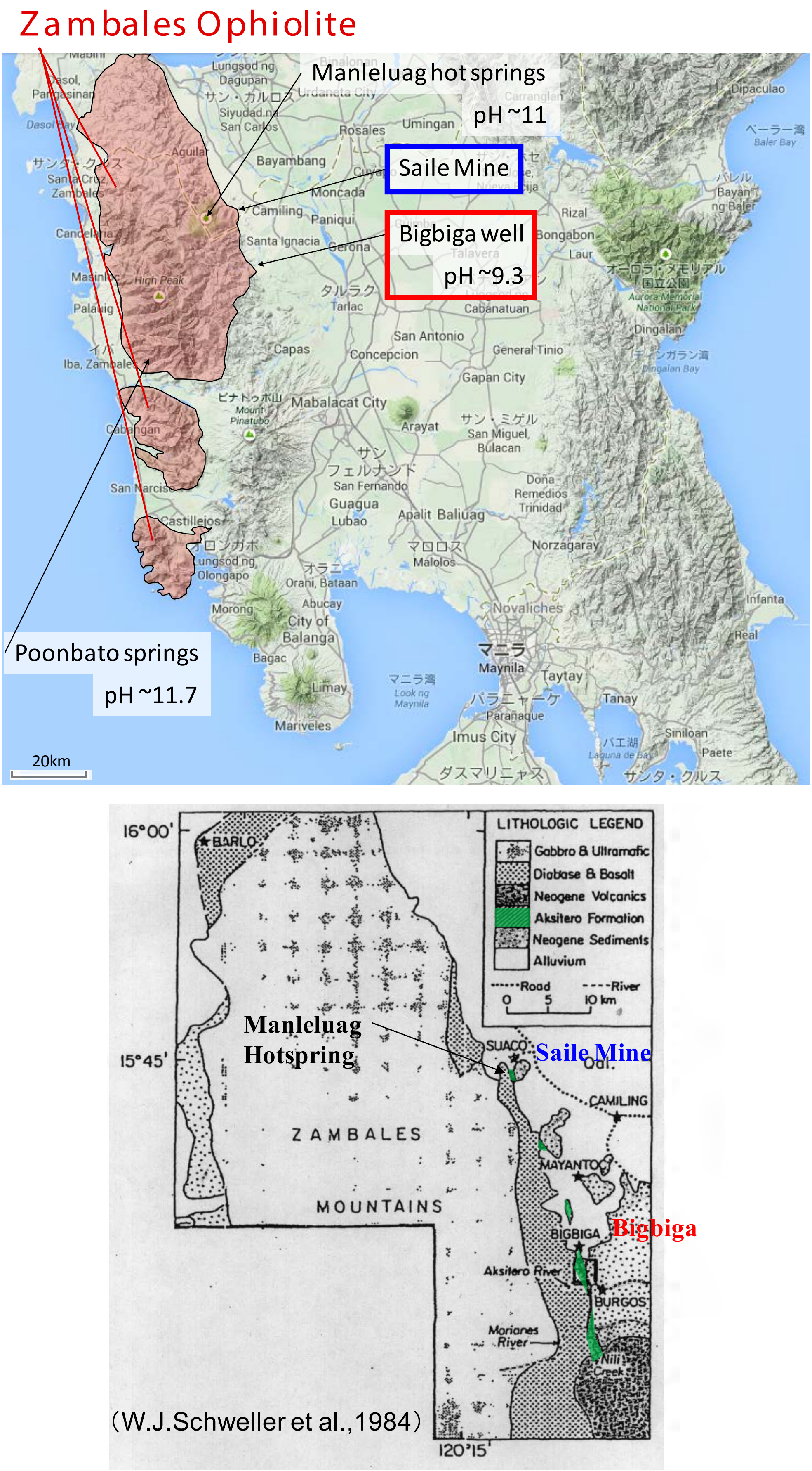

Fig.1. Location map of survey sites and distribution map of the Aksitero formation, Northwest Luzon.

NATURAL ANALOGUE OF THE FOSSIL TYPE

Saile bentonite mine is located in the west-central portion of Luzon Island, close to the Zambales ophiolite. The hyperalkaline groundwater near the mine, in Maleluag hot springs, is generated and mainly evolves by low-temperature serpentinization in the ophiolite, and it flows along faults. The mine can be considered as a natural analogue of past bentonite–cement interaction, because the bentonite was in contact with alkaline groundwater (i.e. no high pH groundwater is currently present in the mine). For this reason, the mine is referred to as a natural analogue of the "fossil type".

The alkaline alteration zone at the contact interface has a width of about 5 mm. It is constituted of Fe-smectite/saponite, K-feldspar, Ca-zeolite and silica minerals in the cation exchange reaction of hyperalkaline groundwater and dissolution-precipitation reaction (Fig. 2). The nontronite and goethite (paragenesis mineral) has formed an iron accumulation band with a density that is higher than the altered bentonite zone and the non-altered bentonite zone. The alkaline alteration zone is limited to the mm scale as a result of clogging of the iron accumulation zone.

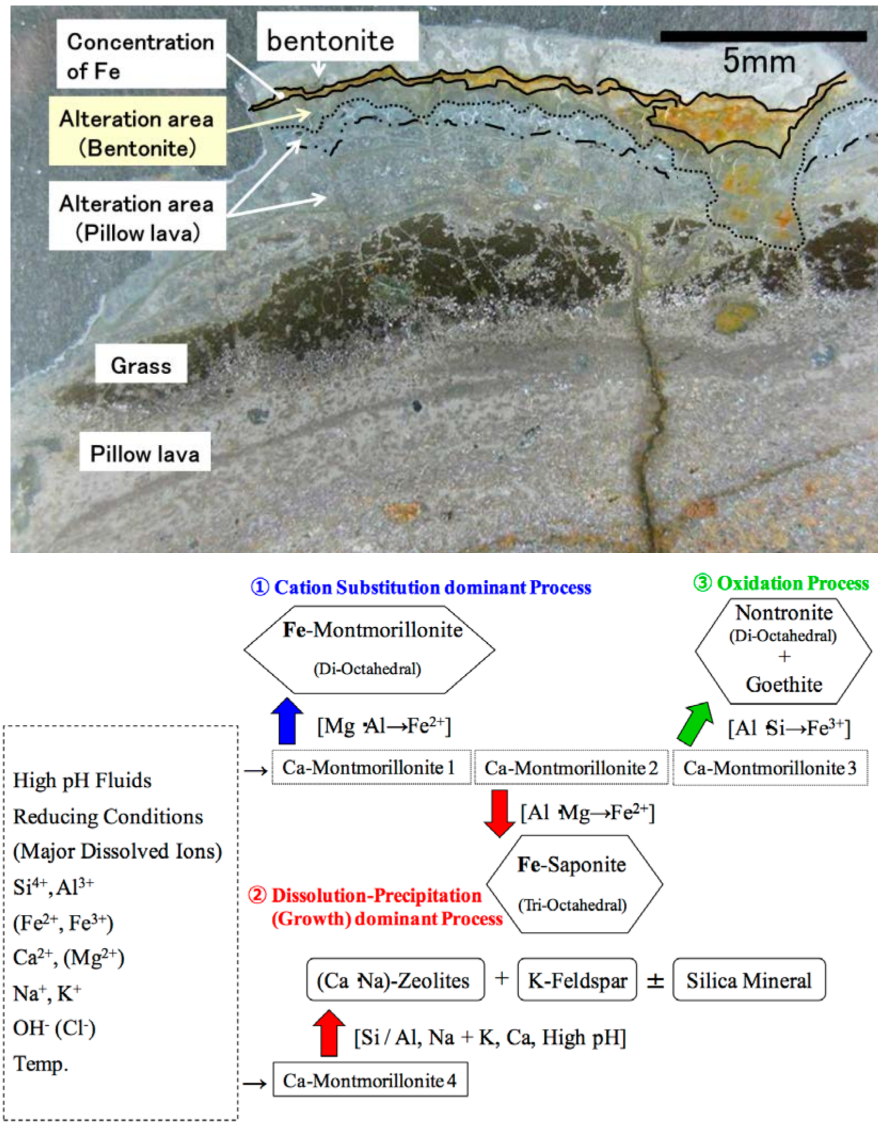

Fig. 2. Mineralogical evolution under hyperalkaline groundwater in the Saile Natural Analogue of the "fossil type".

NATURAL ANALOGUE OF THE ACTIVE TYPE

Bigbiga is located at the eastern end of Zambales ophiolite and 30 km from the Saile mine to the south. The alkaline groundwater (pH ~9.5) in Bigbiga is fissure water in a flowing fracture system developed in micro-gabbro/diabase of Zambales ophiolite, and also flows along the fracture (Slickenside) in the Aksitero Formation contacting with Zambales opiolite (Basement). Consequently, Bigbiga is an NA site of the "active type", where the alkaline groundwater has been interacting with pale brown limy-tuffaceous sandstone/mudstone (bentonic clay) for a long time (Fig. 3).

The alkaline groundwater flows into bentonic sediments, but the pH is up to 9.5, as shown in Figure 3. In laboratory experiments, alkaline alteration has not been observed at pH 9.5. However, from microscopic observations of the borehole core (Fig. 4), alkaline alteration of smectite in bentonic sediments was clearly observed. That is, alkaline alteration of bentonite arises by long-term interaction under low alkaline conditions. The secondary minerals include Ca-zeolite and K-feldspar, and are almost the same as the NA of the Saile mine.

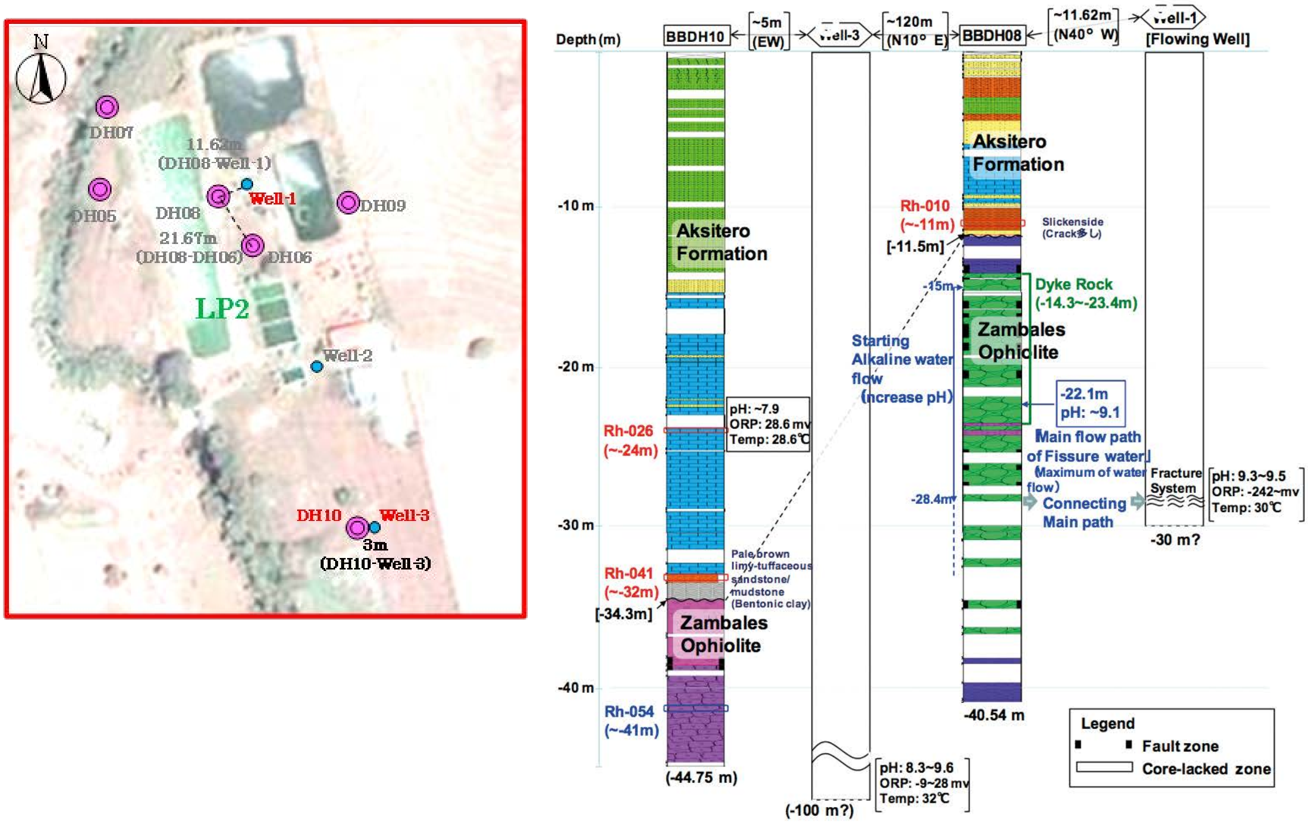

Fig. 3. Location of boreholes and conceptual alkaline groundwater flow in Bigbiga.

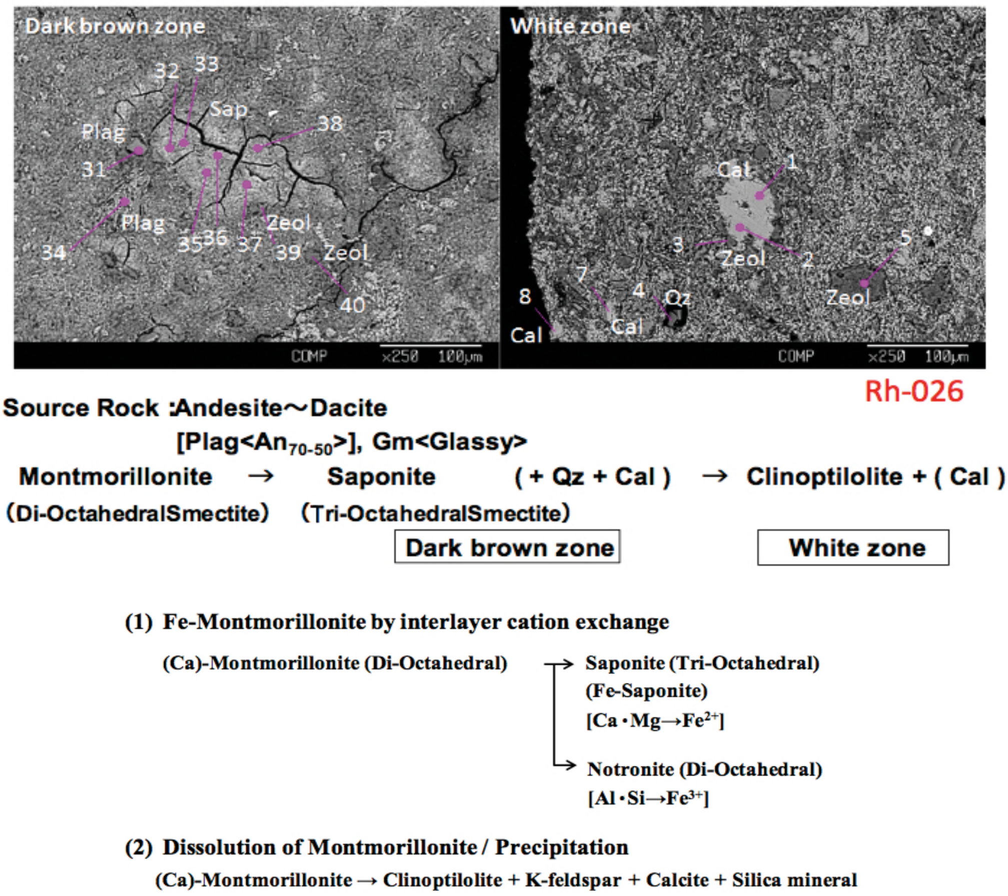

Fig. 4. Alkaline alteration process under low alkaline groundwater in the Bigbiga Natural Analogue.

CONCLUSION

Alkaline alteration of smectite in bentonic sediments at the Bigbiga NA site of the “active type” was observed by EPMA and FE-SEM analysis. The assemblage and occurrence (texture) of alkaline alteration minerals is analogous to that of the NA at Saile mine, which is of the “fossil type”. This NA shows alkaline alteration of bentonite that has arisen through long-term interaction under low alkaline (pH ~9.5) conditions.

Bigbiga is inadequate as an NA site of the "active type" in respect of the following: (1) the pH is low and (2) it is difficult to understand the scale of the alteration, because the contact is too deep (10 to 30 m depth). For this reason, we expect Palawan, where springwater with a pH of over 11 was found at two or more locations, to be a new NA site of the “active type”.

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THERMAL ALTERATION OF BENTONITE (TAB): ISN'T IT TIME WE GOT IT RIGHT?

by

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INTRODUCTION

In the case of thermal alteration of bentonite, cementation can occur due to smectite/illite conversion, where silica is released at higher temperatures (and later re-precipitated) (Leupin 2014). Currently, in many national programmes, the requirements for maximum bentonite temperatures are set around 100 °C (e.g. Posiva 2012). In many cases, there would be interest in increasing this limit, thereby allowing, for example, the placement of waste packages more closely than is currently planned. However, this would require more stringent justification than exists at the moment.

Although Miller et al. (2000, 2006) and Laine & Karttunen (2010) noted a significant number of natural analogue (NA) papers on this theme in the literature, recent re-analyses (Posiva 2013, Reijonen & Alexander 2015) shows that most examples suffer from the same problem: the conditions are not truly representative of a repository. In most cases of contact metamorphism studied, the temperatures have been much too high, around 800–900 °C rather than the repository-relevant maximum of 100 °C in the current Finnish design (Posiva 2013), for example. As illustrated in Figure 1, the degree of saturation of the EBS affects the thermal conductivity of the bentonite, the highest temperatures being met in the "dry" case, i.e. with bentonite water contents being as installed. In Posiva's reference design, the initial water content of the buffer is assumed to be 17% (Posiva 2012).

THERMAL-INDUCED CEMENTATION: SOME EXAMPLES

Several NA studies (e.g. Pellegrini et al. 2002, Woods et al. 2000) have tried to address the potential impact of thermally-induced cementation. Usually, smectite-bearing clays which had been penetrated by dykes/sills were examined and, at distance 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

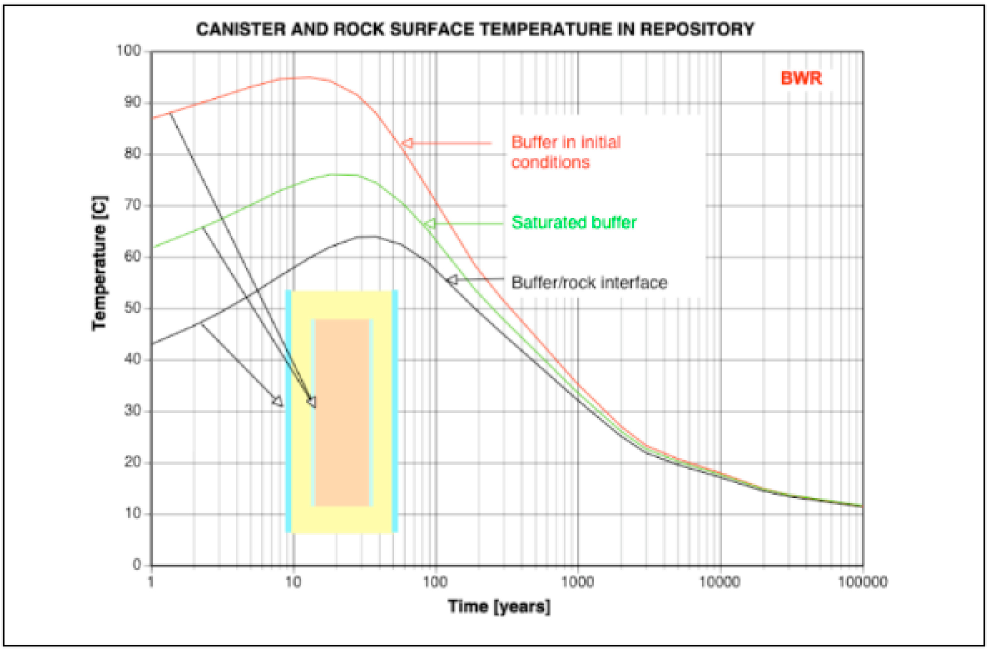

Fig. 1. An example of the likely temperature evolution in the bentonite buffer (for BWR fuel in a centrally located deposition hole in Olkiluoto; Ikonen & Raiko 2012).

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 the safety case, because the full thermal, chemical and pressure histories experienced by these clays were not adequately characterised.

Pusch & Karnland (1988) investigated bentonite from the Busachi site in Sardinia and ran a series of calculations to assess the likely temperature at different places in the bentonite. 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 in most designs), 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 that might adversely affect the containment of radionuclides if it occurred in a repository environment.

However, the calculations were based on a range of assumptions about the site and the bentonite, and these need to be ground-truthed before the results can be used quantitatively in a safety case (cf. discussion in Alexander et al. 2014, Reijonen et al. 2015).

Kolaříková & Hanus (2008) reported on the Ishrini bentonite body in Libya, where the impact of a basalt dome and dykes on the bentonite was assessed by examining isotopic and mineralogical changes. It would appear that the minimum temperature experienced by the bentonite was probably >190°C, but bentonite from outwith this zone (i.e. in the lower temperature area) was not sampled (Laine & Karttunen 2010).

In conclusion, while the probable impact of raised temperatures is minimal (i.e. some local cementation occurs, but the majority of the bentonite remains unaltered), a truly repository-representative analogy has not yet been examined.

THERMAL-INDUCED ILLITISATION: SOME EXAMPLES

In the case of diagenetic illitisation, a number of NA studies have been carried out in the Gulf of Mexico (e.g. Robertson & Lahann 1981), and elsewhere (Pusch & Karnland 1988). These studies suggest the illitisation rate in the natural environment is considerably slower than that predicted by kinetic models, but this is due to the fact that the process depends on the rate of supply of potassium, which may often be limited. This will likely be the case at Olkiluoto, but was shown not to be the case for a potential repository in the north Switzerland crystalline basement (Alexander & McKinley 1999).

However, these studies do not really represent the repository environment because the duration of heating is several orders of magnitude longer than would be the case in most repository designs (cf. Fig. 1).

PROPOSED NEW STUDY

A two-pronged approach utilising URL and NA data (cf. Alexander et al. 1998) is proposed:

URL

- to date, bentonite saturation studies have been highly artificial, with induced groundwater flow
- what is required is an approach where re-saturation is studied under appropriate conditions (i.e. slow, natural re-saturation), only then can we be sure that the 'dry thermal pulse' is realistic

NA

- no more surface sites, time to look at bentonite mines where the bentonite is under appropriate hydrostatic loads
- and is fully saturated
- and fully characterise the material using modern analytical methods.

For example, FEBEX-DP has just dismantled the long-running Grimsel FEBEX experiment (Fig. 2), where the bentonite was allowed to re-saturate under natural conditions. The results of the analyses of the bentonites will provide invaluable

Fig. 2. Excavation of section 36 of the FEBEX bentonite in Grimsel in May, 2015 (image courtesy Nagra; www.grimsel.com).

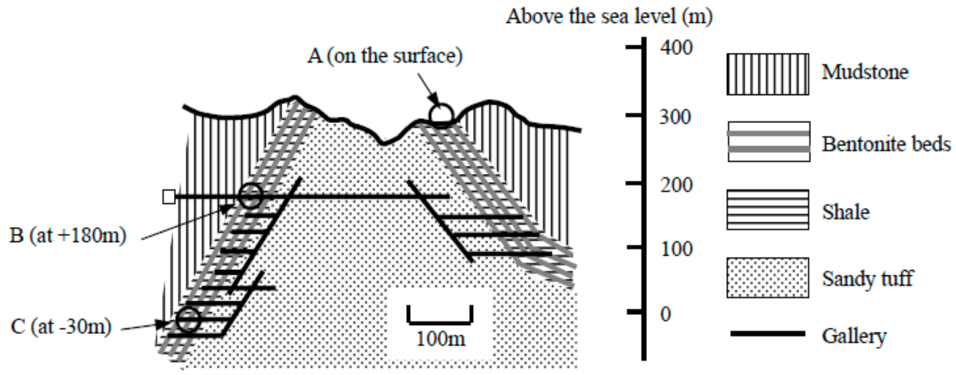

Fig. 3: Schematic cross-section through the Tsukinuno bentonite deposit in northern Japan (Kuno et al. 2002).

data for comparison and combination with NA data from an appropriate site (e.g. the Tsukinuno bentonite deposit in northern Japan; Fig. 3), where bentonite under a hydrostatic load has experienced heating from a local dyke. This will allow, for the first time, a realistic assessment of the likely impact of thermal alteration of the buffer bentonite in a repository for spent fuel and vitrified high-level waste.

CONCLUSIONS

NA studies of thermal alteration of bentonite are legion (and only a few are noted above), but two critical reviews (Laine & Karttunen 2010, Reijonen & Alexander 2015) of the work carried out to date indicate that they are of little value in supporting the safety case. This is because either the conditions are not repository relevant or the information obtained is limited due to:

- the inappropriate temperature range examined (e.g. Ishrini)
- inappropriate timescales for heating of the bentonite, they are usually way too long (e.g. Gulf of Mexico)
- ill-defined (e.g. dyke/sill studies) or assumed (e.g. Busachi) boundary conditions
- under-characterised physico-chemical parameters of the bentonite (the majority of studies to date)
  - use of modern analytical techniques can better define temperature zonation before assessing changes in the smectite (cf. comments in Posiva 2013)
  - define more than just the smectite content (e.g. swelling pressure, saturation state etc.; cf. Alexander & Milodowski 2015, Milodowski et al. 2015)

In addition, certain aspects have never been properly addressed, for example:

- as surface/near-surface sites are normally studied, bentonite under a repository-relevant lithostatic load/hydraulic pressure has not been examined
- again, as surface/near-surface sites are normally studied, fully saturated bentonite has never been examined

Suitable sites have been identified for a new project on the thermal alteration of bentonite (TAB), and these are discussed above. Now, with several new repository safety cases about to be launched, isn't it time we got it right?

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LONG-TERM DURABILITY OF SHAFT SEALING MATERIALS UNDER HIGHLY SALINE GROUNDWATER CONDITIONS

by

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The Nuclear Waste Management Organization is responsible for the implementation of Adaptive Phased Management (APM), the federally-approved plan for safe long-term management of Canada's used nuclear fuel. Under the APM plan, used nuclear fuel will ultimately be placed within a deep geological repository in a suitable rock formation. Both crystalline and sedimentary rock formations are being considered.

The materials proposed for sealing the repository shafts are a bentonite-sand mix, a low-heat, low-pH concrete, and a bitumen-sand-lime mix. A particular requirement in the case of the sedimentary rock formations in the Michigan Basin in Southern Ontario is to consider the high salinity of the porewater. The porewater chemistry within this geosphere is expected to be essentially Na-Ca-Cl brine at 260–370 g/L and near-neutral pH at 10–25 °C.

Natural analogues are of interest with respect to supporting the durability of these materials under saline low-temperature conditions. This is a summary of what may be the most appropriate natural analogues (studied to date) of the core materials: bentonite, concrete, and asphalt.

BENTONITE

The primary material for sealing the repository shafts is expected to be a compacted bentonite-sand mix. As the closed and sealed repository is slowly infiltrated by groundwater, the bentonite will swell and fill any remaining void spaces.

Under postclosure conditions, high salinity of the groundwater is expected to affect the swelling properties of the bentonite; however, it is not expected to alter the mineral stability of the bentonite. Alexander and Milodowski (2014) observed that the Perapedhi bentonites studied under the Cyprus Natural Analogue Project likely remained in a marine, saline environment until the formation of Cyprus and the initiation of fresh groundwater circulation. Figure 1 compares the swelling pressure of the Cyprus bentonite with a range of industrial bentonites, indicating that exposure of the Perapedhi bentonite to marine saline conditions for nearly 90 million years had no significant impact on its swelling capacity.

Bentonite, particularly the swelling clay component (smectite), is unstable under high-pH conditions. In the repository, concrete may produce alkaline conditions locally, affecting adjacent bentonite. However, as the low-heat concrete leachates will have a lower pH (10–11) than Ordinary Portland Cement, it is expected that any reaction with the bentonite will be local.

Significant degradation of smectite under alkaline groundwater has been observed in conditions with large amounts of alkaline groundwater; however, in natural analogue systems with limited groundwater flow, the reaction is limited. For example, results from the International Philippines Natural Analogue Project (Fujii et al. 2010) show that reaction with alkaline water (pH ~11) in the bentonite is restricted to the contact interface, with the maximum width of the reaction zone being 5 cm. In another example, in the Cyprus Natural Analogue Project (Alexander & Milodowski 2014), the groundwater (9 < pH < 11) appears to have been circulating under the bentonite for approximately 500,000 to 800,000 years. In this time, less than 1% of the smectite in the bentonite has reacted, indicating very slow reaction times. The authors note that sufficient swelling pressure remains in the reacted bentonite to minimize further reaction due to pore throat reduction (see also Wilson et al. 2011).

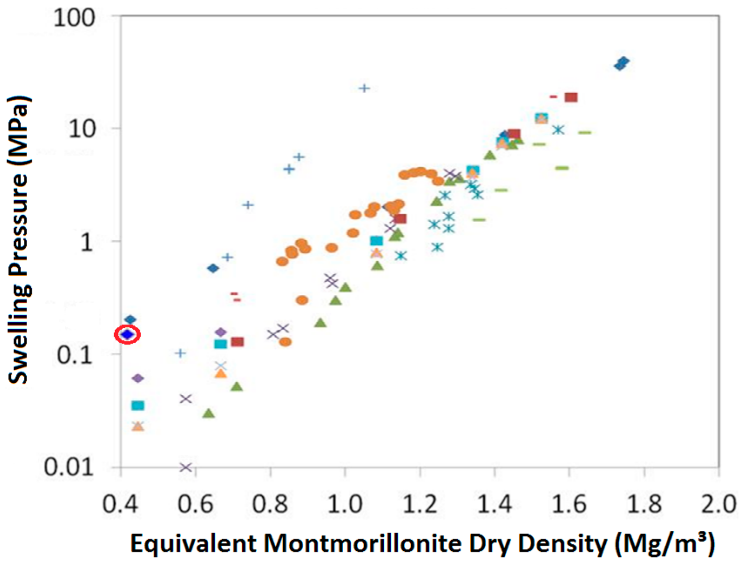

Fig. 1. Cyprus bentonite (marked with a red circle) compared with industrial bentonites.

CONCRETE

Low-heat concrete may be used to provide structural support and confinement to the column of shaft sealing materials. Analogue studies of natural cements suggest that, within tectonically stable systems, the material is durable, with the oldest reported cements at Maqarin in north Jordan being approximately 2 million years old (Alexander 1992). Milodowski et al. (1989) also reported the presence of unreacted natural cements from the Scawt Hill and Carneal Plug sites in Northern Ireland. These cements were produced during the thermal metamorphism of the host limestone and are estimated to be around 58 million years old. In both examples, the natural cements are effectively impermeable and remain unchanged until accessed by groundwaters (through tectonic damage, for example).

If damaged, the tendency is for these systems to reseal, either with secondary calcium silicate hydrate phases (Linklater 1998) or carbonates (Clark et al. 1994).

Of the natural analogue studies reported to date on cements, it is important to note that the natural cements examined are more akin to Ordinary Portland Cement, not low-heat cement (Gray & Shenton 1998). Low-heat cement is essentially the same as the pozzolanic cements developed by the Romans in the 3rd century BCE, or perhaps in Tiryns and Mycenae a millennium earlier (Middleton 1888). Recent studies of Roman cements exposed to marine salinities for about 2000 years suggest little degradation of the cement (Oleson et al. 2004, Vola et al. 2011).

ASPHALT

The shaft seal design concept includes a layer of asphalt as a redundant low-permeability seal which can make good contact with the host rock and provide an effective barrier to the movement of water immediately upon placement.

The Athabasca oil sands located in the McMurray Formation in Athabasca are the largest known reservoir of crude bitumen in the world. Longstaffe (1993) indicates that the bitumen has remained stable for over 10 million years under groundwater conditions that were likely brackish.

The bitumen deposits in sandstone rocks in the Uinta Basin in Utah are believed to be from the late Cretaceous to Eocene period, 70 to 30 million years ago (Schamel 2009), formed under basin waters that were likely comparable to marine salinity. Also within the Uinta Basin in Utah, extensive veins of another natural asphalt called gilsonite were formed by hydrothermal fluids during the Eocene period, 56 to 34 million years ago (Boden & Tripp 2012). Both examples suggest long-term stability of bitumen under likely saline conditions for several tens of millions of years.

Drake et al. (2006) reported natural asphalt (asphaltite) in open and closed fractures at the Forsmark site in Sweden. This asphalt was exposed to brines (45 g/L at present) for at least several million years, suggesting very long-term stability of asphalt under saline conditions.

The Dead Sea is representative of repository groundwater salinity, with values of up to 370 g/L. Large asphalt blocks (up to several hundred cubic metres) have frequently been found floating in the Dead Sea. The use of Dead Sea asphalt in the preservation of Egyptian mummies (Rullkötter & Nissenbaum 1988) stands testament to the durability of the material despite immersion in the Dead Sea brine.

POTENTIAL STUDIES OF BENTONITE DURABILITY UNDER HIGHLY SALINE CONDITIONS

Smectite samples from existing rock cores from the sedimentary formations in the Michigan Basin and field samples of smectite from the coast of the Dead Sea could be analysed for their physico-chemical properties to provide a better understanding of how these properties evolve with palaeohydrological conditions. Posiva Oy is completing work similar to this on cores from their site at Olkiluoto.

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RECENT SCIENTIFIC AND TECHNOLOGICAL ADVANCES IN SITE EVOLUTIONAL MODELLING IN THE JAPANESE URL PROGRAMME: NA INPUT

by

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Geo-stability has been of great concern to safety issues in the geological disposal of high-level radioactive wastes (HLW) in many countries, especially in neighbouring tectonic plates such as Japan. Generally, site evolution models (SEM) that are developed using past geological data on sites may play a central role in a safety case including geo-stability from the geological past to several thousand to hundreds of thousand years into the future.

JAEA (Japan Atomic Energy Agency) has developed/synthesised the SEM methodology utilizing data from two URLs (Mizunami as a crystalline rock type; Horonobe as a sedimentary rock type) collected in the past dozens years. The methodology consists of on-site palaeohydrogeological investigations and the development of integrative modelling methods combining geology, hydrogeology, hydrochemistry and site tectonics, including uplift and erosion processes.

Here, the current status of the SEM models will be presented and their practical applicability and the potential interaction with NA will be discussed.

URANIUM OCCURRENCES AS INDICATORS OF LONG-TERM GEOCHEMICAL STABILITY IN THE FINNISH BEDROCK

by

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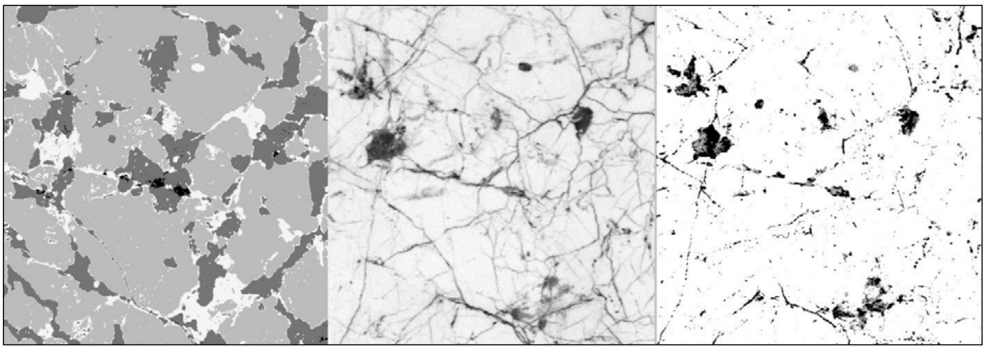
One of the major challenges in the field of geological disposal of spent nuclear fuel is to show confidence in the long-term safety of the repository system. For this, the conditions in the bedrock should remain stable and favorable to the long-term behavior of engineered barrier system and be easily re-established should any disturbance occur, such as the intrusion of dilute water during or after glacial periods. In our presentation, using the age of the findings of uranium minerals associated with the rock matrix structure, we can state with confidence that 1) oxygen penetrates the bedrock to a maximum of a few tens of meters, 2) dilute/



<u>*Petrographic map:*</u> *Chemical staining; light grey = K-feldspar, white = plagioclase, dark grey = quartz and black biotite*	<u>*Porosity map*</u> *C-14-PMMA autoradiography*	<u>*Uranium map*</u> *α-tracks on CR-39 polycarbonate film*

Fig 1. Mineral map of granite sample after manipulation with tresholding program (left); light grey = potassium feldspar; white = plagioclase; dark grey = quartz; black = biotite. Autoradiograph made from the rock surface after C-14 PMMA impregnation (in the middle). Alpha tracks on CR-39 polycarbonate film (right). Sample width 4.6 cm.

fresh water is also buffered within a few tens of meters and even within a few meters, 3) the structure of the rock matrix contributes to transport retardation by offering large reactive surfaces on one hand, and on the other hand by making the transport travel times longer (See Figure 1 below).

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USE OF SELF-ANALOGUES TO COMPLEMENT "SITE STABILITY" EVALUATION AND TO IDENTIFY STABILITY INDICATORS

by

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NUMO'S STAGED IDENTIFICATION OF A REPOSITORY SITE

Providing assurance of the long-term stability of the geological environment is a prerequisite for geological disposal. In Japan, particular concern has focused on the stability, as earthquakes and volcanic eruptions, for example, frequently occur in relation to tectonic evolution over geological timescales. A wide spectrum of natural events/processes is commonly perceived as having potentially disruptive impacts on the geological environment. The selection of a suitable site for a high-level radioactive waste repository is, therefore, one of the biggest challenges in Japan and should take into consideration the dynamic tectonic settings and their temporal and spatial (or 4D) evolution.

The Nuclear Waste Management Organization of Japan (NUMO) initiated the siting process in 2002 with open solicitation of volunteer host municipalities for the identification of a suitable repository site in three stages: a literature survey (LS), preliminary investigation (PI) and detailed investigation (DI) as stipulated in the Act on Final Disposal of Specified Radioactive Waste. A logical and progressive basis for the siting process has been developed, which involves explicit exclusion criteria (NUMO 2004).

The initial LS stage involves the use of all available literature information to ensure that proposed or volunteering areas and their surroundings are not obviously likely to be directly disrupted by natural events/processes. The following PI stage concentrates on extensive surface-based investigations at the preliminary investigation areas (PIAs) that have been identified in the initial target areas. Sufficient information will be gathered on geological structures as well as thermal, hydrological, rock mechanical and hydrogeochemical (THMC) conditions to decide whether there is reasonable likelihood of the geological environments being stable for a long period of time. The detailed investigation areas (DIAs) that meet requirements such as the long-term persistence of favourable THMC conditions will then be identified in the PIAs. The final DI stage proceeds further with site characterisation that includes *in situ* investigations in underground facilities. On the basis of the DI results, the most suitable repository site where the long-term stability of the geological environment must be ensured will be finally identified.

ASSEMBLING OF NATIONWIDE GEOSCIENTIFIC INFORMATION

NUMO have now been assembling nationwide the latest geoscientific information of direct relevance to geological disposal and periodically updating their geoscientific database. State-of-the-art geoscientific knowledge clearly demonstrates that the favourable THMC conditions most likely prevail in deep geological environments in Japan (Table 1). A convincing indication for slow groundwater movements, for example, is the existence of deep groundwater with a very long residence time in some sedimentary formations (Table 2). Basic concepts and criteria for precluding the potentially significant impacts of natural disruptive events/processes on the geological environment are also demonstrated (Ota et al. 2015). Defining explicitly both the favourable THMC conditions and also the exclusion criteria is requisite for the staged identification of a suitable repository site.

Table 1. Prevalence of favourable THMC conditions in deep geological environments.

Conditions	Favourable characteristics/ properties required	Prevailing characteristics/properties identified
T Thermal	Low geothermal gradients for prolonged periods	Geothermal gradients in non-volcanic regions generally lie in the range of 3 °C to 5 °C per 100 m (NUMO 2013, Tanaka et al. 2004), which demonstrates that areas where target temperatures prevail at a depth greater than 300 m are widely distributed
H Hydrological	Slow groundwater movements due to low hydraulic gradients and/or low rock hydraulic conductivities	Values determined for hydraulic gradients in the order of 0.001 to 0.1 and hydraulic conductivities in the order of 10^{-12} to 10^{-6} m/sec (NUMO 2013) show that high hydraulic gradients are normally associated with low hydraulic conductivities and vice versa, which indicates the occurrence of slow groundwater movements
M Rock mechanical	Small rock deformation including rock creep	Rock mechanical datasets updated show that the ratios of uniaxial compressive strength to overburden pressure are generally in the range of 1 to 2 (NUMO 2013, Cho et al. 2009)
C Hydro-geochemical	Groundwater with near neutral pH, reducing conditions and concentrations of dissolved inorganic carbon (DIC) less than 0.5 mol/dm^3	The common occurrence of deep groundwater with pH values between 6 and 9, more negative Eh values and DIC concentrations of smaller than 0.1 mol/dm^3 (NUMO 2013, Oyama et al. 2011) demonstrates the prevalence of favourable hydrogeochemical conditions

Table 2. Groundwater ages.

Location	Host rock	Depth [mbgl]	Age [ky]	Methods	Reference
Horonobe	Miocene sedimentary rock	700	10,000	^{4}He, ^{36}Cl	Nakata & Hasegawa 2010
Yokosuka	Miocene sedimentary rock	210~500	7,000	^{4}He, ^{14}C, ^{36}Cl	Hasegawa et al. 2013
Kushiro	Cretaceous sedimentary rock	750	2,000	^{36}Cl	Mahara et al. 2006
Kobe	Pliocene sedimentary rock	1,500	230~25	^{4}He	Morikawa et al. 2005
Rokkasho	Miocene sedimentary rock	180	80~50	^{4}He	Nakata & Hasegawa 2011
Mizunami	Cretaceous granite	1,000	20~10	^{4}He, ^{14}C	Hasegawa et al. 2010

Natural disruptive event/processes to be taken into consideration here involve igneous activity, the occurrence at repository depths of non-volcanic thermal or deep-seated fluids, disruptive fault movement and significant uplift/erosion. With regard to igneous activity, for example, an exclusion area of 15 km radius is drawn around Quaternary volcanic centres in Japan (METI 2014). Among the key natural system evidence behind this criterion is that low pH (<4.8), high temperature fluids that would potentially perturb the favourable thermal (T) and hydrogeochemical (C) conditions generally occur within a 15 km radius circle around Quaternary volcanoes (Figs 1 and 2). In order to preclude the potentially significant impacts of tectonic-driven uplift/erosion, the amount and rate of future uplift/erosion will be quantified. Detailed nationwide information based on field observations allows the quantification to be made with confidence, which involves, in particular, the current uplift rate (Fig. 3) and the onset time of a constant velocity uplift rate (Fig. 4).

USE OF SELF-ANALOGUES

During the LS stage, information will initially be collected from the geoscientific database, relevant organisations and other available sources, all of which should specifically relate to natural events/processes and geological conditions of the target areas. However, at this stage (and possibly even at the early PI stage), site-specific information may be rather limited and hence there is assumed to be little or no information that directly ensures the long-term stability of the geological environment in the target area (or 'site stability'). It will thus be of help to use relevant supporting information that could complement the evaluation of the site stability (Fig. 5).

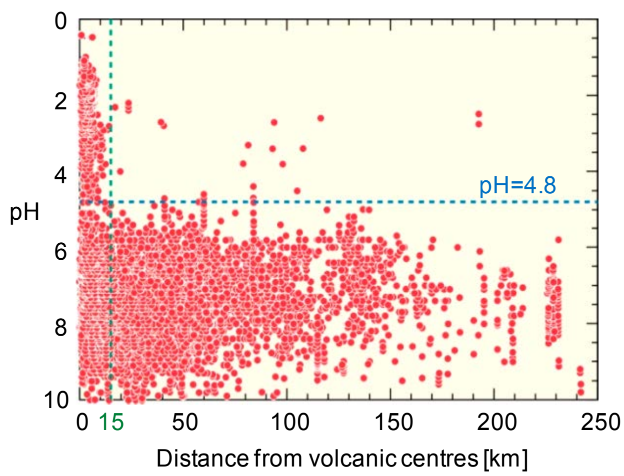

Fig. 1 Correlation between pH and distance from Quaternary volcanic centres (modified from Asamori et al. 2002).

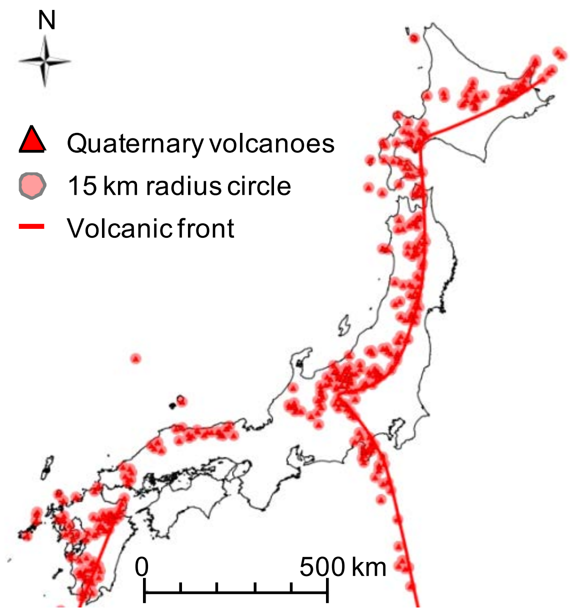

Fig. 2 Exclusion areas around Quaternary volcanoes (modified from AIST 2013).

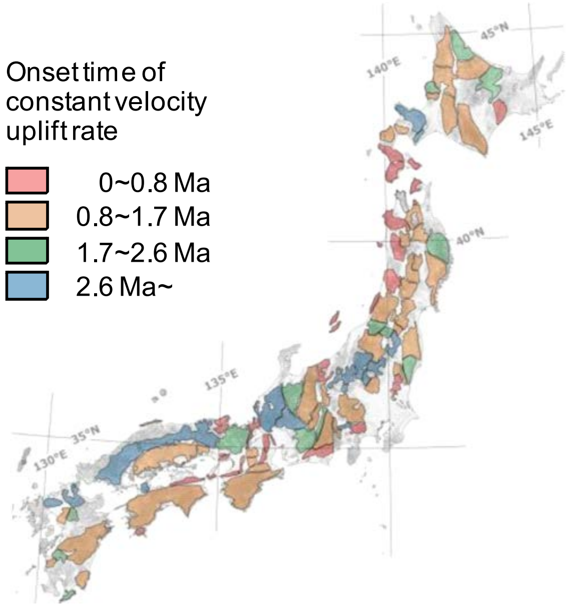

Fig. 3 Onset times of consistent mode and rate of uplift (Yasue *et al.* 2014).

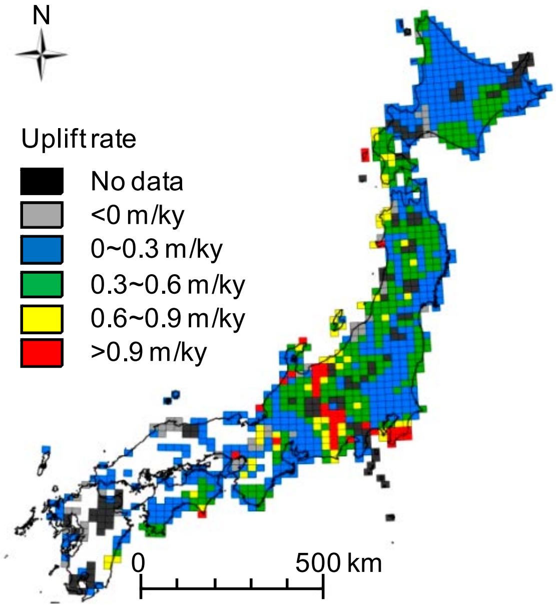

Fig. 4 Distribution of current uplift rates (after JGS 2011).

To this end, NUMO will use relevant self-analogue information that is derived from studies on similar rock types in similar geological settings. Here, it is critical that the entire analogue site (or perhaps even region) should be well supported by multiple lines of natural system evidence. A wide range of parameters are required to fully assess the site stability and, in Japan, are available in a couple of analogues sites; for example, Tono, central Japan, where uranium mineralisation occurs in the host sedimentary formation, and Horonobe, Hokkaido, Japan,

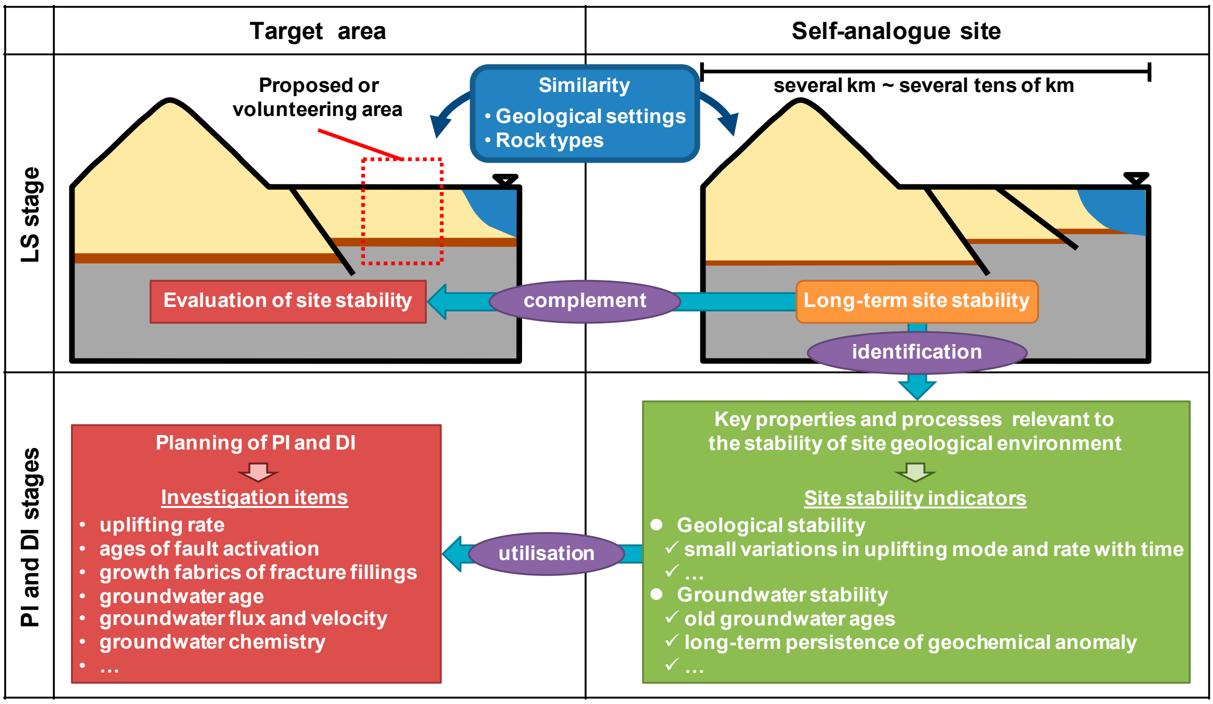

Fig. 5 Illustration of how self-analogues are used in the three-staged siting process.

where the Japan Atomic Energy Agency (JAEA) has been characterising 4D site evolution in their underground research laboratory project (e.g. Ota et al. 2010). At both sites, despite clear evidence of fault movement, constant uplift/erosion and sea-level changes, suitable hydrological (H) and hydrogeochemical (C) conditions have been maintained in the host geological environments over millions of years (e.g. Sasao et al. 2006, Niizato et al. 2010, Amano et al. 2011).

IDENTIFICATION OF STABILITY INDICATORS

The site stability can be defined as the persistence of the THMC conditions favourable for the safety of geological disposal. This does not imply, however, that steady-state conditions prevail in the host geological environment over a long period of time. In fact, the geological environment is constantly evolving as gradual processes develop rather slowly with time over a large spatial scale (e.g. uplift/ erosion and climatic/sea-level changes). It is thus necessary to ensure that the extent over which the favourable THMC conditions vary with time would have an insignificant impact on the safety of geological disposal, thereby assuring the long-term site stability.

NUMO have been establishing an appropriate methodology for ensuring the site stability. As part of this, a variety of self-analogue information that is available particularly at Rokkasho, northeast Japan, where low-level radioactive wastes are disposed, along with Tono and Horonobe, is reviewed in detail. This allows 'stability indicators' to be identified, which involve, for example, key properties and processes currently ongoing in the geological environment and signatures of palaeohydrogeological evolution in groundwater, rocks and minerals, all of which are of strong relevance to the site stability. Here, the stability indicators are discussed (see also Fig. 5), particularly focusing on 'geological stability' and 'groundwater stability', both of which are required to ensure the long-term site stability (see discussion in Alexander et al. 2014).

With regard to the 'geological stability', the focus is on ensuring the stability of both tectonic (or regional geological) settings where the target area is located and also host rock environments vis-à-vis perturbations that have occurred during their history. A couple of the potential indirect indications of the geological stability in Japan are identified; for example:

- The extent of variation in the mode and rate of uplift with time is small;
- Coherent, unfolded sedimentary units present within the otherwise mixed accretionary complex of widespread distribution in Japan.

Since the behaviour of the hydrogeological environment has an impact on groundwater movement and chemistry and probably also the transport of radionuclides released from a repository, the main concern over the 'groundwater stability' involves both hydrogeological and hydrochemical aspects. Direct indications of these are shown as 'favourable characteristics/properties' in Table 1. These are required of the geological environment so that reliance can be placed on key safety functions related to the long-term site stability. Of particular concern in this context is the extent to which favourable hydrological (H) and hydrogeochemical (C) conditions have persisted under any natural disruptive events/processes that have occurred at the site of interest. Signatures of palaeohydrogeological evolution in groundwater, rocks and minerals will thus potentially be regarded as indirect indications of the groundwater stability; for example:

- Deep groundwater has a very long residence time, indicating low fluxes and velocities;
- Geochemical anomalies (e.g. uranium mineralisation) persist for a long period of time, despite natural events/processes, demonstrating the long-term maintenance of favourable pH-Eh conditions.

Developing technical knowledge on the indirect stability indicators will reinforce the scientific and technical bases upon which characterisation of the site stability over geological time will be based. More specifically, the stability indicators allow the type and amount of key information to be explicitly identified in a comprehensive fashion, which would be necessary for ensuring a low likelihood of the significant impacts of natural events/processes and also for characterising the 4D site evolution. As such, NUMO will adopt this concept in the planning of PI and DI by considering the relevance of the indicators to the target geological environments in the PIAs and DIAs.

SUMMARY

On the basis of the state-of-the-art geoscientific knowledge that has been developed to date, NUMO has demonstrated both the prevalence of THMC conditions favourable for geological disposal in deep geological environments in Japan and also basic concepts and criteria for precluding the potentially significant impacts of natural disruptive events/processes on the favourable subsurface conditions. This ensures the feasibility of assurance of the long-term stability of the site geological environment.

During the LS stage, NUMO will use relevant self-analogue information that has been gained in similar rock types in similar geological settings on a larger spatial scale in as effective a fashion as practicable to complement the stability evaluation of the sites of interest. Self-analogue information at a couple of sites where palaeohydrogeological evolution was well characterised has now been reviewed in detail to identify potential indirect stability indicators from the viewpoints of, in particular, both geological stability and groundwater stability. In the detailed planning of PI and DI for ensuring long-term site stability in the conclusion of each stage, NUMO will explicitly identify the type and amount of key information in a logical fashion, taking the indirect stability indicators into consideration.

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*JE: in Japanese with English abstract; **J: in Japanese

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USE OF NATURAL ANALOGUES TO DEVELOP AND TEST MODELS OF RADIONUCLIDE RELEASE AND TRANSPORT IN UPLIFT / EROSION SCENARIOS

by

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Unlike other countries where uplift / erosion is treated as a special perturbation scenario, due to the Japanese tectonic setting, the impact of uplift must be considered as part of "likely" repository evolution. In addition, because safety assessments are currently being developed to support the comparison of possible sites or alternative disposal concepts, the treatment of uplift / erosion must be as realistic as possible in order to identify any significant differences between options. In an initial study, the perturbation of the EBS and modification of the far-field were assessed for a simple uplift and erosion scenario, with resultant changes in solubility and sorption of key radionuclides considered – assuming reversible sorption for U and Np, but allowing for partial reversibility in the case of Cs. Greatest uncertainties were associated with the behaviour of the altered EBS as it approaches the surface.

Although the potential for high doses in the distant future exists, the risk is probably not great for HLW – but could be more problematic for direct disposal of spent fuel (SF). A special challenge here is the development of models (and associated databases) to quantify the impact of rock mechanical, hydrogeological and bio-geochemical changes on radionuclide migration – both in the EBS and in the geosphere. The two main questions here would be: how long is it before uplift causes significant alteration of performance and how is performance modified when the geosphere barrier is significantly modified and afterwards when the repository nears the surface?

Even in Japan, significant impacts will occur only in the far future (after hundreds of ka) and develop very slowly. As such, there are great limitations associated with any laboratory studies to investigate the processes involved and therefore natural analogues have great potential here. This paper discusses the key safety-relevant processes and radionuclides of interest – with a focus on the relatively simple case of disposal of vitrified waste in an inland setting, but with consideration of other waste types (TRU, SF) and also coastal environments.

From this, potential analogue studies are listed – considering both data mining of existing knowledge bases and also new projects that are specifically focused on this application.

The ideal NA would probably consist of an eroding ore body containing relevant trace elements associated with massive iron oxides in a rapid uplift setting with relevant climate conditions. For a preliminary assessment, Poços des Caldas in Brazil is a good analogue. One of the main advantages of this site is the large amount of high-quality data available at all levels, ranging from material for the general public that could be used for communication purposes (including video) to very detailed technical reports.

There are two main sites in Poços des Caldas: the Morro de Ferro near-surface Th/REE deposit and the Otsamu Utsumi U mine. The latter is particularly interesting due to the presence of redox fronts, which have been carefully mapped and associated redox processes investigated. This includes colloidal and microbiological effects on the radionuclide migration and retardation properties. At the Morro de Ferro site, some large magnetite veins are present, which could be a good analogue for a very heavily corroded steel overpack in the repository system. Hence, despite the fact that there is no U ore within magnetite, many of the component geochemical processes highlighted in Japanese uplift scenarios can be examined at these sites.

The past comparison between analytical data from Poços des Caldas with blind predictions using thermodynamic models and databases from different organisations shows that, in general, values for solubilities derived from these databases are over-conservative in both oxidising and reducing conditions (although some clear errors led to correction of the databases involved). Nevertheless, great variation in predicted speciation and incompatibility with field measurements shows the limitation of the databases, which could be crucial for the evaluation of retardation processes (which is now more common than it was at the time of this project). Although there is good agreement on the predicted limiting solids, there are few indications that the pure phases involved actually form at Poços.

The Poços des Caldas NA clearly highlights the importance of microbial activity in catalysing redox processes. It is both an important factor in assuring understanding of the processes involved in redox front development, but also indicates the potentially significant role of bio-catalysed secondary mineralisation in the retention / retardation of relevant elements.

In terms of the potential role of colloids, it is shown at Poços that their effect on radionuclide transport properties is insignificant for transport in sub-surface waters. However, in the case of uplift and erosion scenarios, colloidal effects also need to be considered in surface runoff and the biosphere, which have not been examined to date.

Although much has been done on the Poços des Caldas knowledge base, it still has a large potential for further data mining, possibly complemented by future studies on archive samples or new work at the site. In addition, other possible natural analogues should be checked for better understanding uplift/erosion scenarios. Currently interest in this topic is driven by discussions resulting from new regulatory guidelines in Japan, but the technical issues involved may be of wider interest to other tectonically active countries, indicating a potential for international collaboration.

BENTONITE ANALOGUES FOR THE GEOLOGICAL DISPOSAL OF RADIOACTIVE WASTE – CURRENT STATUS AND FUTURE OUTLOOK

by

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INTRODUCTION

Numerous studies on bentonites, focusing on bentonite interaction with other components of the engineered barrier system (EBS) and a range of host rock environments, are present in the literature. In this article (Reijonen & Alexander 2015a), recent bentonite NA studies were briefly reviewed and gaps in the current literature identified. The aim was to:

1) suggest where relevant new information could be obtained by data mining published bentonite NA studies with a new focus on current safety case requirements
2) collect relevant information by revisiting known bentonite analogue sites and conducting investigations with more appropriate (and up-to-date) analytical techniques
3) identify novel study sites where, for example, bentonite longevity in very dilute to highly saline groundwater conditions can be studied (cf. Reijonen & Marcos 2015).

It must be noted that the use of NAs in safety case development is likely to be site- and repository design-specific in nature, and emphasis is thus placed on the appropriate use of relevant NA data on bentonite longevity (see also discussion in Alexander et al. 2014, 2015a, Reijonen et al. 2015).

CONCLUSIONS

This brief overview of several recent NA studies has shown that, although most processes relevant to the long-term behaviour of EBS bentonite under repository conditions could be addressed by NA studies (Table 1), *few have been carried out to an appropriate level*. Either the boundary conditions have not been fully defined or the environments studied were not as relevant as hoped, or the studies were simply not focused enough on supplying data for the safety case (i.e. bottom-up vs. top-down studies; cf. Alexander et al. 2014). This is reflected in the strength of the qualitative discussion and strong process understanding regarding engineered clay barriers and their longevity in general but, on the other hand, in the lack of usable quantitative data.

Nevertheless, it is clear from Table 1 that many NAs of relevance do exist and much information of use in the safety case, both quantitative and qualitative, could be provided. These data could be produced by a mixture of means, including:

- obtaining relevant information by data mining published NA studies with a new focus on current safety case requirements (cf. McKinley & Alexander 2007, Alexander et al. 2007)
- obtaining relevant information by revisiting known bentonite analogue sites and conducting investigations with more appropriate analytical techniques (cf. Milodowski et al. 2015)
- identifying novel study sites where, for example, the long-term stability of bentonite in very low salinity groundwaters can be studied (cf. Reijonen & Marcos 2015)

Table 1. An overview of the main processes of relevance to the long-term behaviour of bentonite in the repository EBS, backfill and seals with recommendations for focused NA studies.

Process studied	Future recommendations regarding NA studies
Thermal effects on bentonite have been studied through NAs, but no information is available on the relevant temperature regime (Posiva 2012a) and will need to be carefully considered in the design and the development of the safety case (Wilson et al. 2011). Wyoming bentonites hold potential for both thermal and geochemical studies (Laine & Karttunen 2010).	Return to more relevant sites (e.g. Busachi in Sardinia, cf. Laine & Karttunen 2010) and apply a range of more relevant analytical techniques to better understand the site history and temperature profiles (see discussion in section 3). The Clay Spur bed (Wyoming) should be studied in order to better constrain the thermal and hydrogeological boundary conditions preserving the deposit.
Bentonite resaturation: large-scale experiments suggest that the theory and laboratory-based understanding of bentonite resaturation may not be a good description of the process at the full scale. Resaturation times may be significantly longer than previously predicted, and full resaturation may not occur in some cases (Wilson et al. 2011).	Boundary conditions for dry and wet thermal reactions can be obtained via new NA studies at existing sites (such as Busachi in Sardinia).
The evidence of bentonite plasticity is interesting, but this topic is not treated quantitatively enough to warrant use in safety assessments (Posiva 2012a).	The approach of Keto (1999) should be repeated plus the process should be included into current URL experiments (e.g. FEBEX, Gens et al. 2002). In addition, try to find a site where canister sinking could be studied by means of boulder fall on bentonite. See also discussions in Reijonen & Alexander (2015b) and Alexander et al. (2015b)
In the case of iron-bentonite interactions, useful NA data are very limited (cf. Marcos 2003, Wersin et al. 2007). Steel/iron corrosion product could react with bentonite to produce reduced swelling or non-swelling minerals, thereby causing changes in swelling pressure, bentonite cementation, and loss of sorption sites. The kinetics of smectite alteration to iron-rich minerals is not well understood (Wilson et al. 2011). Identified need for future studies (e.g. Pelayo et al. 2011 type of NAs, Morron de Mateo).	Existing NA studies are not relevant enough for KBS-3 or other designs, although general observations from natural systems and the NF-PRO URL experiment (Kickmaier et al. 2005) suggest the uptake of iron could reduce swelling pressures. These data need to be integrated with a more appropriate NA study.
Cu-bentonite interaction: only limited information available (Posiva 2012a)	From Kronan cannon, qualitative data are available, which should be more rigorously integrated with existing/new laboratory data.
Effect on bentonite on waste glass in designs with no container, or a very thin container, is not understood. Glass dissolution may increase in contact with some clays, but mechanisms are poorly understood (Wilson et al. 2011).	Sites where natural glasses are in contact with bentonites exist. However, the differences between natural glasses and vitrified radioactive waste should not be forgotten (cf. Miller et al. 2000)

Table 1. Cont.

Process studied	Future recommendations regarding NA studies
Bentonite freezing and thawing processes: only general considerations are available (Posiva 2012a).	A NA study in glaciated terrain could provide more reliable tools for extrapolating the experimental evidence to safety case time scales.
Chemical erosion of bentonite: There is no NA study to support or discount the process (dilute conditions during glacial melt water intrusion, Posiva 2012a). Further research and development is likely to be needed on the erosion of bentonite, especially for specific conditions (Wilson et al. 2011). New work has just been initiated by Posiva (see Reijonen & Marcos, this workshop)	Montmorillonite has been observed to occur as fracture filling mineral, e.g. at repository sites such as Olkiluoto in Finland and Forsmark in Sweden, providing potential *in situ* NA sites for montmorillonite stability under varying geochemical conditions (see discussion in section 4 of Reijonen & Alexander 2015a) (many other sites for similar studies have also been identified). In addition, studies related to the erosional histories of known bentonite deposits under the effects of the last glaciation or by meteoric waters could be of interest.
Effect of saline groundwater on bentonite: saline conditions may need to be further considered (Wilson et al. 2011). The impact of temperature gradients on the accumulation and dissolution of salts in the vicinity of canisters embedded in clay buffer also needs to be assessed. Experimental/modelling/NA work may be required to increase understanding if bentonite is to be exposed to unusually high Mg concentrations (i.e. Mg-rich brine). See also Milodowski et al. (2015).	The approach proposed in section 5 is worthwhile for bentonite-saline groundwater interaction and understanding the impact of Mg-rich brines. The temperature gradient impact on salt accumulation could be addressed at Busachi (see above).
OPC cement–bentonite interaction: experimental work to reduce uncertainties concerning smectite alteration rates and bentonite degradation rates under highly alkaline conditions needs to be extended to include integrated NA studies (Sidborn et al. 2014)	Little hard evidence, but could be addressed at the Khushaym Matruk site in Jordan (cf. Pitty & Alexander 2011, Alexander 2012). At Cyprus, there is a site where pH 12 is observed (see Alexander et al. 2012).
Low alkali cement-bentonite interaction: NA studies at Cyprus are finalised (Alexander et al. 2012, Alexander & Milodowski 2015, Milodowski et al. 2015) and ongoing in the Philippines.	Results (Cyprus) clearly show limited reaction; additional information from the Philippines study can contribute to confidence in the process understanding.
Coupled processes: a key point to note is that, although for simplicity the reactions and processes are discussed as individual topics, many of the processes of relevance to the long-term behaviour of bentonite will be strongly coupled and cannot be considered, or optimised, in isolation (Wilson et al. 2011).	Focussing on coupled processes should be introduced more strongly in NA research in general. Potential sites exist e.g. for cement-bentonite alteration coupled with thermal alteration and these should be studied.
Assessment of clay materials with respect to coupled THMCB behaviour: (including gas conductivity and ion diffusivity) needs to be made in order to obtain a basis for optimal selection of one specific candidate material.	Despite recent advances through the EU projects DECOVALEX (DECOVALEX 2011), FORGE (FORGE 2013) and PEBS (Schäfers & Fahland 2014), there remains a general need to increase understanding of coupled processes, as seen in the latest stage of DECOVALEX (DECOVALEX 2014).
Performance of bentonite – sand/rock mixtures: these are being considered more as repository designs are optimised	In reality, the physical and mineralogical properties of many natural bentonites are more akin to bentonite-sand or bentonite-rock mixtures (Alexander & Milodowski 2015), allowing the long-term behaviour of these mixtures to be assessed quantitatively. To date, only a tentative assessment has been carried out, but this has shown the potential for further, detailed studies.
Consideration of smectites other than montmorillonites: when considering other clays (e.g. saponite, Wilson et al. 2011 or Friedland clay, Posiva 2012b), the first action should be a study of the source deposit and what boundary conditions can be justified based on this (compare e.g. Clay Spur studies in case of MX-80; Laine & Karttunen 2010).	Alteration patterns for trioctahedral smectites like saponite are well known in nature, providing a lot of potential for NA studies in various geological environments.
Microbial processes: it has been generally felt that the bentonite buffer would minimise microbial populations due to the restriction of groundwater flow, but more recent studies (e.g. Stroes-Gascoyne 2010) of several bentonite-backfill and sealing materials used in large-scale experiments at AECL's Whiteshell URL, have demonstrated the presence of culturable populations in all bulk materials.	These few, URL-based examples should be checked by examining natural bentonites for *in situ* microbial populations. Could most easily be carried out in conjunction with studies on other aspects of bentonite (e.g. saline reaction). Certainly, there is a suggestion of microbial processes in the bentonites studied in the Philippines (Alexander et al. 2008).

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A NEW APPROACH TO SCREENING AND INVESTIGATING POSTGLACIAL FAULTS IN FINLAND

by

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Finland is located within the Proterozoic/Archean part of the Fennoscandian Shield, implying that volcanism is absent and seismic activity is low. From this point of view, the Finnish bedrock conditions are favorable for a spent nuclear fuel repository. However, as Finland is situated at high latitudes, future glaciations may pose potential risks for long-term safety. The slow loading of horizontal stresses during the advance of an ice sheet and rapid unloading of the vertical stresses during the melting stage forces the bedrock to readjust to the changing conditions (e.g. Lund et al. 2009). The recorded present-day uplift rates in Finland are up to 9–10 mm/a, and the overall uplift has been nearly 1 km since the Weichselian glacial maximum. Faults disturbing the overlying Quaternary deposits or offsetting glacially polished bedrock surfaces are taken as evidence of glacio-isostatic compensation of the bedrock for the stresses induced by the ice sheet. Such recently activated faults are called postglacial faults (PGF), regardless of their exact date, whether they are late-glacial or truly postglacial. PGFs have been described from Finland, Norway and Sweden since the 1960s (Kujansuu 1964, Kuivamäki et al. 1998, Lagerback & Sundh 2008). Conventionally, they have been identified from aerial photographs or by map interpretations and field observations. The reported surface traces of the faults are up to 160 km long and the largest observed vertical displacement is about 30 m (Lagerback & Sundh 2008, Kukkonen et al. 2010). The faults are thrust faults striking SW–NE and coincide with ancient deformation zones. If the observed displacements formed in a single seismic event, the earthquakes may have reached magnitudes of up to 7–8. So far, all the reported major PGFs are located in the northern part of Fennoscandia (Fig. 1).

During the past couple of decades, the mapping of PGFs has achieved what has been possible using the available tools. With the advent of airborne LiDAR-based digital elevation models, a new and accurate remote sensing mapping methodology has become available. It allows the rapid and low-cost detection of postglacial features, not only fault scarps, but also other morphological features within the fault's impact area, thus significantly increasing the probability of finding and identifying seismic zones (Sutinen 2005, Sutinen et al. 2007, 2009, Mikko et al. 2015). Such features include paleolandslides (see Figure 2). High-resolution elevation data are also capable of revealing features that are often difficult to notice and identify in the field.

The Geological Survey of Finland and Posiva Oy have jointly launched a country-wide LiDAR-based search for PGFs, paleolandslides and other morphological features of Quaternary deposits possibly related to post- and late-glacial seismic activity (Sutinen et al. 2014a-c, Palmu et al. 2015). The project has three main goals: 1) to reassess the existence of PGFs in the southern part of the country, 2) to continue investigations around and at the extensions of the known PGF zones in northern Finland, and 3) to improve understanding of the reactivation

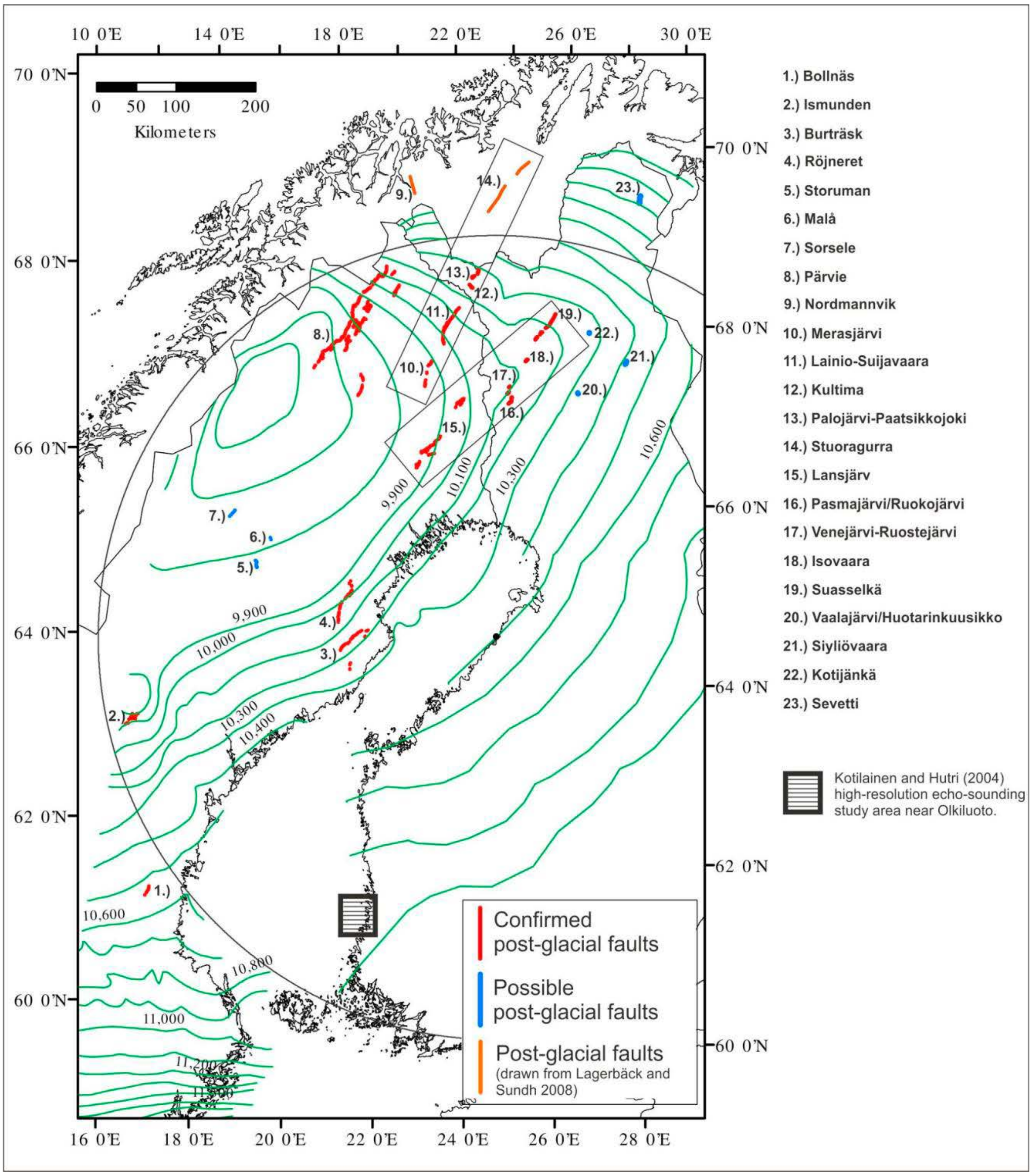

Fig. 1. Post-glacial faults and deglaciation curves of the Late Weichselian ice sheet (Sutinen et al. 2014d). Deglaciation chronology is based on Lunkka et al. (2004) and Lundqvist (2004).

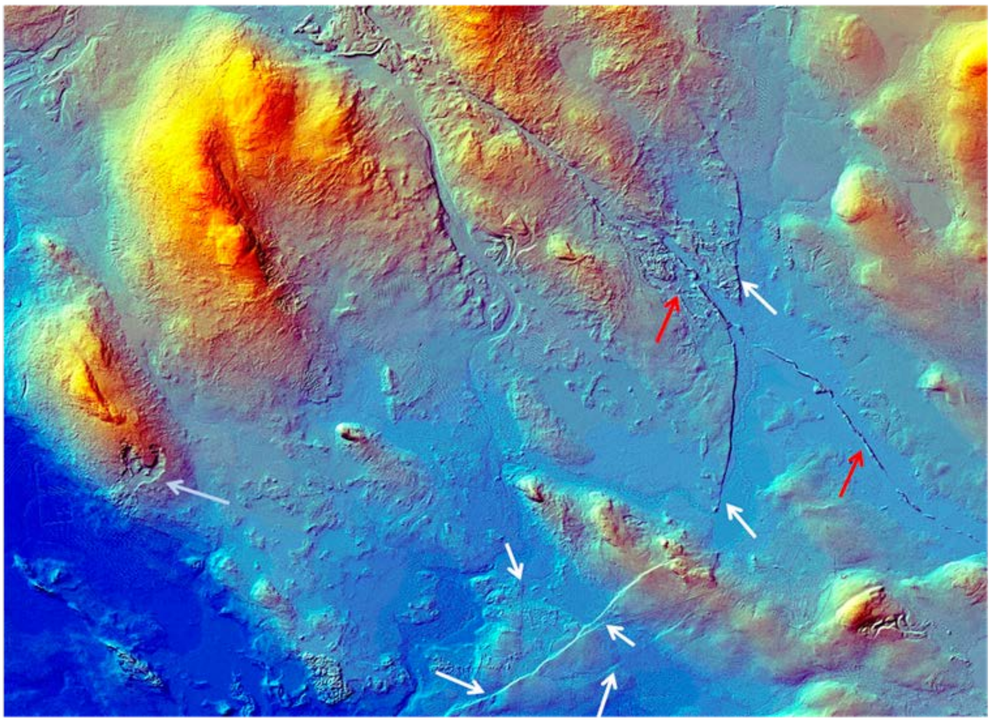

Fig 2. Examples of morphological features assumed to be related to subglacial or postglacial faulting. The white arrows indicate fault scarps showing variable orientations and amounts of displacement. The red arrows point to a well-formed esker, which is seriously disturbed due to seismic activity at the cross-cutting fault zone. The blue arrow on the left-hand side shows a landslide in a sandy till slope. LiDAR image with multidirectional hill shading from NNW. The image area is about 12 km wide. LiDAR DEM © the Finnish Forest Centre.

mechanisms, internal geometry and other properties of the faults. All observations made during the project will be collected and classified into a common geodatabase using ArcGis (© ESRI) software. The outcome of the project will contribute in assessing the seismic risk and stability of the fracture network at Olkiluto during future glacial cycles. A longer term aim is to combine the PGF observations from Nordic countries into a single database in order to facilitate regional interpretations.

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PALEOHYDROGEOLOGICAL INVESTIGATIONS OF GLACIAL WATER EFFECTS IN THE VICINITY OF AN END MORAINE: THE SAIMAA PROJECT

by

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Posiva Oy has carried out preliminary investigations at the second Salpausselkä end moraine in Kyläniemi, eastern Finland, since 2011 in order to investigate the penetration of dilute glacial meltwater in the bedrock, as the ice margin remained stationary for hundreds of years during the deglaciation of Late Weichselian ice sheet.

The hydrogeochemical stability and the buffering capacity in the host rock of the waste disposal facility are important issues affecting the changing future environmental conditions at Olkiluoto. A significant hydrogeochemical change in deep groundwater conditions is expected to occur during the deglaciation of the next glacial period, when a large amount of meltwater will form and the driving factors of groundwater flow will increase compared to the current situation.

The hydrogeological structures and properties (i.e. locations and directions of water-conducting fractures, groundwater salinity) and the velocity of retreat (i.e. how long the meltwater can penetrate into the groundwater system) will determine the hydrogeochemical effects of deglaciation at the geological disposal facility. Although the prevailing hydrogeological and hydrogeochemical properties can significantly reduce and buffer meltwater penetration to the repository depth, it is ultimately a matter of time until the groundwater environment will be determined by the meltwater signature at the repository depth. However, a significant change that would affect the functioning of the disposal facility is the almost complete dilution of groundwater at the repository depth. This would expose the bentonite to erosion.

The aims of this project are to determine 1) the penetration depth and dilution effect of meltwater, 2) the processes that cause groundwater dilution during the extreme deglaciation phase, 3) the oxygen penetration depth during the extreme deglaciation phase, 4) the basic information for the deglaciation scenarios (e.g. bentonite erosion, chemical erosion, post-glacial fracturing), 5) the effects on the fracture network and the groundwater flow rate and 6) the stability of the meltwater disturbance on a timescale of 10 000 years.

The project will be a natural analogue study of an extreme situation in which the ice margin remains stationary while retreating on top of the geological repository at Olkiluoto for a long time period.

THE GREENLAND ANALOGUE PROJECT (GAP)

by

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The *Greenland Analogue Project* (GAP) was initiated in 2008, collaboratively by Svensk Kärnbränslehantering AB (SKB) in Sweden, Posiva Oy (Posiva) in Finland and the Nuclear Waste Management Organization (NWMO) in Canada (Claesson Liljedahl et al. 2015, Harper et al. 2015). The goal of the GAP was to advance the understanding of processes associated with glaciations and their impacts on the long-term performance of a deep geological repository (DGR) in crystalline bedrock. An additional aim of the GAP was to contribute to an increased understanding of a glaciated environment by obtaining an integrated view of ice sheet hydrology and groundwater flow and chemistry. Using the *Greenland Ice Sheet* (GrIS) as a modern analogue for future continental-scale ice sheets in previously glaciated regions, field and modelling studies of the GrIS and subsurface conditions were undertaken. The GrIS was chosen because it is of about the same size as those ice sheets known to have formed, and expected to form in the future in Fennoscandia, which suggests that the scale of processes and response times could be similar during the glaciation and deglaciation phases. Moreover, the bedrock in the study area is crystalline, with similarities to the crystalline bedrock in Sweden, Finland and Canada in terms of composition, fracturing and age. These characteristics make the study site an appropriate analogue of the conditions that are expected to prevail in Fennoscandia and, to some degree, in Canada during future glacial cycles (Claesson Liljedahl et al. 2015, Harper et al. 2015).

The area in which the lack of information is most significant for glacial regions concerns permafrost characteristics and development, groundwater flow and its chemical composition, and bedrock stress and rock mechanical characteristics. The aim of the GAP was to advance scientific understanding of hydrological, hydrogeological and geochemical processes under glacial conditions (Claesson Liljedahl et al. 2015). To achieve the required increase in understanding, GAP research focused on obtaining information that contributes to answering the following six overall project questions:

1) *Where is meltwater generated under an ice sheet?*
2) *What are the hydraulic pressure conditions under an ice sheet, driving groundwater flow?*

3) *To what depth does glacial meltwater penetrate into the bedrock?*
4) *What is the chemical composition of glacial water if, and when, it reaches repository depth?*
5) *How much oxygenated water will reach repository depth?*
6) *Does the discharge of deep groundwater occur in the investigated pro-glacial talik in the study area?*

These questions cover areas where process understanding based on observations from an actual ice sheet setting or the extent of the process (in terms of duration, magnitude or scale) were very limited prior to the GAP. These questions also highlight areas where substantially conservative assumptions have been necessary in safety assessment analyses. To answer these questions, the GAP was divided into three subprojects: 1) surface-based ice sheet studies; 2) ice drilling and direct studies of basal conditions; and 3) geosphere studies, each with specific individual objectives but collectively aimed at contributing to the understanding of the research aims listed above.

The results and research undertaken as part of the GAP have advanced scientific understanding of hydrological processes associated with a retreating ice sheet GrIS, including the temporal and spatial nature of processes occurring on the ice sheet surface, conditions in the ice sheet bed (thermal and meltwater generation) and also interactions between glacial meltwater and the underlying groundwater systems (Claesson Liljedahl et al. 2015, Harper et al. 2015). Hydrological conditions in the ice sheet bed were found to vary across. Between the ice divide and the margin, there is evidence for three different basal zones as defined by the amount and configuration of meltwater: *the frozen zone, the wet bed zone, and the active-drainage zone* (Fig. 1). These zones result from surface, bed, and internal ice flow processes, and are probably representative of Northern Hemisphere ice sheets in a similar stage of development to the current GrIS (Claesson Liljedahl et al. 2015, Harper et al. 2015).

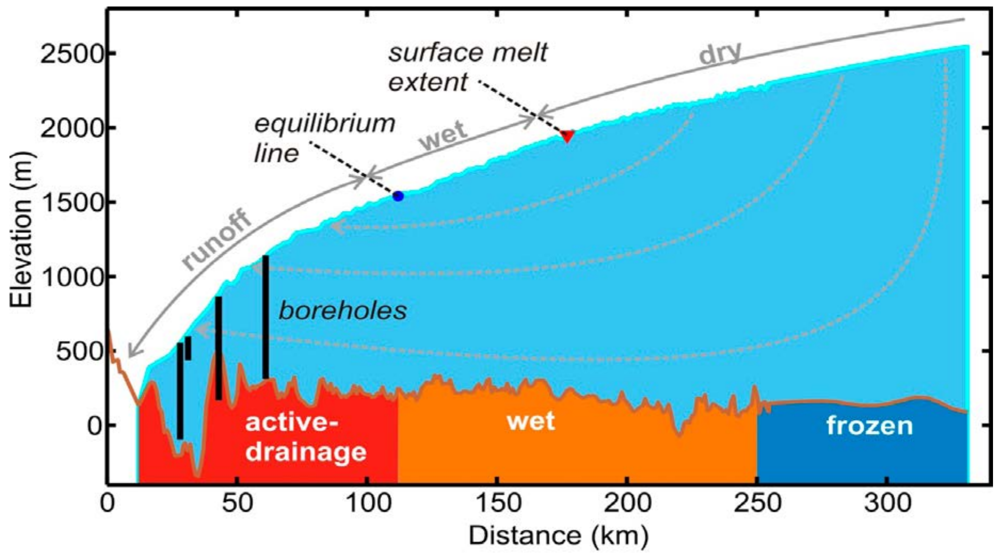

Fig. 1. Between the ice divide and the margin, three different basal zones have been defined by the amount and configuration of meltwater: the frozen zone, the wet bed zone, and the active-drainage zone.

The main part of the base of the ice sheet in the study area, including the marginal areas, has been shown to have basal melting conditions, with the exception of the central parts of the ice sheet. This, together with the glacial history of the region, suggests that permafrost does not exist under the major part of the large warm based areas of the ice, with the exception of the ice margin, where a wedge of permafrost most likely extends underneath the ice for some distance. The existence of unfrozen areas through the entire permafrost thickness, so-called through taliks, in close proximity to a continentalscale ice sheet has been confirmed by scientific investigations conducted on the Talik lake and in borehole DH-GAP01. Several lakes in the investigation area are larger and deeper than the investigated lake, indicating that through taliks are common features in the hydrological system in the study area. Below the permafrost (at depths greater than c. 350 m), reducing conditions are interpreted to prevail in the study area. Past penetration into the bedrock of dissolved oxygen in meltwaters has been limited in depth, as indicated by the presence of iron oxyhydroxides in fractures only in the upper parts of the rock (down to 60 m), with only two isolated occurrences of goethite down to 260 m depth. The stable water isotopic signatures ($\delta\ ^2H$ and $\delta\ ^{18}O$) indicate that the groundwaters sampled in borehole DHGAP04 (depth of at least 500 m) are of glacial meltwater origin. A preliminary interpretation of dissolved He concentrations from the upper section of DH-GAP04 suggests that the deep groundwaters may have residence times exceeding hundreds of thousands of years (Harper et al. 2015).

In assessments of the potential risk to humans and the environment from deep geological repositories for spent nuclear fuel, uncertainties related to process understanding are typically handled using conservative assumptions which over- rather than underestimate the potential radiological consequences. By applying the results from the GAP in future safety assessments, it will be possible to demonstrate a significantly increased process understanding within the fields studied. Such demonstrations of process understanding are a cornerstone in safety assessment analyses. In addition, the increased understanding and associated reduction of uncertainties will reduce the need for pessimistic assumptions in certain areas, and may also allow a re-evaluation of the degree of pessimism in some of the assumptions made in previous safety assessments and modelling work (Claesson Liljedahl et al. 2015).

In this context, some main points of interest are:

- The advance in scientific understanding of the interactions between the hydrology of a retreating ice sheet (GrIS) and the hydrogeology of the bedrock beneath the ice sheet. This gives important information on how the boundary conditions for groundwater flow modelling during glacial cycles should be defined, also in the case of the existence of deep permafrost at the ice sheet margin;
- Evidence of anoxic groundwaters deep in the bedrock under a warm-based ice sheet, conditions which in principle could allow the intrusion of oxygenated water. This gives confidence in the quantification of geochemical parameters at depth, especially on the solubility of solids containing redox-sensitive radionuclides;
- Evidence of the buffering capacity of the bedrock, not only for oxygen, but also pH, to maintain favourable chemical conditions in a repository at depth.

The extent to which the results of the GAP can be applied to safety assessments will depend on the selected repository site(s), the chosen repository concept (e.g. repository depth, type of barriers), the selected safety assessment methodology, and the view of the implementer and regulator with respect to the use of analogue sites (Claesson Liljedahl et al. 2015).

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INDUSTRIAL NORM AND ITS RELEVANCE TO NUCLEAR WASTE DISPOSAL

by

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The majority of natural analogue studies undertaken to date have focused on the geochemical behaviour of uranium, leading to a greatly improved understanding of migration and fixation processes in diverse climatic and geological settings. The findings are now being re-examined to assess the fate, not only of spent nuclear fuel, but also of the large inventories of light-enriched, natural and depleted uranium (DNLEU) that have accumulated in the United Kingdom and other countries over the past few decades.

Despite the emphasis on uranium, it is the longer-lived progeny isotopes, primarily ^{226}Ra, which dominate the dose contribution in post-closure assessments. Relatively little attention appears to have been paid to radium or, for that matter, protactinium, polonium or lead in previous natural analogue investigations. Where radium is addressed, it is commonly assumed to be a mobile element whose transport through rocks and soils can be approximated by an equilibrium adsorption coefficient (K_d). However, evidence from industrial NORM (Naturally Occurring Radioactive Material) sites shows this to be misleading. Although invariably divalent, radium is a strongly redox-sensitive element owing to the influence of the sulphide–sulphate couple. Where precipitated in barite, NORM scales derived from exploitation of mineral deposits can contain very significant levels of radium, in some cases far exceeding the threshold for intermediate level radioactive waste.

The management of NORM is a particular issue for the rare earth, titanium and zirconium industries. In the non-metalliferous sectors, China Clay, phosphate and oil & gas have attracted the most attention to date. There are many similarities in terms of mineralogy and these are highlighted in this presentation, as they largely determine the isotopic composition of scales and other waste residues. Examples are given of base metal mining in Finland and the processing of China Clay in southwest England to show how chemical fractionation and the concentration of radioactive progeny, such ^{226}Ra, ^{210}Pb and ^{210}Pb, could have a major impact on post-closure assessment calculations for the nuclear industry. The wealth of data from previous natural analogue studies provides a useful resource and could be used, in conjunction with evidence from NORM, to improve existing conceptual models of uranium series behaviour.

USE OF NATURAL AND ANTHROPOGENIC ANALOGUES OF CARBON TO SUPPORT THE DISPOSAL OF SPENT EXPERIMENTAL HTR FUEL

by

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This paper sets out a conceptual model for the long-term evolution of carbon (in the form of pyrolytic carbon and graphite) associated with high temperature, gas-cooled reactor (HTR) spent fuel under high-pH conditions in an intermediate-level waste (ILW) disposal vault of a deep geological disposal facility (GDF). Evidence from natural graphite occurrences and industrial processes has been used to support elements of the model and corroborate the expected stability of carbon over a one-million-year timescale.

In HTRs, the carbon is an intimate part of the fuel elements in which (fissile) uranium and (fertile) thorium microspheres ('fuel particles') are embedded in a pyrolytic carbon and graphite matrix. A number of experimental and power-generation reactors have used this technology in the past (AVR, Germany; Dragon, UK; Peach Bottom, USA; THTR-300, Germany; Fort St Vrain, USA) and pebble-bed reactors currently under development use similar fuel technology.

For small-volume spent fuels from experimental reactors, complex processing required to separate the carbon from the spent fuel particles may be less attractive than direct disposal of the intact spent fuel. In the case of historical experimental fuels, disposal as ILW may be considered, as the fuels are no longer significantly heat-producing.

Post-closure criticality safety calculations indicate that the presence of the carbon around the highly-enriched fuel particles acts as a diluent and allows a significantly greater amount of fissile material to be contained in each waste package than if the carbon is not present. However, to take benefit from the presence of the carbon, it must be shown to persist over a timescale of some hundreds of thousands of years.

The key arguments on which the conceptual model for the evolution of the carbon (Neall et al. 2015) is based are:

- Carbon is unreactive, due to unfavourable reaction kinetics, and is only significantly corroded by oxygen or air at high temperature (> 100 °C);

- Reaction of the carbon under ambient GDF conditions requires a surface complexation step in which oxygen (or radiolytic oxidants) binds to the carbon surface. Thus, corrosion will be limited by the very low oxygen availability in the GDF;
- Initial carbon corrosion is due to decomposition of the surface complexes formed by oxygen bound on the surfaces over the storage period. Further carbon corrosion will only take place if radiation dose rates are sufficiently high to generate enough oxidants from water radiolysis to cause corrosion;
- Decay of short-lived fission and activation products will limit the period over which beta/gamma radiolysis of water will occur. By the time that the ILW vault is saturated, substantial decay is expected to have already occurred, reducing the potential for water radiolysis;
- Alpha radiolysis of water around fuel particles will be limited by the shielding effect of the particle coating (SiC) and can only occur after particle failure has exposed the fuel kernel;
- Carbon corrosion will primarily give rise to carbon dioxide rather than methane, owing to the carbon corrosion mechanism;
- Carbon dioxide will be retained in the vicinity of the waste by reaction with the cement-conditioned porewater to form carbonate minerals such as calcite;
- Isotopic exchange of natural groundwater U-238 will cause some loss of U-235 from the fuel, but will prevent U-235 accumulating outside the waste package to create fissile masses;
- Micro-organisms will not significantly affect the stability of the Dragon fuel carbon.

Arguments for the stability of carbon under the low temperature conditions in the ILW vaults draw on observations of graphite from all rocks types: as a primary constituent of igneous rocks but also a nearly ubiquitous component of the clay-size fraction in sedimentary rocks. It is present as a trace component of some of the oldest rocks on earth, such as 3.8 billion-year-old rocks from the Isua Supracrustal belt in Greenland (van Zuilan et al. 2003) and 2.7- to 3.65-billion-year-old quartz-pyroxene rocks from Akilia in Greenland (Papineau et al. 2010). It could be argued, of course, that we see only the remains of more extensive graphite deposits in which the carbon has corroded. However, the detection of fine detrital graphite in billion-year-old metamorphic rocks (Luque et al. 1992) that also contain later metamorphic graphite (which can be distinguished using carbon isotope analysis) suggests this is not the case. There is no evidence that the detrital grains were once larger or more abundant. Despite the later metamorphic events and eventual uplift to the present day surface, the detrital grains have retained their petrographic and geochemical identity.

Evidence for the behaviour of graphite surface complexes formed by oxygen and other oxidants comes from industrial experience of heterogeneous catalysis, where the stability of the graphite surface is of prime importance to the efficiency of the process. Furthermore, Zacharia (2004) documented the relationship between the temperature of desorption of the complexes and the products of the desorption reactions: lower temperature reactions (>130 °C) preferentially give rise to CO_2, whereas higher temperatures (>600 °C) promote CO. This suggests that any desorption taking place under the low temperature (30–50 °C) GDF conditions will produce CO_2 in preference to CO, which could be lost from the vicinity of the waste in a gas phase.

The industrial production and use of graphite also provides insight into the stability of graphite under alkaline conditions that are expected to persist in an ILW disposal vault for many thousands of years due to the presence of large volumes of cement-based materials. Tamashausky (2013) subjected fine-grained (300–850 μm), acid-treated graphite material to one hour of boiling in 2M sodium hydroxide solutions as a method of removing intercalated sulphuric acid. The study showed that the flake graphite exhibits a high degree of chemical stability toward concentrated sodium hydroxide solution. The author suggested that expandable graphite materials are expected to show good *in situ* chemical stability towards other basic systems, and concluded that, based on the test results and past studies using less concentrated alkali, graphite is expected to remain stable when exposed to high pH environments. Although the experiments were of short duration, the treatment with boiling sodium hydroxide under oxidising conditions is quite extreme and certainly far more aggressive than would occur in a geological disposal facility.

Other industrial examples include the use of graphite fibres, powders, colloids and nanomaterials (e.g., carbon nanotubes) as additives in cementitious materials. Such additives are used for various reasons, including improving the thermal and electrical conductivity of the materials, improving their mechanical properties and vibration damping abilities, and enhancing the ability of the materials to provide shielding from electromagnetic radiation (e.g. Cao & Chung 2003, Liu & Wu 2011). The wide range of uses of graphite and carbon in cement-based materials suggests that there is little concern over their chemical stability in such cementitious chemical environments. Although the timescales of these engineering applications are short, probably some decades at most, it should be noted that a number of the applications rely on the use of very fine graphitic components (e.g. carbon nanotubes that are essentially rolled graphene sheets, or graphite colloids that are sub-micron-sized particles) incorporated within the cement matrix. Even small amounts of corrosion of the carbon component could be deleterious to the desired properties, which suggests that there is confidence that there is no on-going reaction between the cement and the carbon under the conditions of the application.

The occurrence and apparent persistence of graphite within the serpentinites provide a longer-term perspective on graphite stability under high pH conditions. In ophiolites, the graphite may have originated from graphitisation of organic material or breakdown of carbonates in intercalated sediments associated with the pillow lavas. Graphites are known from serpentinised ophiolites around the world, but Morrell et al. (2013) have documented specific examples of graphite from rocks associated with Californian serpentinites. These rocks, tens or hundreds of millions of years old, are now uplifted to the earth's surface where the graphite is exposed to high pH, reducing pore fluids formed as the serpentinisation reactions continue at low temperatures due to percolating groundwater. The persistence of graphite over geological timescales under these conditions gives weight to the experience from industrial applications of graphite that it is very resistant to reaction under alkaline conditions.

In summary, the use of natural and industrial analogues has helped to build confidence that the graphite and pyrolytic carbon associated with HTR spent fuel will remain predominantly in its original form in the vicinity of the enriched uranium over the timescale of interest. A minor amount of oxidation cannot be ruled out, due to the radiolysis of water providing a source of oxidants after the ILW vaults are saturated with reducing groundwater. However, there is also strong evidence from natural hyperalkaline systems to support the expectation that any carbon dioxide produced as a corrosion product will be retained in the form of carbonate minerals very close to the spent fuel due to reaction with the cement-conditioned porewater in the ILW vaults.

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POTENTIAL LONG-TERM ALTERATION OF NSF AND MINOR RADIONUCLIDES. INFORMATION FROM NNAA

by

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Evidence from laboratory experiments and natural analogue studies have indicated that the most likely solid phases to form from uraninite (UO_{2+x}) alteration under reducing conditions are secondary uraninite and coffinite ($USiO_4 \cdot nH_2O$). All the coffinitization processes observed in nature occurred at temperatures in the range 130°C to 300°C. The ages of some of the coffinitization events are not very far from the times of interest for the deep geological repository, according to the natural sites reviewed. Much evidence of the presence of minor elements in both secondary uraninite and coffinite has been found. The different coffinite compositions reported in the literature contain U, Th, Y, REE, Ca, Zr, Hf, Fe, Si, P and S.

The results from the calculation of the evolution of the fuel composition over time indicate that, at the moment of canister failure (considered to happen after 10^5 years), the content of REEs, Th, Np, Pu and Zr is always lower than the content reported in secondary uraninites and coffinites, thus indicating that these secondary phases can potentially retain REEs, Th, Np, Pu and Zr released due to the fuel alteration. Elements present in the fuel as metallic inclusions will not be so prone to be retained in these solid phases, but to form individual secondary solids, mainly sulphides, as evidenced by field observations.

The three main conditions identified as potentially causing the formation of the secondary solid phases studied in this work are: i) the likely presence of high concentrations of Si (or silica) due to the alteration of vitrified wastes, or from the surrounding clay (bentonite) backfill material or surrounding host rocks; ii) the potential development of an alkaline plume leading to high pH values in the system due to the possible co-location of wastes and iii) the development of highly reducing conditions (or the prevention of the development of oxidising conditions) due to the anoxic corrosion of steel canisters, with subsequent hydrogen generation.

NATURAL ANALOGUE STUDY RUPRECHTOV: AN EXPERIENCE REPORT

by

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The preferred option for the long-term management of high-level and long-lived radioactive waste is disposal in deep geological formations. The realization of such disposal concepts is based on both technical feasibility and long-term safety. The latter requires a wide range of technical and scientific data for assessing the long-term performance of disposal systems and sites, which is supported by additional complementary information in a Safety Case. For the latter, natural analogues are valuable.

The main value of natural analogue studies is to provide information on a geological system (evolution), i.e. the characteristics of processes occurring over very long time scales. In general, the direct use of quantitative information from natural analogue studies in a Safety Case has been mostly limited, because it is very difficult to extract hard numerical data from complex natural systems, where initial and boundary conditions cannot be fully defined. Indirectly this information could, however, be very valuable in a supportive sense. Therefore, natural analogues are an integral component of the Safety Case in many national repository programmes.

The experiences from more than 15 years of research with the natural analogue Ruprechtov have been compiled and documented in a joint report of GRS and ÚJV. Besides a brief overview on the different roles of analogues in national repository programmes and the evolution of analogue application in Safety Cases, the intentions and objectives of this report are to:

- compile and critically discuss the decisions regarding the selection of the Ruprechtov site as a natural analogue,
- classify the Ruprechtov site with regard to the type of uranium accumulation,
- display the iterative steps, decisions and evolution of knowledge during the investigation of the site,

- describe the experiences obtained, particularly in the selection and application of experimental laboratory and field methods,
- outline the scheme by which these methods have contributed to understanding and characterizing the main features of the site,
- illustrate the main findings relevant for a Safety Case for a radioactive waste repository, and
- outline recommendations for future research and development (R&D) from the lessons learned.

The report structure reflects the experimental/methodological approach applied in conducting the project (see Fig. 1). The existing knowledge at the onset of the project is compiled and discussed. It includes knowledge concerning uranium deposits and natural analogue studies, previous information on the geology at the Ruprechtov site and repository systems in Germany and Czech Republic at that time. Subsequently, the methods applied and the key experiences are summarized with a corresponding standardized description of these methods in the annex. A wide variety of methods are introduced, including drilling, sampling, characterization, *in situ* measurements, analytical methods, isotope investigation and modelling tools. Each method is well described in a standardized scheme. Furthermore, the investigation of key scientific issues by application of different methods is described and major Ruprechtov results are summarized. We identified five main topics: identification of groundwater flow patterns, geochemistry, microbiology, organic matter and uranium transport and enrichment processes. Next, the evaluation of the selected key topics provided the basis for reaching a comprehensive system understanding of the Ruprechtov site. It comprises the current situation, the geological development and the key processes that are occurring or have occurred during the geological past at the Ruprechtov site. In addition, new knowledge from other research became available and was incorporated into the synthesis of the system. Finally, the report discusses how these results could be used in supporting a Safety Case. One key objective of this report is also to illustrate and include experiences gathered during the project, which cannot always be found in technical or scientific reports.

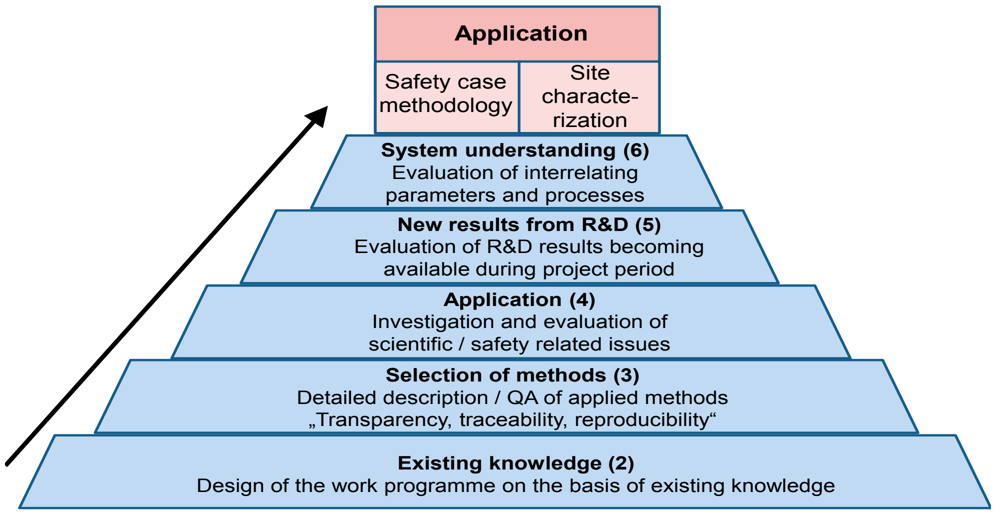

Fig. 1. Structure of the project steps and corresponding chapters of the report.

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CURRENT NATURAL ANALOGUE ACTIVITIES IN GERMANY

by

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INTRODUCTION

In 2013, a new Site Selection Act (Standortauswahlgesetz) became operative in Germany. The aim of this Act is to find a disposal site for high-level waste in Germany in a scientifically-based, transparent process. The geological formations included in this process as potential host rocks will be rock salt, clay and crystalline rock. Important elements in the site selection process will be the safety cases for selected potential repository sites/formations. Due to the German requirements for the disposal of heat-generating radioactive waste (BMU 2010), a safety case needs to demonstrate that containment of the waste within the so-called containment-providing rock zone will be achieved. Therefore, safety concepts are based on the safety functions of containment and to some extent retardation. The key barriers providing these safety functions are the geological formation and geotechnical barriers. Thus, underpinning the integrity of these barriers is crucial, and natural and anthropogenic analogues consequently play an important role, alongside laboratory experiments and process modelling.

This study introduces the top-down approach used to identify relevant analogue studies to be used in the safety case. For a potential repository in rock salt, a systematic analysis was started few years ago and is described in Wolf et al. (2015). Since then, some aspects have been further developed and some new results have become available. Brasser et al. (2014) have compiled detailed reviews for the following NA-relevant topics: the behaviour of competent formations, compaction of crushed salt, composition of fluid inclusions, thermal and mechanical stability of rock salt, impact of earthquakes on a rock salt formation/repository, qualified shaft and drift sealings, iron corrosion, and microbial processes. The reviews include the relevance for the safety case, knowledge from laboratory and field experiments and modelling, and potential natural analogues, with an evaluation including the time frame, uncertainties, limits of application, suitability for communication, open questions and references. Selected new aspects on geomechanics are briefly discussed here.

SELECTED NEW ASPECTS FOR A REPOSITORY IN ROCK SALT

Mechanical stability and self-sealing of rock salt

An important aspect for the disposal of radioactive waste in rock salt formations is the viscoplastic behaviour of the salt. By the convergence process in several 100 m depth, the pore volumes in backfilled areas and in the excavation-disturbed zone will be reduced and, depending on the convergence rate and temperature, completely sealed after a few thousand years. Analogues can be used to demonstrate these properties and the influence of corresponding processes under repository conditions. Analogues from mining and geological analogues are considered to be particularly suitable in this respect.

One example demonstrating the self-sealing capacity of rock salt is the analyses of the post-gas-frac situation at the Merkers site after a rockburst at Völkershausen (Popp et al. 2007). After the disastrous event of barrier rupture due to CO_2 expulsion, a time-dependent recovery in the integrity of the mechanical and hydraulic barrier was, at least partly, demonstrated during over a short period of only 18 years. It has to be mentioned that, in addition to the rock burst itself and the associated event of a gas-frac, the subsequent sealing could be simulated by the performed rock mechanical back-analysis of this scenario (for details, see Popp et al. (2007).

The self-sealing of the excavation disturbed zone is also observed in a technical analogue at Asse mine, where a drift was excavated in 1911 and 25-m-long part of it was subsequently sealed by a steal liner and concrete. Permeability measurements around the sealed drift demonstrate that it decreased from approx. 10^{-15} m^2 after excavation (and still today observed in the EDZ of the open drift) to 10^{-19} m^2 today (Wieczorek 2003). The observations of this technical analogue, which covers a significantly larger time frame than laboratory and field experiments, have now been simulated by constitutive models describing the thermo-mechanical deformation of rock salt (Hampel et al. 2015), and might consequently serve for the qualification of these models. Similarly, fractures observed in the Sigmundshall salt mine and probably originating from the formation of the salt dome and its tectonic situation are also completely filled with secondary halite crystals, and have been efficiently, hydraulically sealed (Fig. 1, middle).

Another illustrative example showing the convergence process is the embedding of tools or other mining materials as, for example, observed in the Hallstadt salt mine in Austria for more than 3000-year-old material (see Fig. 1, right). Examples like this might help to visualize and communicate the convergence process and the principle of containment of the waste in rock salt, particularly to non-technical audiences.

Fig. 1. Technical analogues from mining and geological analogues for convergence and self-sealing of rock salt: A drill core with sealed rock salt after gas-frac (left, Popp et al. 2007); fractures at the 350 m level in the Sigmundshall salt mine (Bokeloh, middle, Hammer et al. 2012) and an old wooden staircase, 1300 BC, Salzkammergut (Austria, right).

Earthquakes

The impact of an earthquake might impair the integrity of the host formation and has to be addressed in a safety case. Although a repository site in Germany will certainly not be located in a seismically very active area, there is a need to demonstrate that the containment-providing rock zone will not be jeopardized by the dynamic strain of an earthquake and keep its safety function. Besides geomechanical model simulations, technical analogues, namely rock bursts, have been identified as strong arguments to estimate the effects of an earthquake on rock salt. Therefore, twelve salt mines in which rock burst have occurred during the last century have been compiled by Minkley et al. (2010). Investigations showed that the geological barrier of rock salt and salt clay had retained its integrity against overlying aquifers in all cases. From the analyses of the rock bursts, it became clear that they provide a significantly higher dynamical strain compared to that from an earthquake. A general conclusion, which is discussed in more detail in Minkley et al. (2010), is that a repository in rock salt is earthquake-resistant for a sufficiently thick salt barrier.

ANALOGUES FOR A REPOSITORY IN CLAY FORMATIONS

According to the German safety concept for a repository in clay, the containment of the radioactive waste is primarily achieved by hindering radionuclide transport by chemical and physical processes resulting from the positive properties of the clay host rock in combination with geotechnical barriers (Rübel et al. 2014). This primary goal is achieved by a set of goals listed in the safety concept. Currently, a systematic analysis has been started in which analogues can be used for a German Safety Case for a repository in clay. This approach is based on the

Table 1. Subset of goals for the safety concept for a repository in clay, and analogue information contributing to underpinning the goals.

Safety concept: Subset of goals	Potential analogue information
The transport of pollutants through the CPRZ is hindered and retarded after their mobilization from the waste. The CPRZ will keep its barrier function over the whole assessment time frame	Relevant processes that have occurred through geological time can be deduced from tracer profiles in the rock pore water. Clay formations in contact with alkaline pore water form natural geological environments to demonstrate the clay buffer capacity. Resealing of fractures might also be supported by analogues.
The maximum temperatures in the host rock are limited to a value that the barrier function of the host rock is not inadmissibly affected by	Analogues where the clay was exposed to elevated temperatures in the past, e.g. from magma intrusion, to assess the effect of elevated temperature on the rock.
The gas pressure development in the repository is limited in a way that the integrity of the host rock is unaffected by the gas pressure or by gas flow through the rock	Clay formations under a pressure load from gas can be found in natural and technical environments. Examples include natural or technical gas resources in or covered by clay formations.
Microbial processes should be limited as much as possible	Anthropogenic analogues and re-investigation of existing natural analogues with respect to microbial processes

formulation of a subset of goals, i.e. how analogues can contribute to demonstrating that these goals are met. In the first step, a subset of goals has been identified where arguments from analogues are expected to be useful. These and the first ideas for potential analogues are listed in Table 1. This work will be further substantiated in the future.

ACKNOWLEDGEMENT

This work was financed by the German Federal Ministry of Economics and Technology (BMWi) under contract nos. 02 E 10055, 02 E 10719. We would also like to thank Wolfgang Minkley and Till Popp from IFG Leipzig for providing their results for this compilation.

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COMPLEMENTARY CONSIDERATIONS FOR THE SAFETY CASE

by

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The Complementary Considerations (CC) report was published as a part of the Posiva's TURVA-2020 safety case that was produced to support the construction license application for the spent nuclear fuel repository to be built at Olkiluoto, Finland (see Posiva 2013 and Reijonen et al. 2015). The next phase in the disposal programme is the operational license application, for which the safety case will be updated.

The requirement for such a document comes from the Finnish regulations (YVL Guide D.5). According to these, 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, which may include analyses by simplified methods, comparisons with natural analogues or observations of the geological history of the disposal site.

The TURVA-2012 safety case and CC were considered by the regulator to be at sufficient level for the construction license application, but requirements were set for updating the safety case for the operational license application.

The updates needed include better integration of the discussions in CC with the safety functions and performance targets, meaning in general better integration of the CC and reports that it mainly supports, these being the Features, Events and Processes report, Performance Assessment and Formulations of Scenarios.

Updates will also be carried out regarding, for instance, design updates and new information on natural analogues. The final report should provide more in depth, site-specific discussion on natural analogues providing support especially for conceptual understanding within the safety case. The main results are also presented in the synthesis report, providing another line of evidence and arguments for the safety assessment supporting the overall safety case.

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USING NATURAL ANALOGUES TO BUILD STAKEHOLDER CONFIDENCE IN GEOLOGICAL DISPOSAL – A CATALOGUE OF NATURAL ANALOGUES FOR RADIOACTIVE WASTE MANAGEMENT

by

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As part of its generic Disposal System Safety Case (NDA, 2010), Radioactive Waste Management Limited (RWM, the UK implementor of a Geological Disposal Facility (GDF)) notes the following in relation to analogues:
"*Analogues can be helpful in demonstrating understanding of aspects of GDF performance and provide evidence that certain materials can survive for long periods. However, they do not provide conclusive proof that these materials will survive for the required periods in the environments of a particular GDF, as the conditions under which the analogue material has survived may not match those expected to occur or evolve in a GDF. Therefore, analogues will be used with caution, and can only ever provide supporting arguments in an Environmental Safety Case. Nevertheless, appropriate analogues can be helpful in providing a long-term practical demonstration to support the theoretical and mathematical arguments*".

To provide a reference source of safety-relevant examples from natural systems that could be used to support a safety case, British Geological Survey (BGS) and Bedrock Geosciences are working with RWM to produce a catalogue of natural analogues for radioactive waste disposal (BGS & Bedrock Geosciences 2015). Aimed at regulators, policy makers, managers, decision-makers and others involved in the waste management process who are not necessarily specialists in natural systems, the catalogue will be in the format of a set of "flyers" summarising the key points of each natural analogue. It is intended to be suitable for the general scientific audience, and its scope includes all aspects of disposal relevant to the UK situation (a range of waste types and potential GDF host rocks are considered; examples are grouped into four main sections - the engineered barrier system, natural barrier system, radionuclide migration in natural systems, and natural analogues and the safety case).

For each analogue considered, information is provided as follows: (i) a description of the specific aspects of components of a GDF that the natural or archaeological analogue system represents or is applicable to, (ii) a summary description of the geoscientific characteristics of the natural analogue, (iii) key safety-relevant observations, (iv) a summary of limitations of the use of the analogue, (v) images of systems that can be used in RWM media with appropriate acknowledgement/ accreditation, and (vi) a list of references where further information can be obtained.

Figure 1 presents a snapshot of some pages from the catalogue, covering one of the analogues considered therein.


<u>*Category 1B:*</u> *Container performance: Long-term stability of copper canister*

Littleham Cove, south Devon, United Kingdom – Corrosion of native copper in Permian mudstones

Overview

Copper is one of the materials which may be used in a repository to contain high-level waste (HLW) and spent fuel.

The use of copper waste canisters in a repository

- The Swedish KBS-3 repository concept for the disposal? of HLW and spent nuclear fuel, consists of copper canisters with a cast iron insert containing the waste.
- Each canister is emplaced into a hole excavated in granitic rock at depths of 400-700 m, and sealed in place with compacted bentonite (a type of clay?) which protects the container.
- Copper is chosen as the outer canister material because theoretical and experimental studies show that it will be resistant to corrosion in the mildly alkaline, reducing groundwater environment predicted to be persistent in this repository environment. As a result, the copper will be expected to isolate and protect the waste for long periods of time.

The Littleham Cove Natural Analogue Study

Naturally occurring copper sheets are preserved in the Permian age Littleham Mudstone Formation, at Littleham Cove, in south Devon, Southwest England. They can be studies to understand the long-term stability of copper enclosed in clay – as could be found in a repository where copper canisters are used.

ABOV: Uranium-vanadium-rich concretions (surrounded by grey reduction halos) in red mudstone containing sheets of well-preserved native copper (INSET). LEFT: Location of the Littleham Cove Natural Analogue Site.

3

- At Littleham Cove, the copper is 99.9% pure and occurs in discrete plates (160 mm diameter and up to 4 mm thick) enclosed in a silty clay matrix. Each plate is formed by a stack of thin copper sheets, varying in thickness from <0.1 to 2 mm. These sheets grew in situ within the mudstone, along originally more permeable sandstone and siltstone bedding planes. and were formed about 176 million years (Ma) ago.
- Since it was formed, the copper has altered and corroded but detailed studies have shown that this occurred soon after the copper was buried.
- Between 30-80% of the original thickness of the copper remained unaltered and preserved within the clay 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.

Uncertainties and differences

- Unlike a purpose-designed bentonite clay barrier system in a repository, the natural clay of Littleham Mudstone Formation has a different mineral composition to bentonite and has not be engineered to provide a good seal as would be required in a repository.
- The Mudstone will probably have greater permeability and than engineered bentonite buffer in a repository.
- The porewater in the Littleham Mudstone Formation may have been significantly different to those anticipated in a repository. However, the chemistry does show that the porewater remained reducing following burial so that the copper and other sulphide and arsenide minerals were preserved until uplift and erosion exposed the mineralisation to the current weathering environment.
- It also seems likely that the porewaters in these Permian strata would have been sulphate-rich and highly saline, possibly halite- and anhydrite saturated. The groundwaters in the deeper parts of the Permo-Triassic basins of the United Kingdom, including the Wessex Basin in southern Britain, are characterised by highly saline groundwaters. Furthermore, halite and anhydrite deposits and matrix cement minerals are known to have been formerly present in the overlying Triassic strata, and are still preserved in the deeper parts of the basin to the east. Such highly saline groundwaters may potentially be more react more with copper than more dilute groundwaters in some repository environments.
- The natural analogue study has not provided any information on corrosion rates and provided no quantitative data to support safety assessments. However, it does provide qualitative support to the copper canister concept.

Relevance – what we have learnt

- The Littleham Cove Natural Analogue Study demonstrates that copper metal buried in a compacted clay environment can remain stable and resist corrosion for a very long period of time.
- In this particular case, after early corrosion and alteration of copper during burial, the remaining copper (representing 30-80% of the original copper mass) effectively remained inert and isolated from further corrosion within the naturally-compacted mudstone for at least 170 Ma, until uplift and erosion exposed it to alteration by the present near-surface weathering environment. This is well in excess of the timescales (up to 1 Ma) considered in radioactive waste management safety assessments.

4

- The preservation of copper metal in this natural environment provides support to the prediction that copper canisters can potentially resist corrosion within the repository environment for the safety assessment design goal of 100 000 years.

Further reading

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Additional illustrations

Additional illustrations for the Littleham Cove Natural Analogue Study, together with captions, are available in the appendices included on the CDROM

Fig. 1. A snapshop from the RWM Natural Analogue Catalogue. Information is presented succinctly, such that a series of leaflets for stakeholders could be produced, covering a range of analogues.

Once complete and published, the catalogue could be used in conjunction with other forms of communication (web-based, social media) as part of a multi-pronged approach to reach a range of stakeholders. It could also act as an education tool.

Examples of recent other work RWM has published in relation to analogue studies are Alexander & Milodowski (2013), Alexander et al. (2013) and Alexander & Milodowski (2014), relating to the Cyprus Natural Analogue Project.

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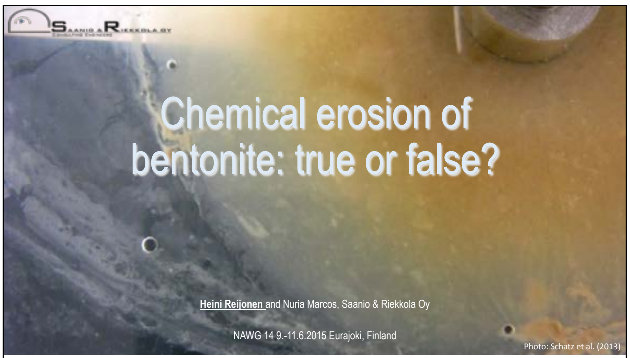
APPENDICES



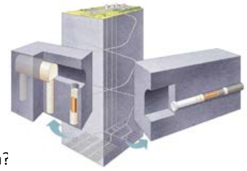
Chemical erosion of bentonite: true or false?

Heini Reijonen and Nuria Marcos, Saanio & Riekkola Oy

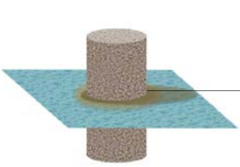
NAWG 14 9.-11.6.2015 Eurajoki, Finland

Contents

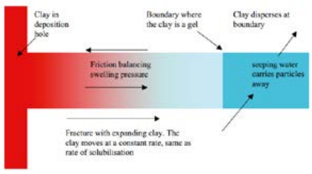
- Background
  - Basis for the FEP
  - Assessing the impact
- Natural analogues for chemical erosion?
  - Development of dilute groundwater conditions
  - Erosion of bentonite
- Conclusions



Basis for FEP – chemical erosion



Posiva (2013a)



Schematic illustration of buffer extrusion and erosion in an intersecting, transmissive fracture From Liu & Neretnieks (2006)

Basis for FEP – chemical erosion

- Two sides to think about:

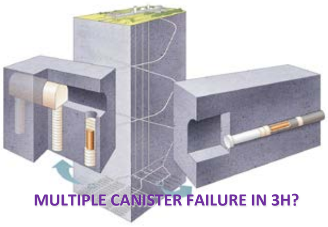
1) Development of the groundwater conditions at repository depth at Olkiluoto site

2) The process of chemical erosion
   - Laboratory experiments and modelling
     - Limited amount of experiments
     - Modelling capabilities limited
   - See e.g. Schatz et al. (2013)

General understanding is that bentonite erosion does not occur when total charge equivalent of cations in groundwater is higher than 4mM, which is reflected in the long-term safety requirements (e.g. SKB 2011, Posiva 2013a).

Assessing the impact (TURVA-2012 safety case)

PERFORMANCE ASSESSMENT (3V):
- Modelling of the groundwater evolution
  - Simplifications and uncertainties
- Modelling of the bentonite erosion
  - Uncertainties, ongoing model development
- FEP was assessed in radionuclide release scenarios due to these uncertainties (Posiva 2013b)



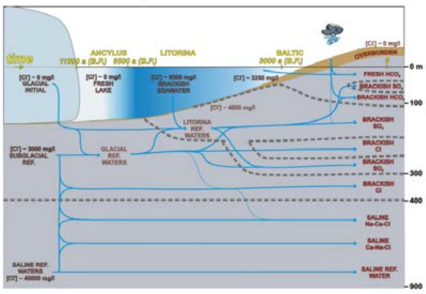
Natural analogues

FOR

1. Development of host rock conditions

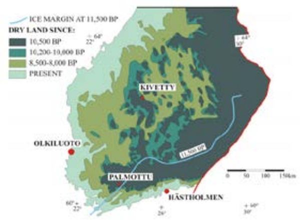
2. Erosion of bentonite

Development of dilute groundwater conditions 1 – on site



Posiva (2013c)

Development of dilute groundwater conditions 2 – reaching out



Pitkänen et al. (2002)

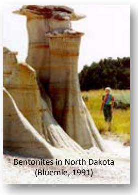
Development of dilute groundwater conditions - notes

- By investigating other sites, process understanding can be increased.
- Modelling development of the groundwater and mixing/ water-rock interaction processes is important!
  - We don't want to pile up uncertainties and go over conservative
- However, no other site can be truly analogous in relation to groundwater systems, due to heterogeneities in geological and climatic controls.
- Discussion and multiple lines of evidence to complement modelling (this was done in the last PA)

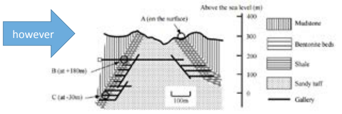
Natural analogues for erosion of bentonite 1– Fetching far

1. Bentonite deposits

- Erosion of bentonite deposits by fresh water
- Limitations: multiple erosional mechanisms, e.g. weathering, drafting.



Bentonites in North Dakota (Bluemle, 1991)

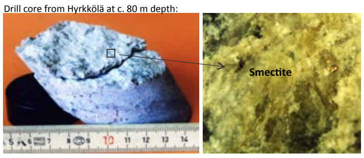


Example of a situation where natural bentonite erosion could be studied. Schematic cross section through the Tsukinuno bentonite mine, northern Japan. Locations of sampled groundwater (A - C) and their depths (above the sea level) are indicated. Kuno et al. (2002).

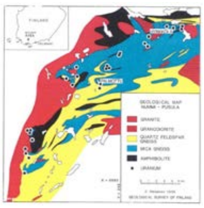
Natural analogues for erosion of bentonite 2– Coming closer…

2.1 Hyrkkölä site in Finland

Drill core from Hyrkkölä at c. 80 m depth:



Marcos (2002)



Marcos and Ahonen (1999)

Current groundwater composition (mgL^{-1}) at this depth:

pH	TDS	Ca	Mg	Na	K	Si	Fe	U	Cl	SO_4	HCO_3	NO_3	O_2
6.6	116	14.4	4.3	7.1	2.8	5.9	0.05	0.093	3.8	10	57	10	4

19/11/2015

2.2 Päijänne tunnel

Providing drinking water to Helsinki area since 1982, tunnel depth between 30 – 130 m.

During pre-construction characterization, montmorillonite was found as fractures filling – no or only very thin grouting or reinforcenment carried out

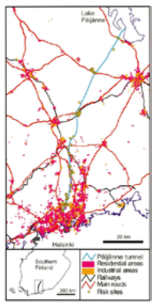
1997 cave-in due to montmorillonite swelling and erosion; rock fall at 60 m depth along 20 m distance.

Flow rate 2.9 m^3/s

pH	TDS mg/L	Ca mg/L	Mg mg/L	Na mg/L	K mg/L	Si mg/L	Fe µg/L	Al µg/L
7.2/8.5	116	18	1.7	5.2	1.3	5.9	77	6

https://www.hsy.fi/

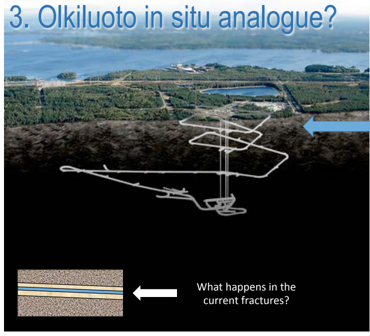
19/11/2015



Lipponen (2002)

Natural analogues for erosion of bentonite 3– Arriving Olkiluoto?

3. Olkiluoto in situ analogue?



– preliminary studies ongoing on fracture smectites (see Reijonen & Alexander 2015 for overall idea)

		Fraction <2 µm	2-20 µm	20-63 µm
Sample PL70				
Montmorillonite		x		x
Sample PL81				
Montmorillonite		x	x	
Sample PL87				
Montmorillonite		x	x	x
Sample PL295				
Montmorillonite			x	
Sample PL957				
Montmorillonite		x		
Sample PL960				
Montmorillonite		x	x	

Montmorillonite occurrences from ONKALO (Gehör 2007)

”Self-analogues” elsewhere?

- Smectites are observed at least at:
  - Other site selection sites in FINLAND:
    - Hästholmen (Gehör et al. 1997a, Front et al. 1999)
    - Kivetty (Gehör et al. 1997b)
    - Romuvaara (Kärki et al. 1997)
  - SWEDEN
    - Oskarshamn and Forsmark sites (Drake et al. 2006 and Sandström et al. 2008)
  - CANADA
    - Bruce site, (Koroleva et al. 2009)
- Smectite is rather wide spread → crystalline and sedimentary environments

Conclusions – dilute or not?

- Dilute water intrusion is site specific process
- Studies on other sites can help in process understanding, but are not direct analogues
- Studies are needed to better describe the processes expected to occur
  →**conceptual understanding on the ”dilute” scenarios**
- Modelling needs to be developped to provide more realistic results

Conclusions – eroding or not?

- **Smectite occurrences are wide spread**
- With focused studies their occurrence can give valuable insight, also in relation to their behaviour in dilute GW conditions
- Natural analogue studies can help to understand the whole process of erosion, which is currently understood only via very limited experience in laboratory scale

Way forward

- To look for smectite occurrences
  - in repository relevant (site) conditions
  - In dilute gw conditions
- To analyse
  - Mode-of-occurrence
  - Age?
  - Alterations?
- To understand all the pieces in the puzzle
- **We can start by looking at the on site smectites**

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British Geological Survey
NATURAL ENVIRONMENT RESEARCH COUNCIL
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Montmorillonite alteration by alkaline groundwater in Cyprus: a natural analogue for bentonite reaction in low alkali cement pore fluids

The Cyprus Natural Analogue Project [CNAP]

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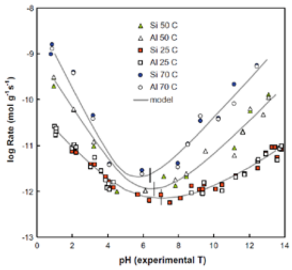
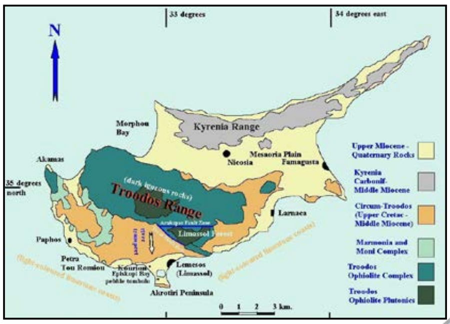
Introduction



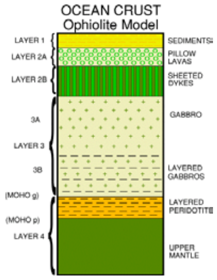
Rozalén et al., 2009, GCA 73, 3752–3766

- High pH silicate minerals – including smectite (the main component of bentonite) have increased solubility
- Potential alteration of bentonite by cement pore fluids leaching from waste form and construction materials
- Potential loss buffer swelling and sealing
- Potential loss of chemical retardation properties: ion exchange, sorption

Geological Setting



Generic ophiolite model



Natural alkaline groundwaters (pH 9-12) are associated with low-temperature sepentinisation reactions

Main reaction pathways to serpentine involve olivine ($(Fe,Mg)_2SiO_4$)* interaction with CO_2 → serpentine ($Mg_3Si_2O_5(OH)_4$)

Additional products can include:

- magnetite (Fe_3O_4)
- magnesite ($MgCO_3$)
- brucite ($Mg(OH)_2$)
- methane (CH_4)
- hydrogen (H_2)

* in all cases, pyroxene group minerals can replace olivine as precursor phase

High Troodos: Alkaline groundwaters at Allas Springs

Troodos Ophiolite sequence

Peripedhi Formation:

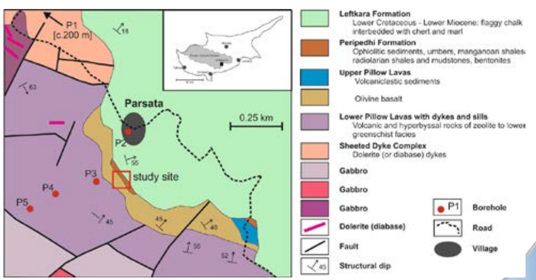
Ophiolitic sedimemts: deep ocean floor - umbers, manganoan shales, finely-banded radiolarian shales, mudstones,and bentonite

Sequence represents uplifted and eroded "window" of ancient **ocean crust and mantle** rocks

Schematic lithographic section of the Troodos ophiolite

[From West, 2007]

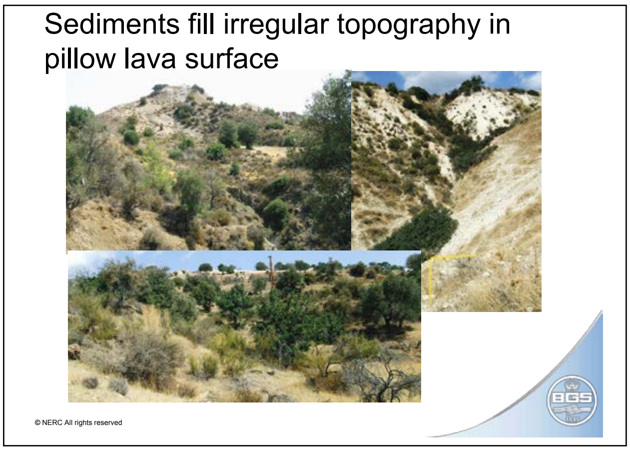
Geology of the Parsata site

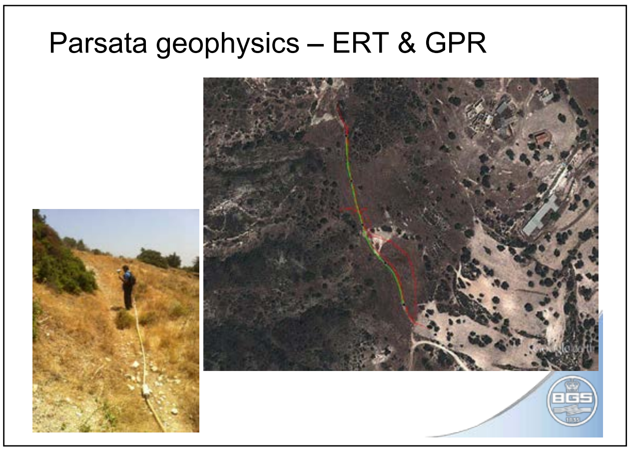


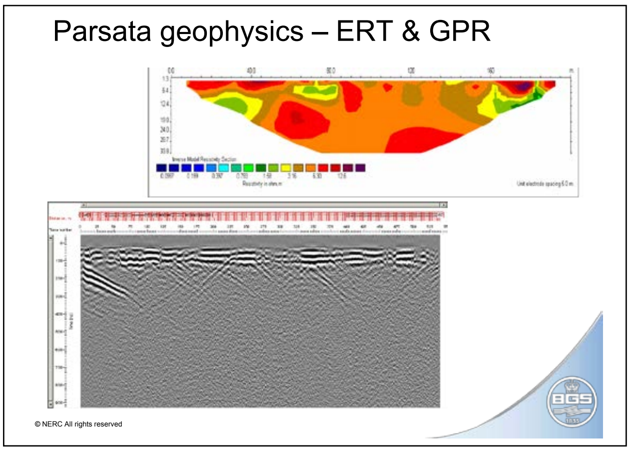
[Modified after Gass et al., 1994]

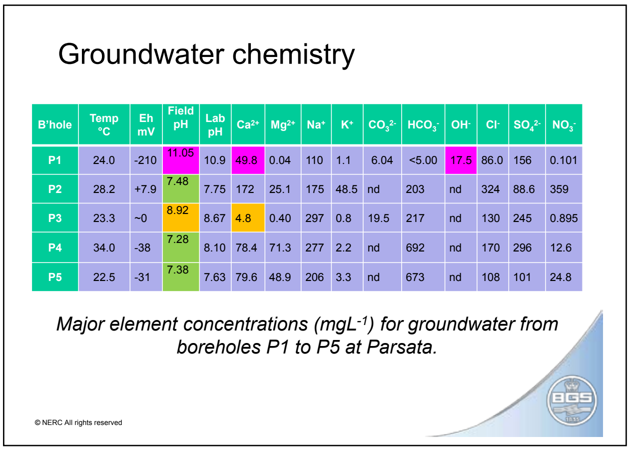
The Circum-Troodos sedimentary sequence

Age (Ma)			Formation	Lithology
2.0	Pleistocene		'Fanglomerate' Apalos Kakkaristra Athalassa	Conglomerates and Sandstones, Calcarenite, Sandstones, Conglomerates
5.2	Pliocene			
			Nicosia	Marls, Silts, Muds, Sandstones, Conglomerates
23.3	Miocene		Kalavasos	Evaporites
		Upper	Pakhna	Reefal and Bioclastic Limestone
		Middle		Pelagic Chalks, Marls, Calcarenites, Conglomerates
		Lower		Reefal and Bioclastic Limestone
35.4	Oligocene		Upper Lefkara	Pelagic Chalk and Marls
56.5	Eocene		Middle Lefkara	Massive Pelagic Chalks
65.0	Palaeocene			Pelagic Chalks, Replacement Chert
74.0	Maastrichtian		Lower Lefkara	Pelagic Chalks
83.0	Campanian		Kanaviou	Volcaniclastic Sandstones, Bentonite
90.4	Turonian		Perapedhi	Umbers, Radiolarites
Ophiolitic Basement				





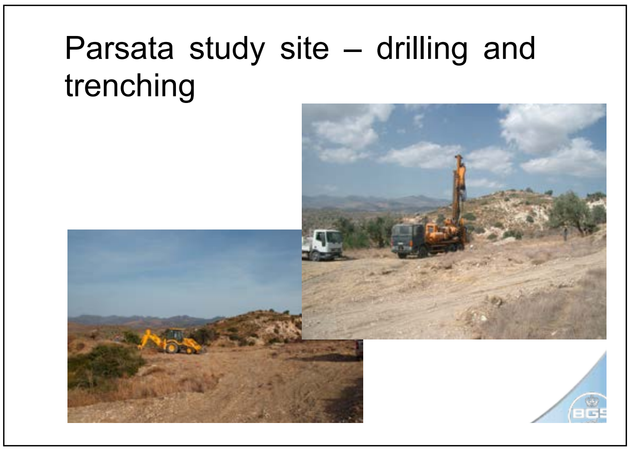


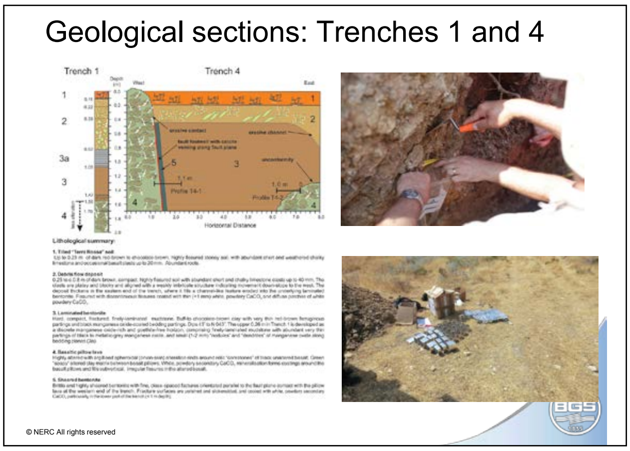


Groundwater chemistry

B'hole	Temp °C	Eh mV	Field pH	Lab pH	Ca^{2+}	Mg^{2+}	Na^{+}	K^{+}	CO_3^{2-}	HCO_3^{-}	OH^{-}	Cl^{-}	SO_4^{2-}	NO_3^{-}
P1	24.0	-210	11.05	10.9	49.8	0.04	110	1.1	6.04	<5.00	17.5	86.0	156	0.101
P2	28.2	+7.9	7.48	7.75	172	25.1	175	48.5	nd	203	nd	324	88.6	359
P3	23.3	~0	8.92	8.67	4.8	0.40	297	0.8	19.5	217	nd	130	245	0.895
P4	34.0	-38	7.28	8.10	78.4	71.3	277	2.2	nd	692	nd	170	296	12.6
P5	22.5	-31	7.38	7.63	79.6	48.9	206	3.3	nd	673	nd	108	101	24.8

Major element concentrations (mgL^{-1}) for groundwater from boreholes P1 to P5 at Parsata.




Geological sections: Trenches 1 and 4
Trench 1
Trench 4
Horizontal Distance
Lithological summary
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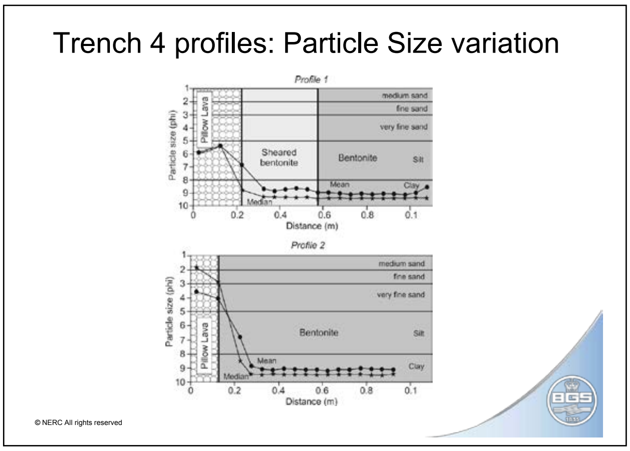
Trench 4 profiles: Particle Size variation
Profile 1
Profile 2
Particle size (phi)
Pillow Lava
Sheared bentonite
Bentonite
medium sand
fine sand
very fine sand
Silt
Clay
Mean
Median
Distance (m)
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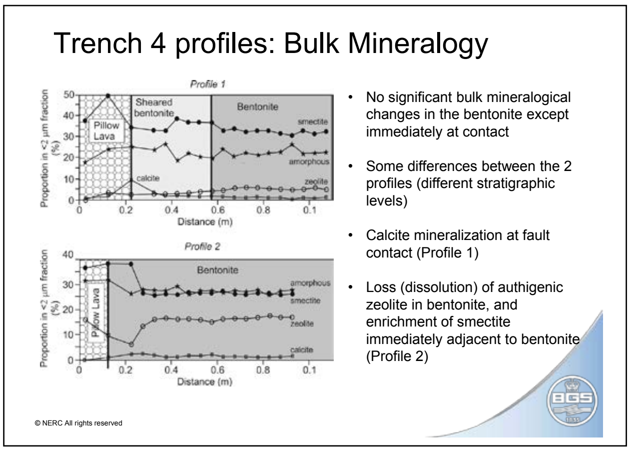
Trench 4 profiles: Bulk Mineralogy
Profile 1
Profile 2
Proportion in <2 µm fraction (%)
Pillow Lava
Sheared bentonite
Bentonite
smectite
amorphous
zeolite
calcite
Distance (m)
• No significant bulk mineralogical changes in the bentonite except immediately at contact
• Some differences between the 2 profiles (different stratigraphic levels)
• Calcite mineralization at fault contact (Profile 1)
• Loss (dissolution) of authigenic zeolite in bentonite, and enrichment of smectite immediately adjacent to bentonite (Profile 2)
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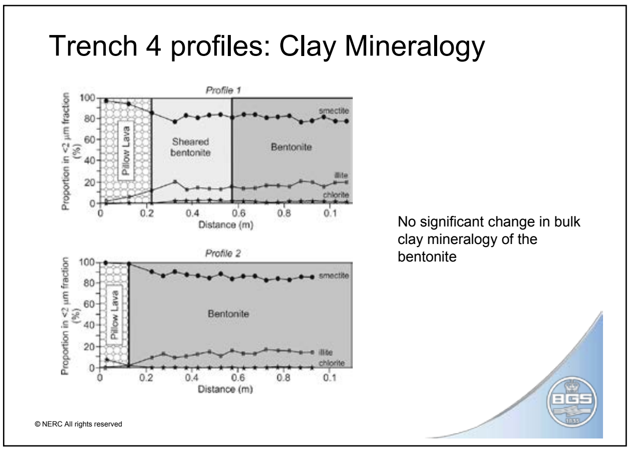
Trench 4 profiles: Clay Mineralogy
Profile 1
Profile 2
Proportion in <2 µm fraction (%)
Pillow Lava
Sheared bentonite
Bentonite
smectite
illite
chlorite
Distance (m)
No significant change in bulk clay mineralogy of the bentonite
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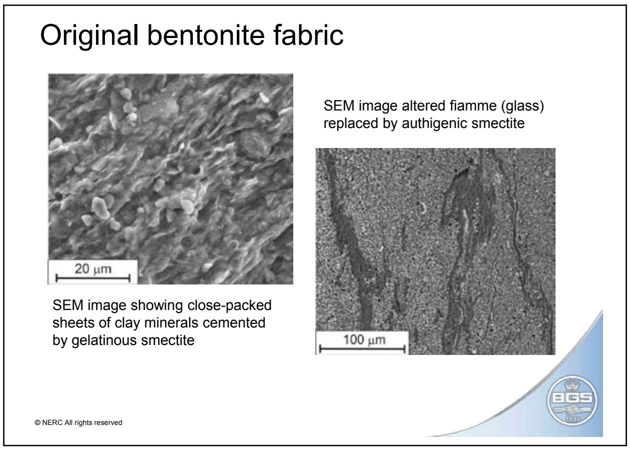
Original bentonite fabric
SEM image altered fiamme (glass) replaced by authigenic smectite
20 µm
100 µm
SEM image showing close-packed sheets of clay minerals cemented by gelatinous smectite
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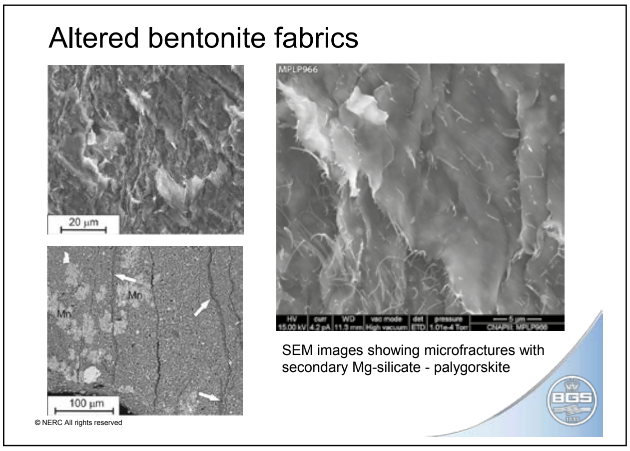
Altered bentonite fabrics
MPLP966
20 µm
Mn
100 µm
SEM images showing microfractures with secondary Mg-silicate - palygorskite
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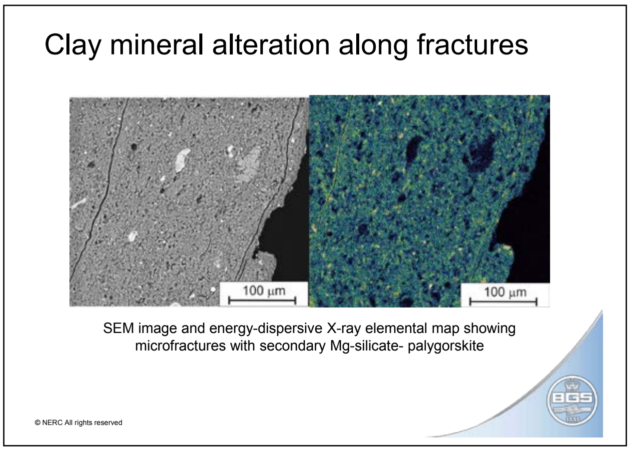
Clay mineral alteration along fractures
100 µm
100 µm
SEM image and energy-dispersive X-ray elemental map showing microfractures with secondary Mg-silicate- palygorskite
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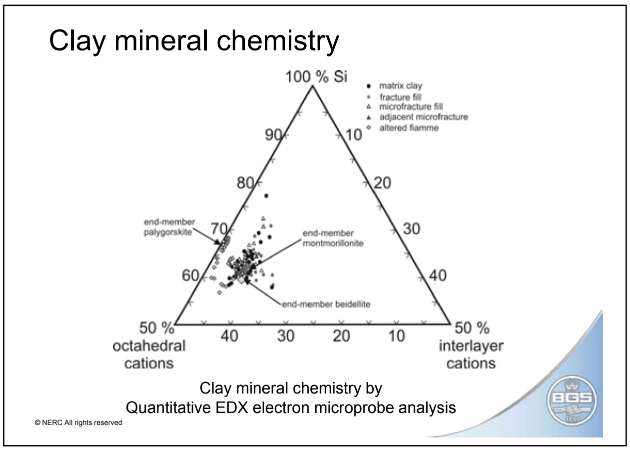
Clay mineral chemistry
100 % Si
matrix clay
fracture fill
microfracture fill
adjacent microfracture
altered fiamme
end-member palygorskite
end-member montmorillonite
end-member beidellite
50 % octahedral cations
50 % interlayer cations
Clay mineral chemistry by
Quantitative EDX electron microprobe analysis
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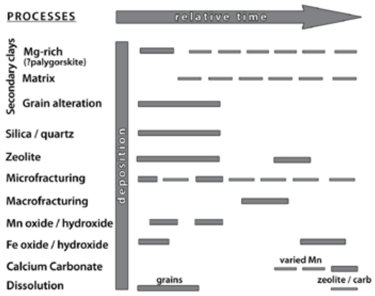
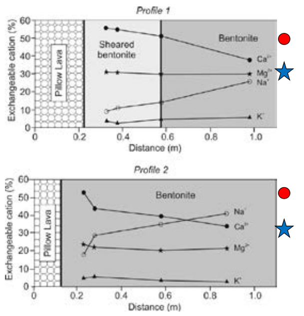
2. Milodowski, Norris & Alexander: Montmorillonite alteration by alkaline groundwater in Cyprus: a natural analogue for bentonite reaction in low alkali cement pore fluids.

Poly-episodic mineralization sequence



Alteration of bentonite exchangeable cations

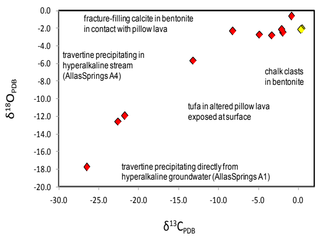


Overall change from Na-dominated exchangeable cation chemistry in the background bentonite analogue (Trench 5) to Ca-dominated or Ca- and Mg-dominated exchangeable cation chemistry in the bentonite analogue in Trench 4

Close proximity of the bentonite in Trench 4 to the PLV is consistent with a model whereby $Ca(OH)_2$-rich groundwater penetrating the bentonite from the underlying PLV, with Ca (and Mg) exchanging with (and displacing) Na in the original bentonite

Background bentonite: Na ● Ca ★

Stable isotopes

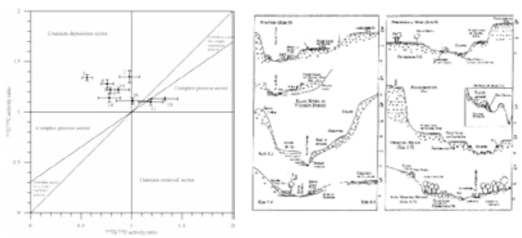


(i) Red symbols – background data from Alexander et al. (2012)

(ii) Blue= calcite mineralisation associated with fractures in Peraphedi Formation bentonite close to the contact with UPL, Trench 4;

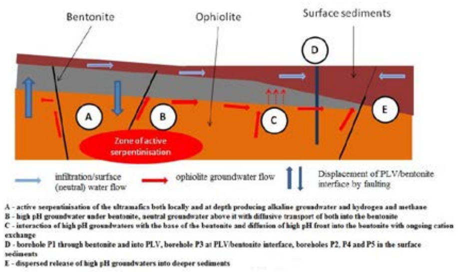
(iii) Yellow symbols – typical detrital micritic limestone or chalk clasts of typical Lefkara Formation chalks included within the bentonitic debris flow lin Parsata Trench 4

Time constraints



- Geomorphological evidence suggests that groundwater flow was initiated in the Troodos Massid ~0.8 Ma ago, and probably at a similar time in the Parsata area
- Natural series data indicate ongoing water-rock interaction at Parsata over at least the past 500 Ka

Conceptual model



Conclusions

- The results suggest that there has been
  - Cation exchange (Ca for Na) for some distance into the bentonite
  - Minimal volumetric reaction (smectite-to-palygorskite transformation) confined to the base of the bentonite, probably due to alkaline groundwater
- Geomorphology suggests groundwater system operational since 500-800 ka
- U/Th/Ra data rule out early hydrothermal alteration, but indicates overall reaction in bentonite at 10^5 - 10^6 a
- Reaction of bentonite is restricted to close to the UPL contact (and very close to fractures)

Conclusions

- This is probably not due to a limited supply of OH^- as groundwater has been flowing 10^5 - 10^6 a (although with fluctuations)
- Probably more likely due to 'tight' bentonite
  - URL experimental evidence that bentonite resaturation does not occur easily – this could also be the case with natural bentonite (cf. very low moisture contents)
- Palygorskite is a common authigenic clay mineral in "alkaline lake" environments, evaporative soil environments (where alkaline pH implicated), serpentinisation in modern oceanic floor

Conclusions

- It is possible that the smectite-to-palygorskite transformation is a fast reaction (cf. BGS experiments and field evidence), but this may be misleading as neither are representative of repository physico-temporal conditions

2. Milodowski, Norris & Alexander: Montmorillonite alteration by alkaline groundwater in Cyprus: a natural analogue for bentonite reaction in low alkali cement pore fluids.

Conclusions

- That Mg-silicate reaction phases have not been considered in previous waste disposal studies is simply a reflection of the limited range of repository EBS designs and host rock types examined to date. In any case, the precise nature of the secondary phase may well prove less important to the long-term performance of the repository than the limited extent of alteration of the bentonite which points to a EBS which is robust enough to survive for the timescales of concern to the repository safety assessment.

Acknowledgements

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  - *In particular:* Patrick Sellin, Petri Korkeakoski
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- Thanks also to the Geological Survey Department of Cyprus (GSD) for their full support in Cyprus
  - *Including:* Michael Rigas (Hydrogeologist), Dr E.G.Morisseau (Director) and Dr C.A.Constantinou (Head of Hydrology)
- Co-workers *including:*
  - Alistair Pitty (EIA Consulting),
  - Simon Kemp, Charles Gowing, Kay Green, Melanie Leng, Ian Miller, Ian Mounteney, Dan Parkes, Jeremy Rushton, Hilary Sloane, Doris Wagner, (BGS)
  - Gus MacKenzie and Suzie Hardie (SUERC, East Kilbride)

Thank you for your attention

AMPHOS 21

Scientific and strategic environmental consulting

Revisiting the bentonite deposits from Cabo de Gata

David Arcos and Jordi Bruno

20 YEARS FLOWING

Introduction

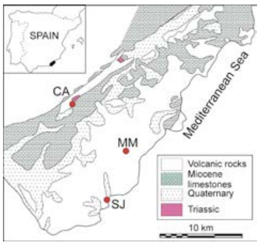
- In PA exercises of KBS-3 repositories it is assumed that the bentonite barrier will be saturated a few years after closure of the repository
.
- This is definitively true for the average behaviour of the clay barriers, but there is a possibility that some portions of the repository with lower flow fields will remain unsaturated for longer periods.
- In this event, the unsaturated bentonite buffer will be exposed to a heat transient for longer periods under dry conditions.
- It is not known to which extent the heat transient will impose mineralogical changes that will alter the hydraulic conditions of the buffer and if these changes are reversible once the buffer is resaturated

Introduction

- In this context, it would be very interesting to investigate these processes in the temperature and time regimes expected in the repository. These conditions can only be attained in natural formations of bentonite, which have been exposed to heat transients.
- In the late 90's, we were deeply involved in the Natural Analogue programmes of Enresa and SKB .
- One of the sites investigated were the bentonite deposits of Cabo de Gata (Almeria) in southwest Spain where the clay deposits have been originated in a post-volcanic environment
- In that site there are a number of locations where bentonite has been exposed to a temperature transient around 120 ºC and therefore there is a possibility to study the mineral alterations that had occurred and more importantly the reversibility of these alterations once the bentonite became saturated

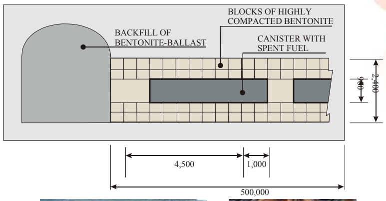
Bentonites from Cabo de Gata (SE Spain)

Alteration of volcanic rocks (white tuffs) from the volcanic belt of Cabo de Gata.



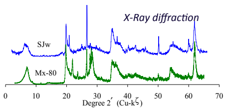
Bentonites from Cabo de Gata (SE Spain)

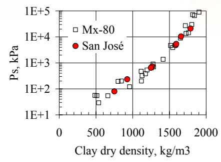
- Bentonites from Cabo de Gata were selected in the former ENRESA NF concept.
- A large number of experiments (FEBEX) were designed with bentonite from Cabo de Gata.



Comparison against MX-80 (effect of high salinity water)

Karnland et al. (2003). Presented at MRS

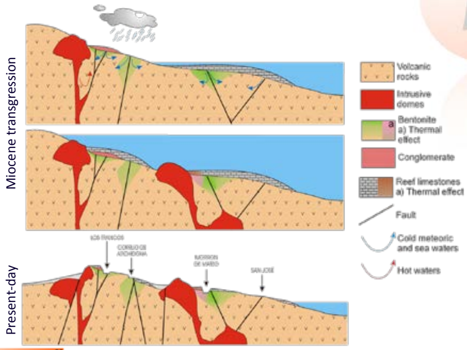




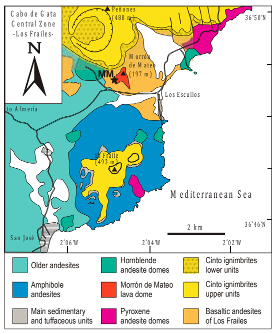
Swelling pressure

Despite some differences in accessory mineral composition, both materials are dominated by dioctahedral smectite with practically the same layer charges, measured as CEC, and the resulting physical properties are similar.

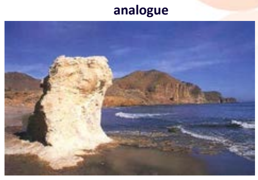
Geologic evolution of Cabo de Gata (SE Spain)



The Morrón de Mateo bentonite



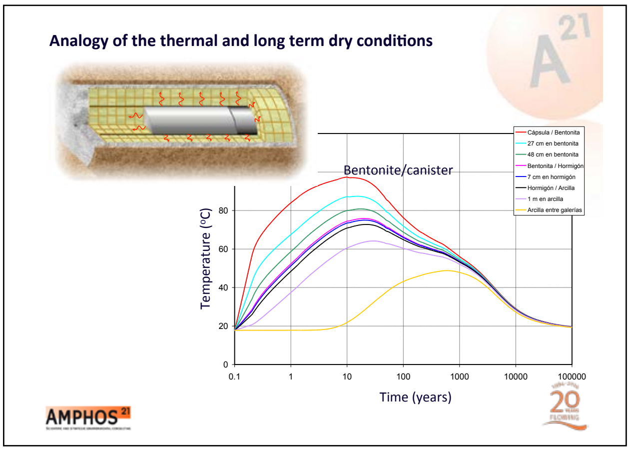
BARRA PROJECT: Bentonite barrier analogue

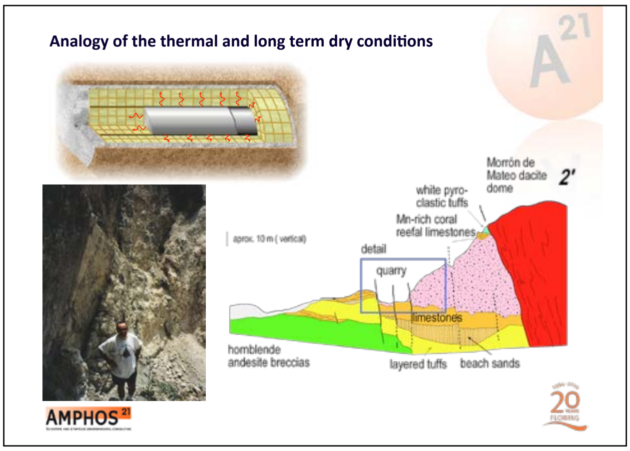


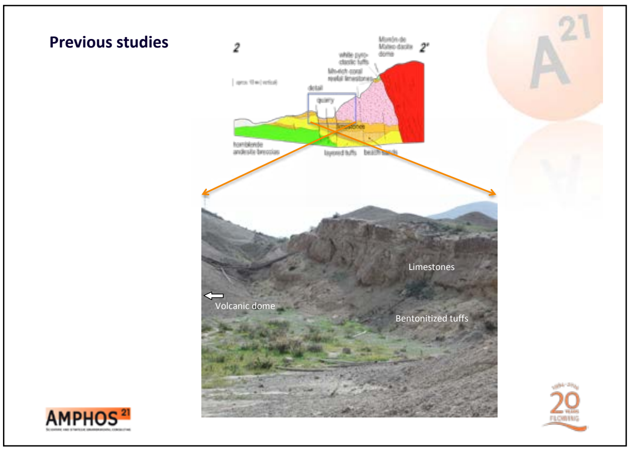
Participants

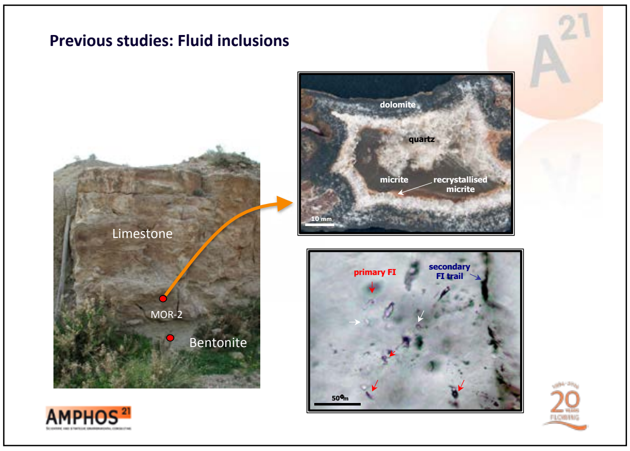
ENRESA: CIEMAT, CSIC, Univ. Granada, Amphos 21
SKB: Clay Technology AB
IRSN: ERM
RAWRA: Charles Univ.

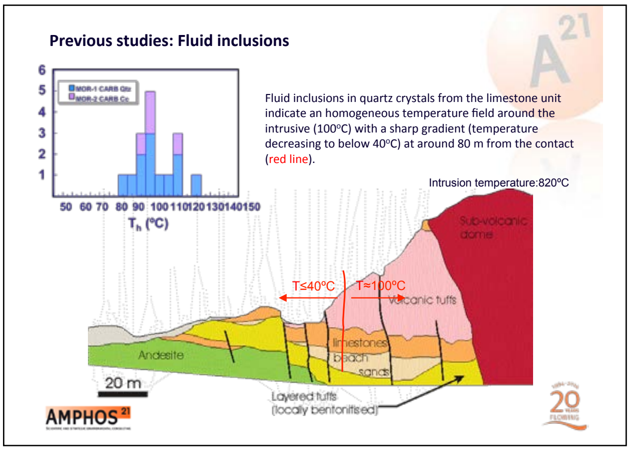

The Morrón de Mateo bentonite
Near the small village of La Isleta del Moro
AMPHOS 21

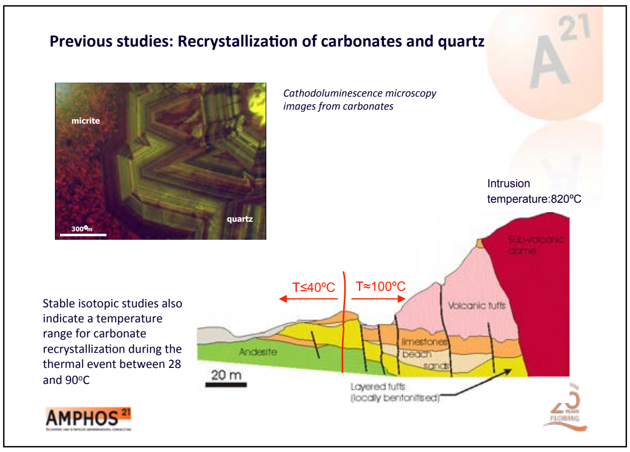

Analogy of the thermal and long term dry conditions
Bentonite/canister
Temperature (ºC)
Time (years)
0.1
1
10
100
1000
10000
100000
0
20
40
60
80
AMPHOS 21


Analogy of the thermal and long term dry conditions
Morrón de Mateo dacite dome
2'
white pyro-clastic tuffs
Mn-rich coral reefal limestones
aprox. 10 m (vertical)
detail
quarry
limestones
hornblende andesite breccias
layered tuffs
beach sands
AMPHOS 21


Previous studies
2
2'
Limestones
Volcanic dome
Bentonitized tuffs
AMPHOS 21


Previous studies: Fluid inclusions
Limestone
MOR-2
Bentonite
dolomite
quartz
micrite
recrystallised micrite
primary FI
secondary FI trail
AMPHOS 21


Previous studies: Fluid inclusions
MOR-1 CARB Qtz
MOR-2 CARB Cc
50 60 70 80 90 100 110 120 130 140 150
T_h (ºC)
Fluid inclusions in quartz crystals from the limestone unit indicate an homogeneous temperature field around the intrusive (100ºC) with a sharp gradient (temperature decreasing to below 40ºC) at around 80 m from the contact (red line).
Intrusion temperature:820ºC
Sub-volcanic dome
T≤40ºC
T≈100ºC
Volcanic tuffs
limestones
beach sands
Andesite
20 m
Layered tuffs (locally bentonitised)
AMPHOS 21


Previous studies: Recrystallization of carbonates and quartz
micrite
quartz
Cathodoluminescence microscopy images from carbonates
Intrusion temperature:820ºC
Sub-volcanic dome
T≤40ºC
T≈100ºC
Volcanic tuffs
limestones
beach sands
Andesite
20 m
Layered tuffs (locally bentonitised)
Stable isotopic studies also indicate a temperature range for carbonate recrystallization during the thermal event between 28 and 90ºC
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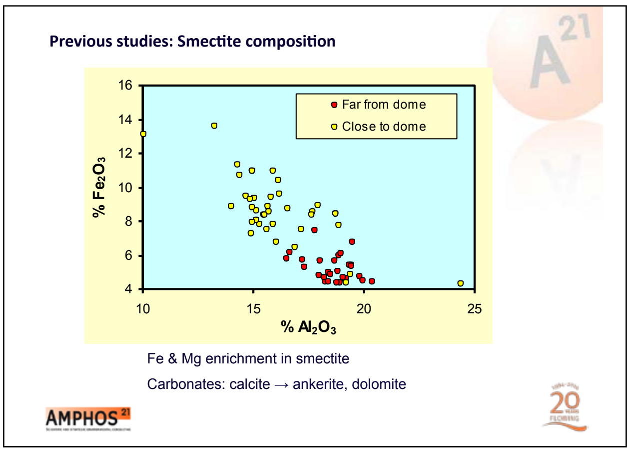
Previous studies: Smectite composition
Far from dome
Close to dome
% Fe_2O_3
% Al_2O_3
4
6
8
10
12
14
16
10
15
20
25
Fe & Mg enrichment in smectite
Carbonates: calcite → ankerite, dolomite
AMPHOS 21

Conclusions

- Bentonite from Morrón de Mateo is a natural analogue of:
  - the thermal effect, with temperatures near 100ºC close to intrusion, and
  - long term dry conditions (emerged conditions since Miocene times)
- Chemical changes occurred in the bentonite, as well as to the other lithologies of the area (mainly replacement of Fe and Mg)
- The iron replacement in the bentonite buffer could also bring some light about the potential effects of canister-buffer interactions
- Therefore we propose to complement previous investigations by sampling bentonite closer to the thermal intrusion, establishing carefully the thermal gradients and doing a mineralogical and chemical characterization of the changes occurred during the thermal period.

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Preliminary study on Korean bentonite to provide the basis for use of natural analogues in supporting safety cases for radwaste disposal

Tae-Jin Park, Min-Hoon Baik, Seung-Yeop Lee, Geon-Young Kim, and Kyungsu Kim
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Phone: 042-868-2049

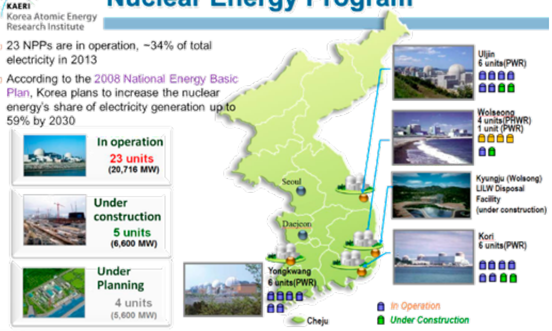
14th NAWG Workshop, Olkiluoto, Finland
June 2015

한국원자력연구원 KAERI Korea Atomic Energy Research Institute

Nuclear Energy Program

KAERI Korea Atomic Energy Research Institute

- 23 NPPs are in operation, ~34% of total electricity in 2013
- According to the 2008 National Energy Basic Plan, Korea plans to increase the nuclear energy's share of electricity generation up to 59% by 2030



2

Spent Fuel Storage at NPPs

- Spent fuels stored at reactors sites reached 12,629 tons as of the end of 2012, which consist of 5,744 tons of PWR and 6,885 tons of CANDU.
- All nuclear sites will reach their capacity within this decade if no further changes are made.

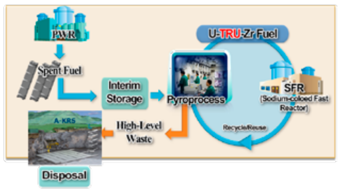
* As of Sep. 2012 (Unit : t)

Site	Capacity	Saturation	Accum.
Kori (6)	2,691	2016	1,981
Yongkwang (6)	3,318	2019	2,061
Uljin (6)	2,328	2021	1,702
Wolseong (5)	9,660	2022	6,885
Total	17,997		12,629 (70%)

3

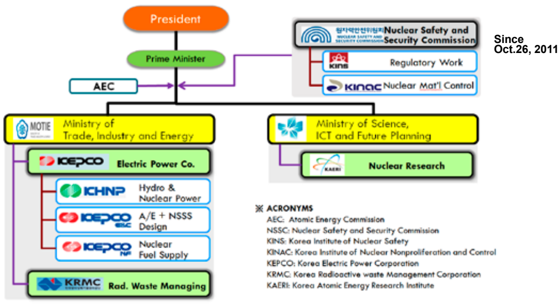
R&Ds on Back-end Nuclear Fuel Cycle

- KAERI is developing closed fuel cycle with the combination of deep geological disposal system, pyroprocessing processes and sodium-cooled fast reactors.
- Based on enhanced safety and nonproliferation principles, the closed fuel cycle program helps to
  - ✓ minimize waste generation up to 1/20,
  - ✓ reduce disposal area by a factor of 100,
  - ✓ shorten the radiotoxicity up to 1/1,000,
  - ✓ and maximize energy production.



4

Nuclear Energy Organization



※ ACRONYMS
AEC: Atomic Energy Commission
NSSC: Nuclear Safety and Security Commission
KINS: Korea Institute of Nuclear Safety
KINAC: Korea Institute of Nuclear Nonproliferation and Control
KEPCO: Korea Electric Power Corporation
KRMC: Korea Radioactive waste Management Corporation
KAERI: Korea Atomic Energy Research Institute

5

Natural Analogue in Safety Case

KINS, a Korean regulatory body, recently starts requiring a safety case report for licensing a radioactive waste repository.

NSSC/KINS is plan to include natural analogues (NA) as complementary consideration, into the safety case for the HLW disposal.

KAERI is carrying out safety case development for a Korean HLW disposal system (e.g., A-KRS).

NA study for Korean HLW disposal system should be carried out.

6

Natural Analogue Study in Korea

- **Feasibility study (2000)**
  - KAERI and KIGAM jointly investigated the feasibility of NA studies focusing on uranium ore deposits in Korea
  - The distribution and characteristics of Korean uranium ore deposits
    - → A potential site for NA studies → Deokpyung-ri uranium ore deposit
    - → No further study after this → only small lab-scale studies
- **ASARR project from 1996 to 1999**
  - KAERI participated in ASARR project at Koongarra uranium ore body, Australia
  - Research items
    - Development of thermodynamic sorption model which can apply to the sorption of U onto soils from Koongarra site (Jung et al., J. Radioanl. Nucl. Chem. 242, 405-412, 1999)
    - Development of a reactive transport model and its application and validation (Keum et al., Computers & Geosciences, 29, 431-445, 2003)

⇨ In terms of radioactive waste disposal, no large scale NA study in Korea

7

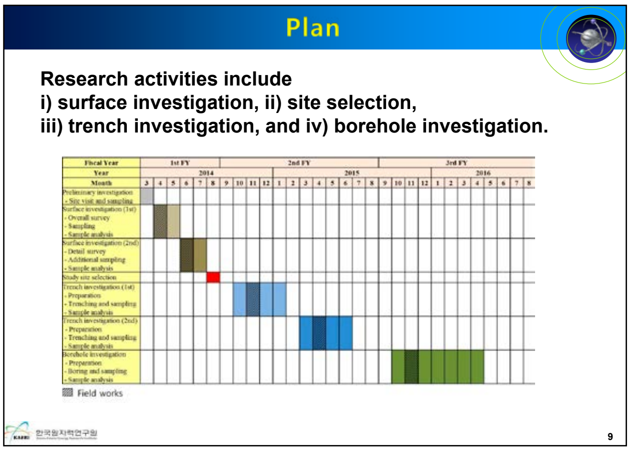
Bentonite Natural Analogue Project

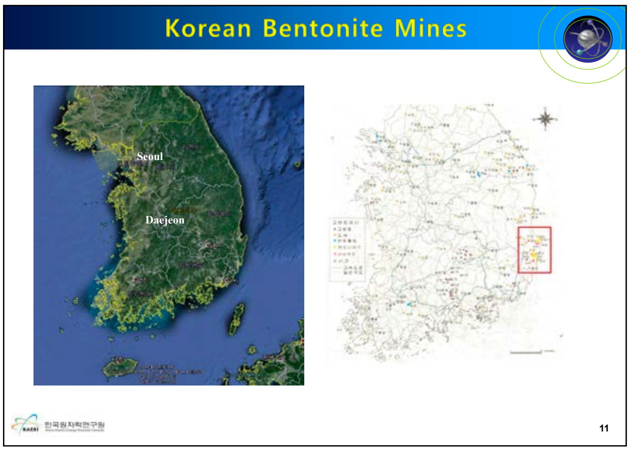
Started (2014) as a subtask of "Development of technology for enhancing the safety of radioactive waste disposal" project funded by NSSC.

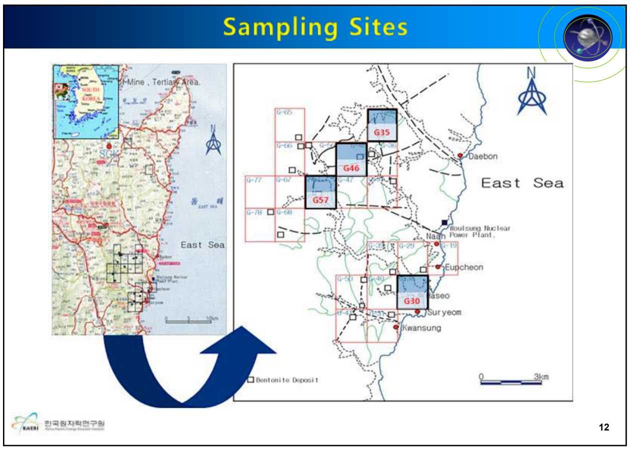
- Natural analogue research for the long-term safety assessment of HLW repository: Degeneration and hydraulic/chemical properties of bentonite.
- Safety verification technology applicable for providing the regulation guidelines and technical standards
- Long-term safety assessment of radioactive waste repository by means of natural analogue research (e.g., bentonite NA)

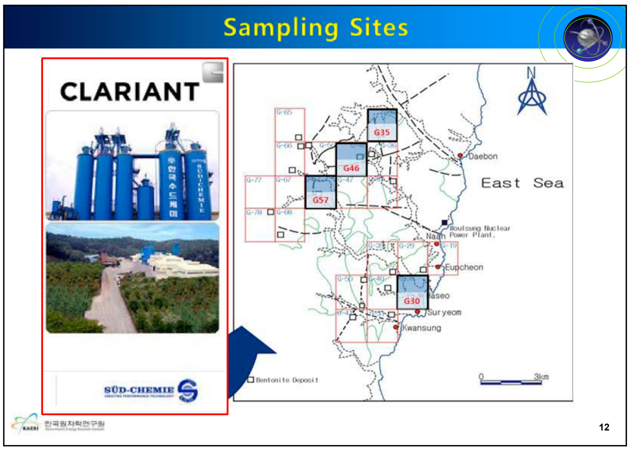
8

4. Park, Baik, Lee, Kim & Kim: Preliminary study on Korean bentonite to provide the basis for use of natural analogues in supporting safety cases for radwaste disposal.









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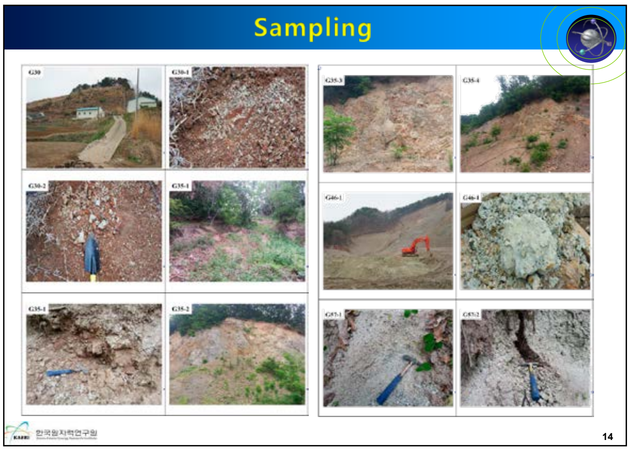

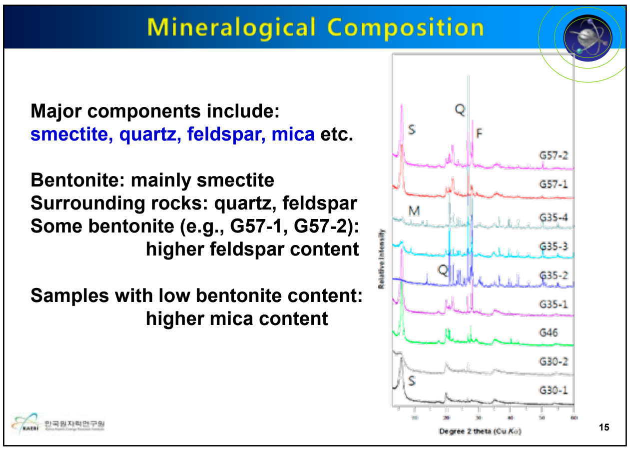

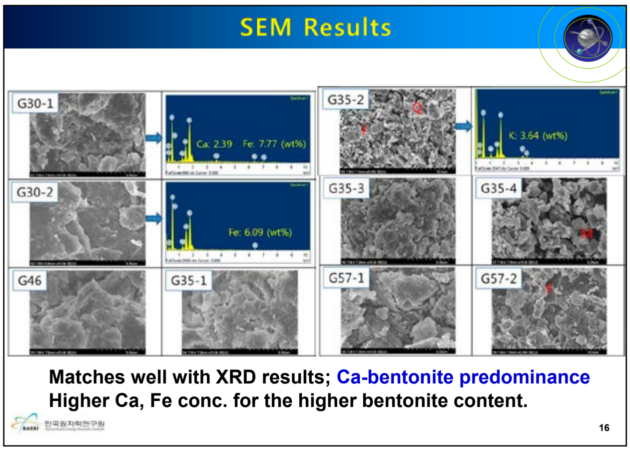

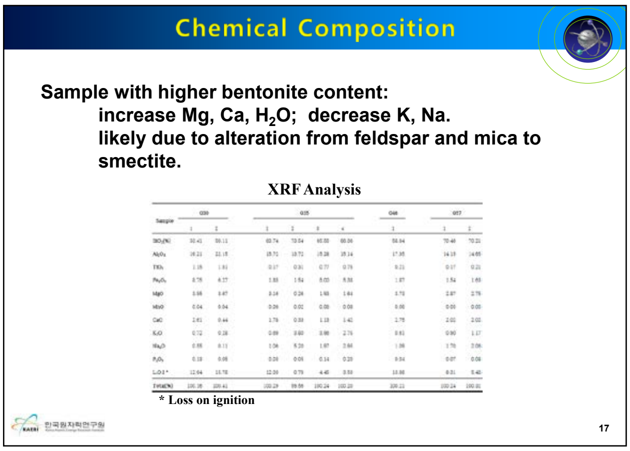

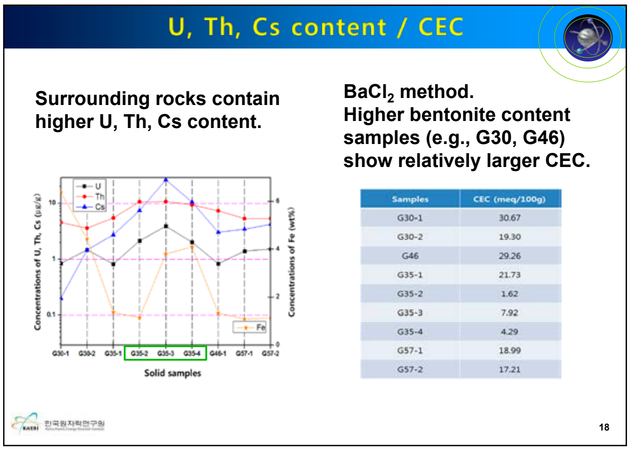

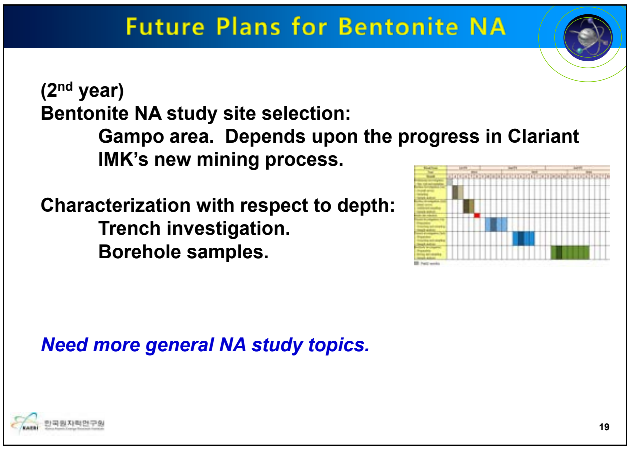

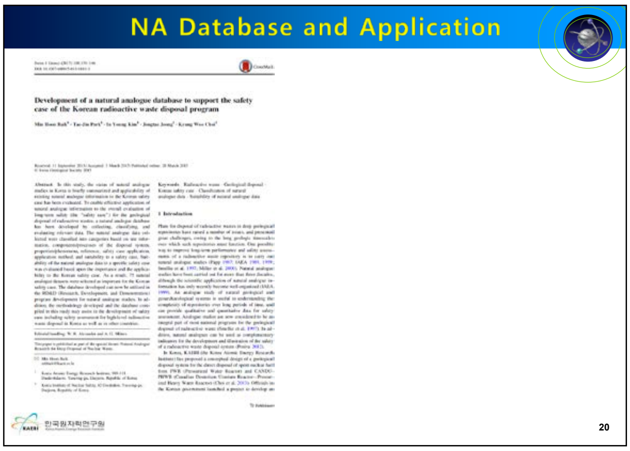

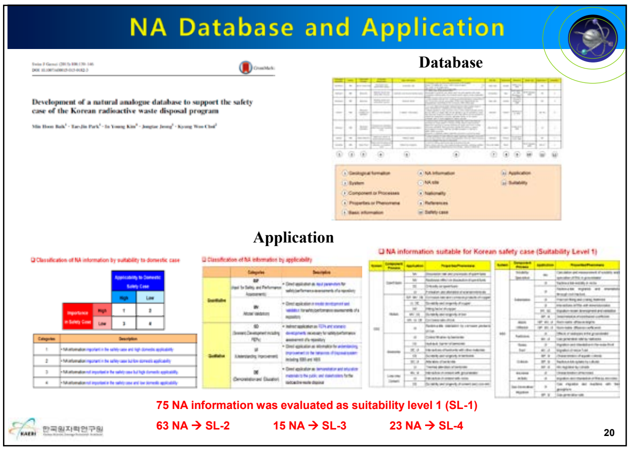

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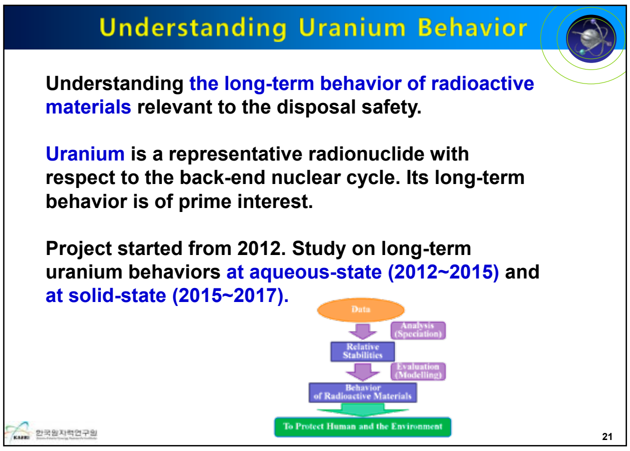
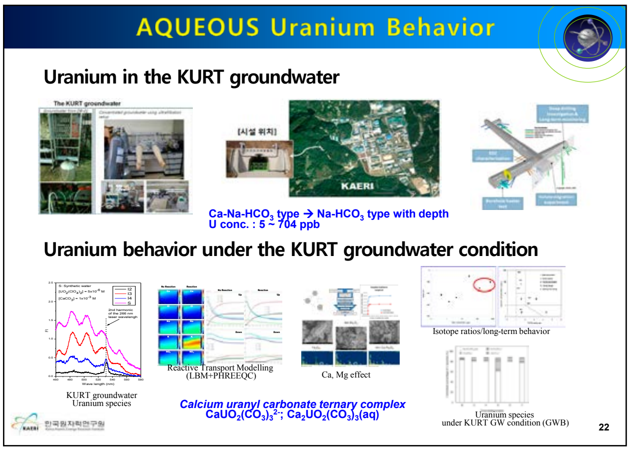
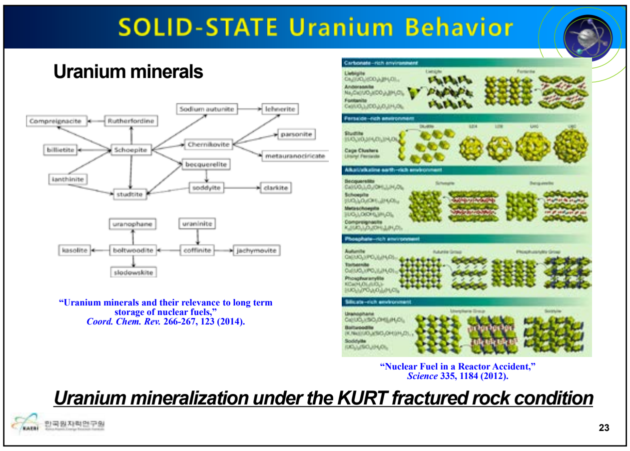
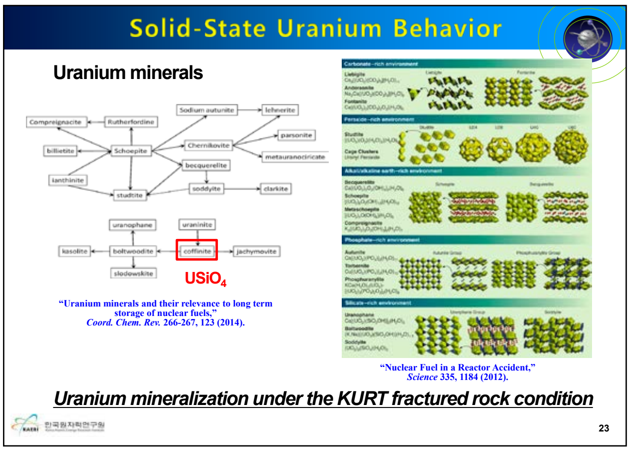
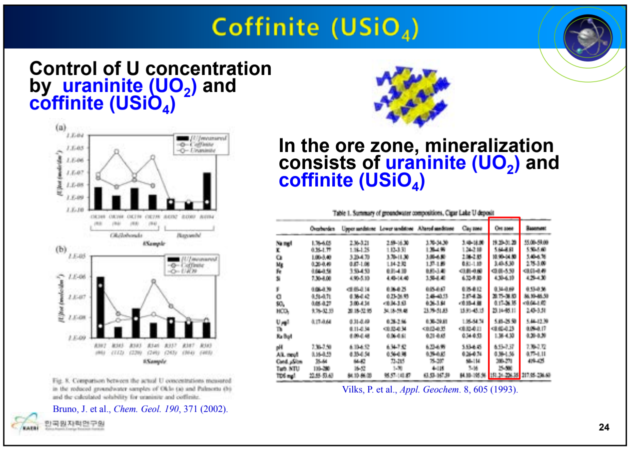
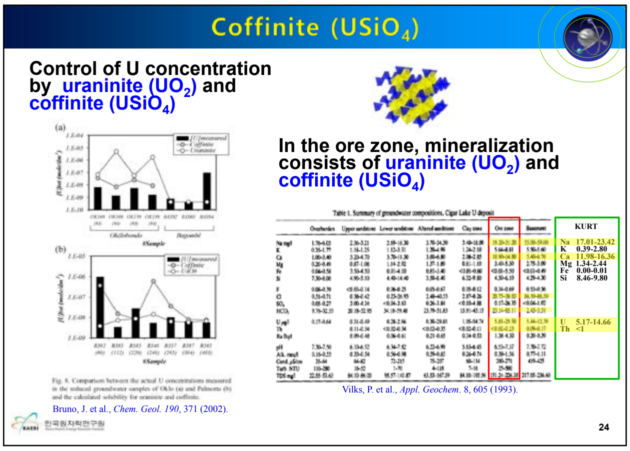


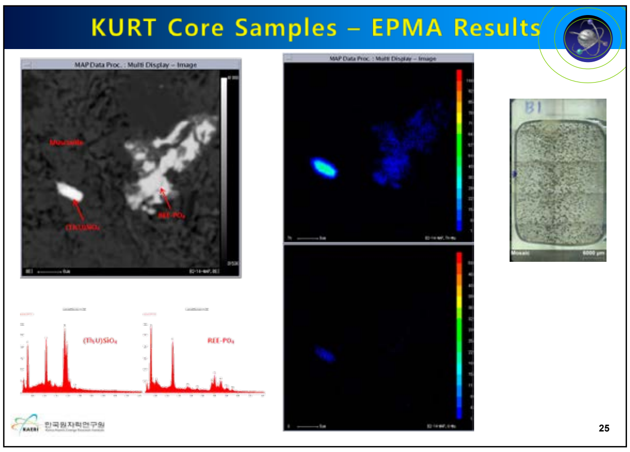


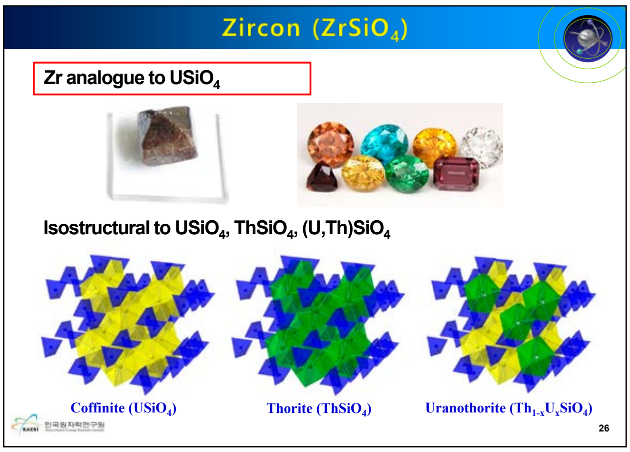








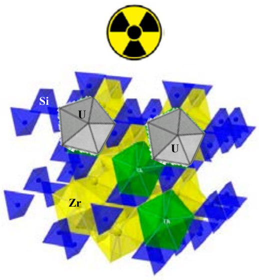




4. Park, Baik, Lee, Kim & Kim: Preliminary study on Korean bentonite to provide the basis for use of natural analogues in supporting safety cases for radwaste disposal.

Metamictization

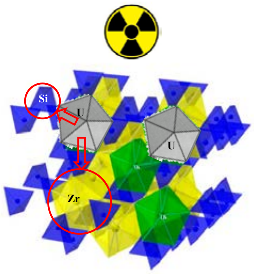
A natural process resulting in the loss of a mineral's crystalline structure due to the radioactivity within it



27

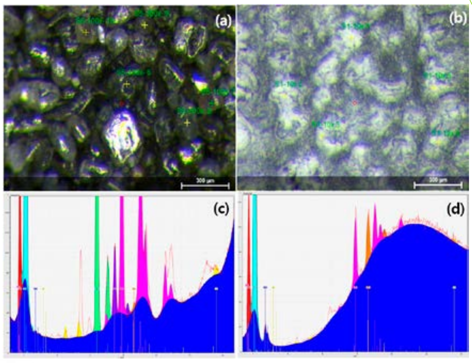
Metamictization

A natural process resulting in the loss of a mineral's crystalline structure due to the radioactivity within it



27

메타믹트화 작용



28

Summary

In Korea, the necessity for NA study is increasing.

Preliminary Korean bentonite natural analogues (NA) study has been performed. The bentonite NA study site is selected, but the study is waiting for funding.

On-going efforts to understand the long-term behavior of uranium in aqueous and solid-state phase is introduced.

Outcome from this work will be fundamentally used to provide information relevant to the safety case development for HLW disposal.

Also, used to communicate among stakeholders (e.g., expert, non-nuclear expert, and the public) on disposal safety.

29

Acknowledgements

- Korea Institute of Nuclear Safety (KINS) through Nuclear Safety R&D Program of Nuclear Safety and Security Committee (NSSC) of South Korea.

- Nuclear R&D Program of Ministry of Science, ICT & Future Planning (MSIP) of South Korea.

30

5. Fujii, Yamakawa, Owada, Nishimura, Sato, Otomo, Nakata, Arcilla, Satoh, Nagakawa & Alexander: Alkaline alteration processes of bentonic sediment in the Philippines natural analogue and perspective of the survey of active type.

Alkaline Alteration Processes of Bentonic Sediment in the Philippines Natural Analogue and Perspective of the Survey of Active Type

The 14th NAWG Workshop

13th- 16th June, 2015, Olkiluoto, Finland

○N. Fujii, M. Yamakawa, J. Owada (RWMC, Japan)
M. Nishimura (Obayashi Co., Japan)
T. Sato (Hokkaido Univ., Japan)
K. Otomo, M. Nakata (Tokyo Gakugei Univ., Japan)
C.A. Arcilla(Univ. of the Philippines, Philippines)
H. Satoh (Mitsubishi Materials Co., Japan)
K. Nagakawa (Kiso-Jiban Consultants Co., Japan)
R. Alexander(Bedrock Geosciences, Switzerland)

This research was initiated within a project to develop an integrated Natural Analogue Program in Japan(FY2013~2014), which was funded by the Ministry of Economy Trade and Industry (METI), Japan.

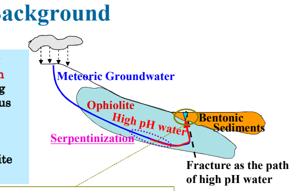
Contents

1

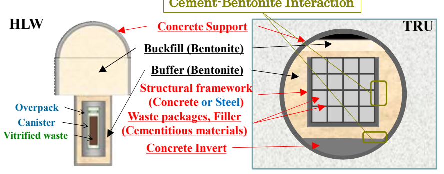
1. Background

The Natural Analogue Concept

- Hypealkaline Groundwater evolved by Serpentinization of Ultramafic rocks in Ophiolite is high pH, high Ca, reducing and 30~40°C (Groundwater is analogous to cement leachate.)
- The NA Concept of Cement-Bentonite Interaction is that Hypealkaline Groundwater have infiltrated Bentonite over a long duration.



Cement-Bentonite Interaction



High-level waste and TRU waste Repository in Japan

2

2. Objectives and NA sites

Objectives

This NA project has field survey in the Philippines and chemical and mineralogical analysis,

- To better understand Long-term stability of Bentonite under the Hyperalkaline Conditions
  - Geochemical properties of Hyperalkaline Groundwater and Environmental Conditions
  - Alteration(Mineralogical evolution) Process of Bentonite in contact with Hyperalkaline Groundwater
  - Reaction time
- To establish comprehensive scenario of Bentonite Reaction

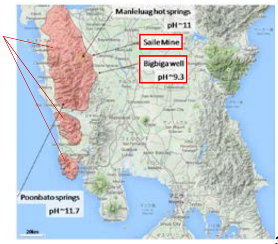
NA sites in Luzon Island

•Zambales Ophiolite in west central area of Luzon in the Philippines

Saile Mine

Outcrop identified 6 Faults in Saile Mine Quarry

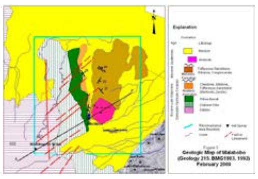
Alternation of a Bentonite bed and Zeolite bed



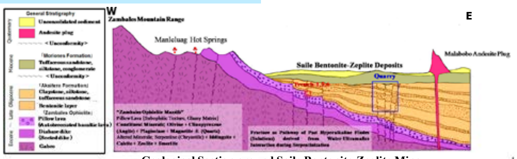
3

3. NA in Saile Mine -Geology and Formation of Bentonite Beds-

- Saile bentonite/zeolite mine is situated at the eastern edge of the Zambales ophiolite. Manleluag Hot Springs (pH 11) exist about 2.5 km southwest of the quarry, situated in the gabbro of the Zambales ophiolite.
- The Aksitero formation, which includes bentonite layer, directly overlies on volcanic basaltic breccias and pillow lava at the top of Zambales ophiolite and is deposited horizontally.
- Bentonite is supplied by primary materials of volcanoclastics (pyroclastics such as pumice, glass, ash and tuff) and have been obviously formed by diagenesis processes.



Geological map around Saile mine

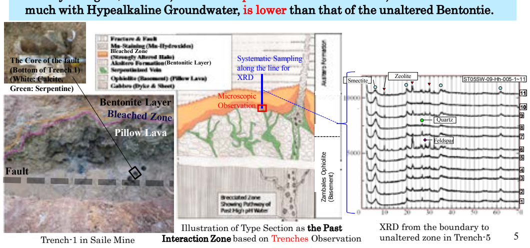


Geological Section around Saile Bentonite-Zeolite Mine

4

3. NA in Saile Mine –Mineralogy of Bleached Zone in Bentonite-

- We found the obvious Geological and Mineralogical Evidences (Serpentine, Calcite, Mn-hydroxide and K-feldspar etc.) that Hyperalkalline Groundwater had infiltrated into bentonite.
- The bleached zone of bentonite distributes in pararell with the same distance of 30-40cm along the wavelike surface of the basaltic pillow lavas and mineral composition of bleached zone is not different from that of unaltered bentonite zone except for the part of heterogeneous calcite.
- The porosity of the Bleached Zone is smaller than that of the non-altered Bentontie (the density is higher). That is, Mass Transport of the Bleached Zone, which had interacted much with Hypealkaline Groundwater, is lower than that of the unaltered Bentontie.



Trench-1 in Saile Mine

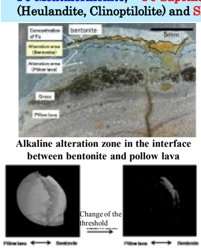
Illustration of Type Section as the Past Interaction Zone based on Trenches Observation

XRD from the boundary to unaltered zone in Trench-5

5

3. NA in Saile mine –Microscopic Observation of the Interaface

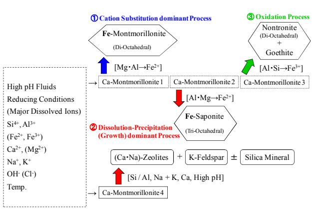
- The Alkaline Alteration Zone had formed by interaction with Hyperalkaline Groundwater was found in the contact interface between bentonite and basaltic pillow lava, but the width is a maximum of 5 mm.
- The concentarton band of Fe, which is higher density by clogging nontronite and goethite, was found in the front of the alkaline alteration zone.
- The Alkaline Alteration zone of Bentonite formed by cation substitution reaction, dissolution-precipitation reaction and reduction-oxidation reaction, and constitutes of Fe-Montmorillonite, Fe-Saponite, Notronite(, Goethite), K-Fldspar, Ca-Zeolites (Heulandite, Clinoptilolite) and Slica Minerals.



Alkaline alteration zone in the interface between bentonite and pollow lava

X-ray CT of the interface

High density part



Mineralogical Evolution under Hyperalkaline groundwater

6

4. Long-term stability of bentonite from NA in Saile mine

① Range and Degree of Bleaced Zone

- The width of bleaced zone in bentonite layer is about 20~40cm. The shape suggests that mass transportation was controlled by diffusion.
- The mineral assemblage, CEC and swelling in the Bleached zone do not differ notably from those in unaltered bentonite except the part of heterogeneous calcite.

② Scale of alkaline alteration and mass transfer

- The alkaline alteration zone had formed by interaction with hyperalkaline groundwater was found in the contact interface, but the width is a maximum of 5 mm. And it constitutes of Fe-type Smectite(Fe-Montmorillonite, Fe-Saponite), Notronite(, Goethite), K-Fldspar, Ca-Zeolites and Slica Minerals.
- The Fe-accumulated band consisted of Nontronite and Goethite was formed in the front of the alkaline alteration zone. The clogging by these minerals prevented mass transfer, and restricted alteration.

③ Fact of the field in a macroscopic viewpoint

- In spite of having been in contact with hyperalkaline water flowing out actively now still on the basement rock with many faults, bentonite deposit has been maintained for a long period beyond the evaluation time of waste disposal.

Because of these facts, the long-term stability of bentonite under highpealkaline condition(pH11) will probably be enough maintained.

7

5. Problems of NA of "Fossil Type"

① The "actually" chemistry of Hyperalkalline Groundwater

- Although we assume hyperalkaline groundwater had reacted in Saile mine is the same as that of Manleluag hot springs (2.5 km southwest from Saile mine), it is imposible to mesure the chemistry of "Fossil Type".

② The sequence of formation of secondary minerals

- The sequence of formation of secondary minerals reacted hypealkaline groundwater is uncertain.
- C-S-H (Calcium Silicate Hydrate), which is often observed in an experiment, is not detected in the NA in Saile mine.

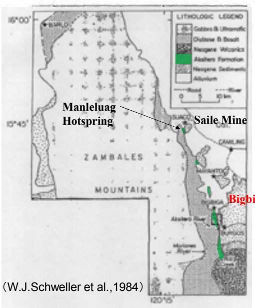
③ Reaction Time

- Althogh we can evaluate the time to stop the hyperalkalline groundwater by dating of TL(Thermo-Luminescence), it is imposible to the reaction time, especially the bigining time to supply hyperalkaline groundwater.

NA site of "Active Type" where environmental condirions(groundwater chemistry etc.) is clearer than the site of "Fossil Type".

8

6. NA in Bigbiga -Geology-



Distribution Map of **Aksitero Formation**, Mangatarem District, Northwst Luzons

NA site at Fish Pond area in Bigbiga

9

6. NA in Bigbiga –NA Survey-

[Geological Survey] : Observation of outcrops along Aksitero river, XRD
[Geophysical exploration] : TEM
[Boring] : DH01~DH10, Core observation, XRD, XRF, Microscopic Analysis(EPMA, FE-SEM)
[Trench] : BBT02~BBT02, Observation of Wall Surface, XRD, XRF
[Groundwater Survey]

- **pH, ORP, Temp., EC, DO, Dissolved Ions, δD, δO**
- **Pumping Test**

Data of Borehole or Well

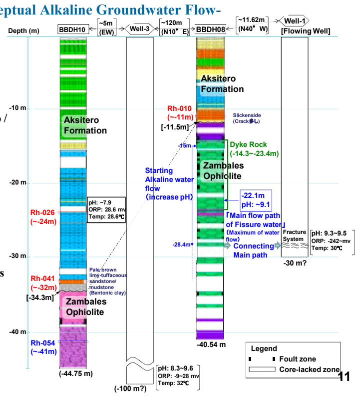
Location	Groundwater Level (GL～)	Depth (GL～)	pH(Temp.℃)	
Well-1	flowing well	-19m	9.01 (29.4)	H25 ORP: -48 mV
	flowing well	-19m	9.52 (29.2)	ORP: 8 mV
Well-2	+0.65m	*-56.75*m	7.58 (32.5)	H25 ORP: 150 mV
Well-3	-3.42m	-100m	8.37 (32.2)	H25 ORP: 28 mV
	-4.05m	*-79.0*m	8.93 (29.2)	ORP: 47.9 mV
DH05	-1.93m	-10.07m	7.05 (29.2)	H25
DH06	-3.1m	-27m	6.16 (29.4)	H25
DH07	-3.2m	-18.8m	6.90 (28.8)	H25
DH08	flowing well	-40.54m	9.16 (31.9)	H25 ORP: 63 mV
DH09	-2.45m	-15.40m	7.13 (29.7)	H25 ORP: 135 mV
DH10	-4.75m	-91.0m *-79.0*m	7.91 (28.6)	ORP: 55 mV

Location of Borehole（DH5~DH10）

10

6. NA in Bigbiga –Conceptual Alkaline Groundwater Flow-

(1) Alkaline grondwater (pH~9.5) is fissure water flowing fracture system developed in micro-gabbro / diabase of Zambales ophiolite.
(2) Alkaline groundwater also flow along the fracture (Slickenside) in Aksitero Fm contacting with Zambales Opiolite (Basement) .
(3) The alkaline groundwater has been interacting with Pale brown limy-tuffaceous sandstone/ mudstone (Bentonic clay) for a long-term.

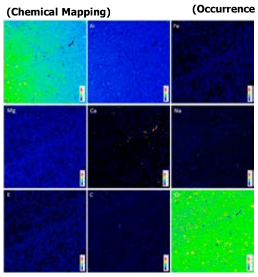


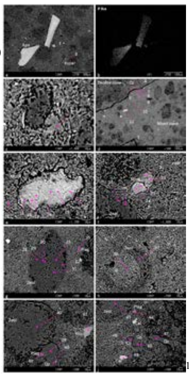
11

6. NA in Bigbiga -Microscopic Observation-

(1)BBDH08-13-Rh-010（Pale brown limy-tuffaceous sandstone/mudstone）

(Chemical Mapping) (Occurrence of minerals)





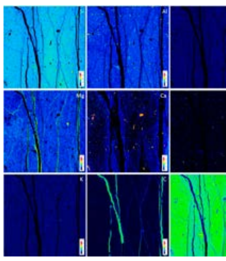
- **K-Feldspar formed by alkaline alteration under low temperature condition**
- **Nontoronite, Ca-Zeolite and Silica mineral as a secondary minaral by Alkaline alteration**

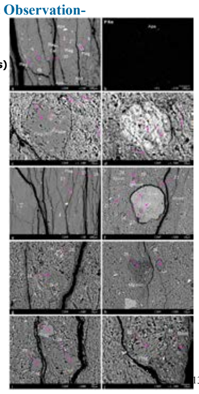
12

6. NA in Bigbiga -Microscopic Observation-

(1)BBDH10-14-Rh-041（Pale brown limy-tuffaceous sandstone/mudstone）

(Chemical Mapping) (Occurrence of minerals)



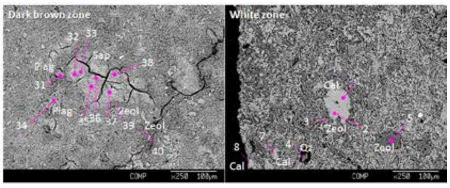


- **K-Feldspar formed by alkaline alteration under low temperature condition**
- **Nontoronite, Ca-Zeolite and Silica mineral as a secondary minaral by Alkaline alteration**

13

6. NA in Bigbiga –Alteration Process under Low Alkaline Condition-

(1)BBDH10-14-Rh-026（Pale brown limy-tuffaceous sandstone/mudstone）



Source Rock : Andesite～Dacite
[Plag<An_{70-50}>], Gm<Glassy>

Montmorillonite → Saponite (+ Qz + Cal) → Clinoptilolite + (Cal)
(Di-OctahedralSmectite) (Tri-OctahedralSmectite)

Dark brown zone | White zone

14

7. Summary

1. **Alkalline alteration of smectite in bentonic sediments at Bigbiga NA site of "Active Type" was observed by EPMA & FE-SEM analysis.**

(1) Fe-Montmorillonite by interlayer cation exchange

(Ca)-Montmorillonite (Di-Octahedral) → Saponite (Tri-Octahedral) (Fe-Saponite) [Ca･Mg→Fe^{2+}]
→ Notronite (Di-Octahedral) [Al･Si→Fe^{3+}]

(1) Dissolution of Montmorillonite / Precipitation

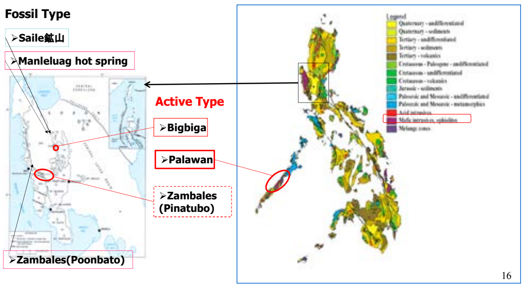
(Ca)-Montmorillonite → Clinoptilolite + K-feldspar + Calcite + Silica mineral

- **Assemblage and Occurrence (Texture) of alkaline alteration minerals is analogous to that of NA of "Fossil Type" at Saile mine.**
- **In laboratory experiments, alkaline alteration is not observe under pH 9.5. But this NA show alkaline alteration of bentonite arises by a long-term interaction under low alkaline conditions.**

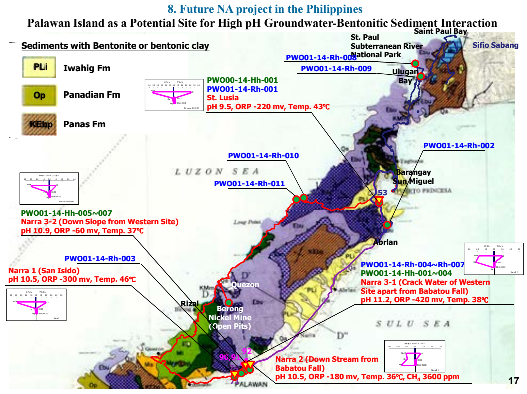
15

8. Future NA project in the Philippines

- Bigbiga is inadequate in respect of the following as NA site of "Active Type". (1)The pH 9.5 is low. (2)The scale of the alteration cannot be understood because the contact is too deep(-10～30m depth).



16



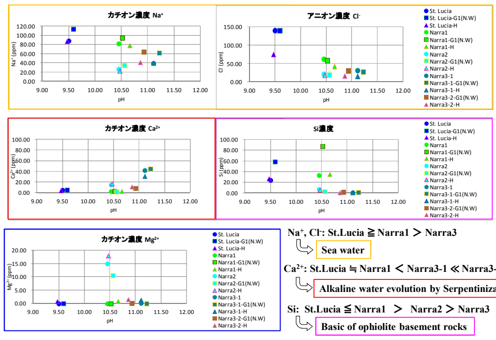
8. Future NA project in the Philippines

Groundwater Chemistry of Palawan and the other survey sites

Site	Poonbato	Manleluag HSp. - M1	Bigbiga Well-1	Bigbiga Well-3	Bigbiga DH10	Palawan Narra-1	Palawan Narra-2	Palawan Narra-3.1	Palawan Narra-3.2	Palawan St. Lucia	low alkali cement leachates (PNC 1997)*
Sample No.	PB-10-KWP-01	M1-11-KWP-01	P-22	P-32	P-32	P-42	P-52	P-62	P-72	P-82	
pH	11.62	10.84	9.52	8.93	7.91	10.52	10.56	11.22	10.93	9.59	11.09
ORP(Eh) [mV]	27.6	-662	8	40	55	-302	-177	-414	-60	-215	-
Temp [℃]	27.6	34.1	29.2	27.8	28.6	45.2	35.5	37.5	37.0	42.7	60
CH_4 [ppm]	>5000	>5000	0 (0~560)	0	0	0	340~3600	0	0 (>5000)	-	-
H_2 [ppm]	50~1320	62	0 (0~130)	0	0	0	0	0	0 (>700)	-	-
Na^+ [ppm]	23.6	1.58	100.6	131.5	94.35	104.7	34.2	61.6	64.0	115.3	43
K^+ [ppm]	6.85	0.28	1.05	1.62	3.07	1.08	1.66	3.52	3.47	3.13	13
Ca^{2+} [ppm]	37.4	23.6	1.34	8.43	1.28	2.95	1.47	44.0	6.88	5.19	16.8
Mg^{2+} [ppm]	0	0.17	0.06	1.05	7.14	0.01	7.04	0.03	0.13	0.08	-
Si^{2+} [ppm]	1.40	11.5	72.3	6.00	35.1	87.6	2.9	1.1	1.7	58.7	
Al^{3+} [ppm]	0	20.3	0.01	0.04	0.45	0.43	0.35	1.79	1.31	0.01	0.3
Cl^- [ppm]	-	16.6	4.50	20.0	17.0	58.0	19.0	27.0	30.0	140.0	-
SO_4^{2-}[ppm]	-	-	48.0	34.0	22.0	3.90	0.52	0.12	0.47	11.0	-
HCO_3^- [ppm]	124.1	73.5	135.6	0.3	12.9	95.5	15.5	0.5	261.2	501.8	-

18

Groundwater Chemistry of Palawan and the other survey sites



Na^+, Cl^-: St.Lucia ≧ Narra1 ＞ Narra3

⮡ Sea water

Ca^{2+}: St.Lucia ≒ Narra1 ＜ Narra3-1 ≪ Narra3-2

⮡ Alkaline water evolution by Serpentinization

Si: St.Lucia ≦ Narra1 ＞ Narra2 ＞ Narra3

⮡ Basic of ophiolite basement rocks

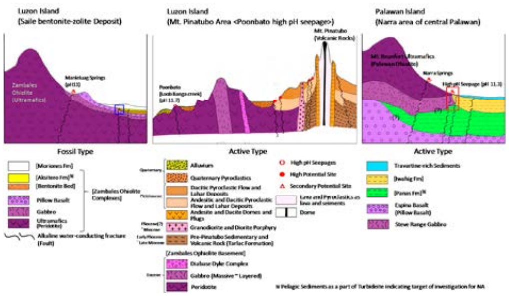
Mg^{2+}: Narra2 ≫ St. Lucia, Narra1, Narra3 ≒ 0

⮡ Narra2 is currently serpentinizing.

19

8. Future NA project in the Philippines

Geological Section of NA sites



20

Thank you for your kind attention.

21

6. Alexander, Takano, Futakuchi & Reijonen: Long-term performance of bentonite (LPB): potential load displacement of the bentonite buffer around radioactive waste containers.

Long-term performance of bentonite (LPB) & Thermal alteration of bentonite (TAB)

Long-term performance of bentonite (LPB): potential load displacement of the bentonite buffer around radioactive waste containers

W.Russell Alexander[1], Hitoshi Takano[2], Katsuhito Futakuchi[2] & Heini M.Reijonen[3]

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2. Dia Consultants Co. Ltd, 2-272-3, Yoshino-Cho,Kita-Ku, Saitama City, 331-0811, Japan
3. University of Helsinki/Saanio & Reikkola, Helsinki, Finland

Introduction (Concept of subsurface disposal)

Subsurface disposal

Wastes settle on the bentonite layer in subsurface disposal repository.
→ It will generate a pressure of about 0.4 MPa.
→ Concern about performance degradation of bentonite due to settlement of the wastes.

Conditions of bentonite

- Bentonite: Kunigel GX
- Dry clay density: 1.6 Mg/m^3
- Thickness of bentonite: 1 m
- Grain size: Maximum 10 mm

Mineral composition (w%)

Smectite	49
Quartz, Chalcedony	17
Feldspar	12.2±10.0
Calcite	1.8
Zeolite	7.2
pyrite	0.88

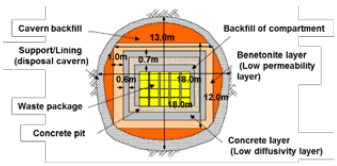
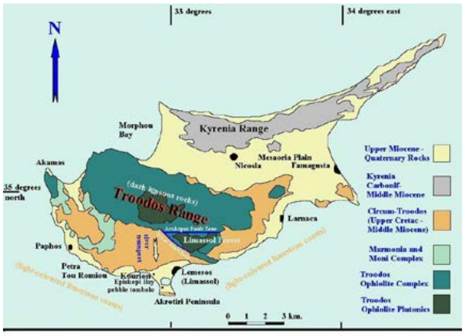


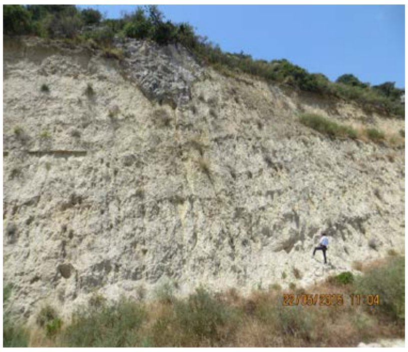
Fig. Subsurface disposal repository scale model (Japan society of civil engineers, 2008)

Overview of the geology of Cyprus



Aphrodite's Rock

Pissouri Jetty



Simple MAA scoring table

Site	Boulder	Caprock	Logistics (incl. site investigation)	Relevance of bentonite	Score
Aphrodite's Rock	2	5	5	7	19
Armenochori	0	5	5	9	19
Choletria	0	4	5	9	18
Episkopi	5	5	5	0	15
Governor's Beach	5	5	2	7	19
Monagroulli quarry (Zoopigi)	0	5	5	10	20
Monagroulli village	5	3	1	9	18
Parsata	2	5	3	10	20
Pierides	3	5	2	9	19

Sampling was planned.....

....then came the rains

LPB reports

W.R.Alexander, H.M.Reijonen & A.Siathas (2015). Long-term performance of bentonite (LPB): potential load displacement of the bentonite buffer around radioactive waste containers. Phase I - site reconnaissance. Bedrock Geosciences Technical Report BG-TR15-01, Bedrock Geosciences, Auenstein, Switzerland.

H.M.Reijonen & W.R.Alexander (2015). Long-term performance of bentonite (LPB): potential load displacement of the bentonite buffer around radioactive waste containers. Phase IIa – preliminary sampling and further site reconnaissance. Bedrock Geosciences Technical Report BG-TR15-02, Bedrock Geosciences, Auenstein, Switzerland.

Thermal alteration of bentonite (TAB): isn't it time we got it right?

W.Russell Alexander[1] & Heini M.Reijonen[2]

1. Bedrock Geosciences, 5105 Auenstein, Switzerland (russell@bedrock-geosciences.com)
2. University of Helsinki/Saanio & Reikkola Oy, Helsinki, Finland

TAB: background

- in the case of thermal alteration of bentonite, cementation, caused by smectite/illite conversion where silica is released at higher temperature (and later reprecipitated) can occur
- currently, in many national programmes, the requirements for maximum bentonite temperatures are set around 100°C why?

TAB: background

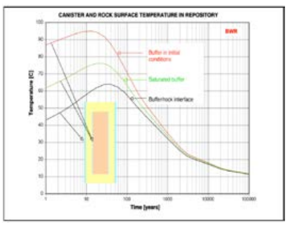
- in many cases, there would be interest in increasing this limit, so allowing, for example, the placement of waste packages more closely than is currently planned
- but this would require more stringent justification than currently exists

TAB: background

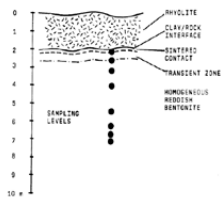
- although a significant number of NA papers on this theme are available, a recent critical re-analysis (CCR) showed that most examples suffer from the same problem: the conditions are not truly representative of a repository
- in most cases of contact metamorphism studied, the temperatures have been much too high (800-900°C)

TAB: background

The degree of saturation of the EBS affects the thermal conductivity of the bentonite, the highest temperatures being met in the "dry" case, i.e. bentonite water contents being as installed (e.g. in Posiva's reference design, the initial water content of the buffer is assumed to be 17%)



TAB: previous work



- numerous studies where smectite-bearing clays which had been penetrated by dykes/sills were examined
- mineralogical changes were associated with subsequent fracturing of the cemented clays as the interstitial waters were driven out.

6. Alexander, Takano, Futakuchi & Reijonen: Long-term performance of bentonite (LPB): potential load displacement of the bentonite buffer around radioactive waste containers.

TAB: previous work

- *unfortunately, these observations cannot be applied directly to the safety case because the full thermal, chemical and pressure histories experienced by these clays were not adequately characterised*
- *no information on the degree of saturation*

TAB: previous work

- the Ishrini bentonite body (Libya) was examined and the impact of a basalt dome and dykes on the bentonite was assessed
- minimum temperature experienced by the bentonite was probably >190°C but bentonite from outwith this zone (i.e. in the lower temperature area) was *not* sampled

TAB: previous work

- *in conclusion, while the probable impact of raised temperatures is minimal (i.e. some local cementation occurs, but the majority of the bentonite remains unaltered), a truly repository-representative analogy has not yet been examined*

TAB: previous conclusion

- NA studies of thermal alteration of bentonite are legion (and only a few are noted above), but a critical review (CCR) of the work carried out to date indicates that they are of little value in supporting the safety case because
  - either the conditions are non-repository relevant
  - the information obtained was too limited

TAB: previous conclusion

- inappropriate temperature range examined (e.g. Ishrini)
- inappropriate timescales for heating of the bentonite, they are usually way too long (e.g. Gulf of Mexico)
- ill-defined (e.g. EU study) or assumed (e.g. Busachi) boundary conditions
- under-characterised physico-chemical parameters of the bentonite
  - *use of modern analytical techniques can better define temperature zonation before assessing changes in the smectite*
  - *define more than just the smectite content (e.g. swelling pressure, saturation state etc)*

TAB: what do we need to do now?

In addition, certain aspects have never been properly addressed, for example:

- as surface/near-surface sites are normally studied, bentonite under a repository-relevant lithostatic load/ hydralulic pressure has not been examined
- again, as surface/near-surface sites are normally studied, fully saturated bentonite has never been examined

Zoopigi

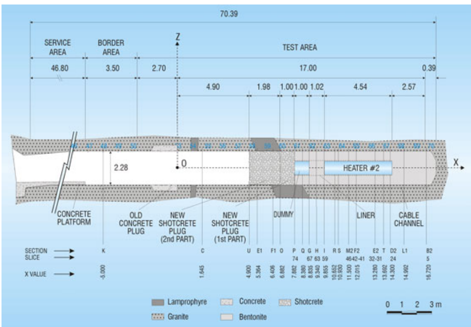
TAB: what next? Coupled projects

We have proposed a two pronged approach:

URL
- to date, bentonite saturation studies have been highly artificial, with induced groundwater flow
- what is required is an approach where resaturation is studied under appropriate conditions, only then can we be sure that the 'dry thermal pulse' is realistic

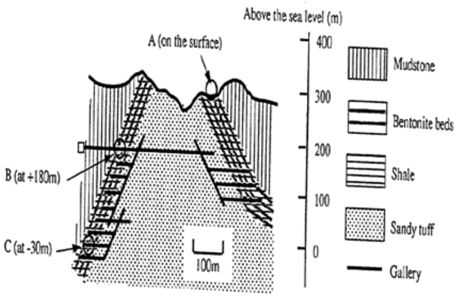
NA
- no more surface sites, time to look at bentonite mines where the bentonite is under appropriate hydrostatic loads
- and is fully saturated
- fully characterise the material using modern methods

TAB: what next? Coupled projects



TAB: what next? Coupled projects

TAB: what next? Coupled projects



TAB: references

Alexander, W.R., Gautschi, A., Zuidema, P. (1998). Thorough testing of performance assessment models: the necessary integration of *in situ* experiments, natural analogues and laboratory work. *Scientific Basis of Nuclear Waste Management XXI,* 1013-1014

Reijonen, H.M., Alexander, W.R., Marcos, N. & Lehtinen, A. (2015). Complementary considerations for safety case – recent developments and future outlook. Swiss Journal of Geosciences,108, 111-120. (DOI 10.1007/ s00015-015-0181-4)

7. Kramer: Long-term durability of shaft sealing materials under highly saline groundwater conditions.

nwmo
NUCLEAR WASTE MANAGEMENT ORGANIZATION SOCIÉTÉ DE GESTION DES DÉCHETS NUCLÉAIRES

Long-Term Durability of Shaft Sealing Materials under Highly Saline Groundwater Conditions

Erik Kremer
Natural Analogue Working Group, Workshop 14
June 2015

Contents

Background
- About the NWMO
- NWMO Deep Geological Repository

Long-Term Durability of Shaft Sealing Materials
- Building Confidence using Natural Analogues

Summary
- Conclusions
- Future work

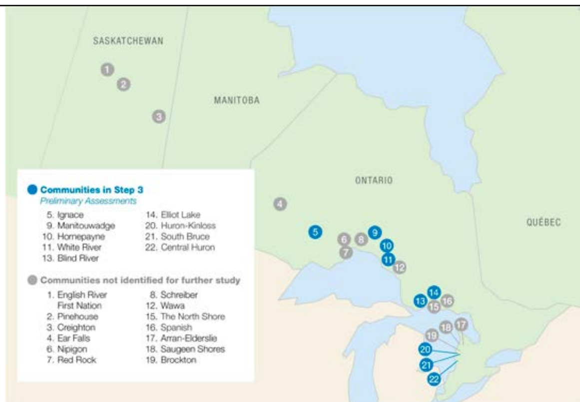
2 nwmo

About the NWMO

- Formed in 2002 as required by *Nuclear Fuel Waste Act*
- Funded by Canada's nuclear waste owners
- Operates on a not-for-profit basis
- Responsible for the safe long term management of Canada's used nuclear fuel
- Repository site selection process launched in 2010, currently in the process of evaluating willing host communities

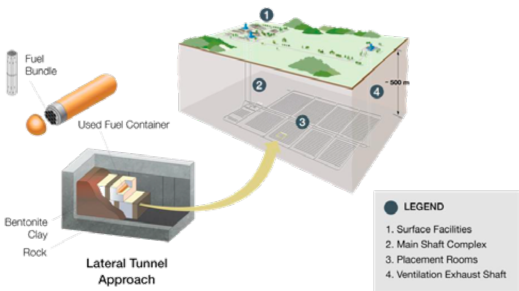
Our mission is to develop and implement collaboratively with Canadians, a management approach for the long-term care of Canada's used nuclear fuel that is socially acceptable, technically sound, environmentally responsible, and economically feasible.

3 nwmo



4 nwmo

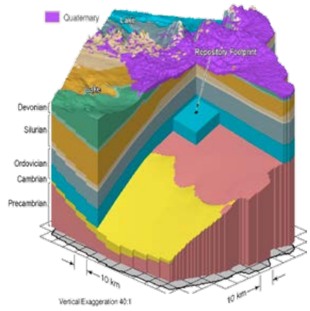
NWMO Deep Geological Repository



5 nwmo

NWMO Deep Geological Repository

- Crystalline and Sedimentary Rock Formations in the Siting Process
- Sedimentary Rock Formations are in the Michigan Basin in Southern Ontario
- Limestone at the Repository Horizon
- Continuous and Relatively Uniform
- Lacking Permeable Fractures or Fault Zones

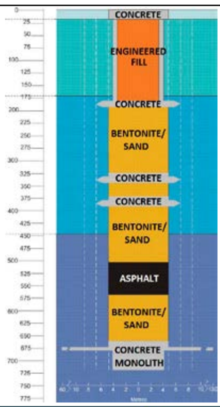


6 nwmo

NWMO DGR

Expected Porewater Chemistry
- 260-370 g/L Na-Ca-Cl Brine
- Near-Neutral pH
- 10-25 °C

Shaft Sealing Materials
- Bentonite-Sand Mix
- Low-Heat Concrete
- Bitumen-Sand-Lime Mix



7 nwmo

Long-Term Durability of Shaft Sealing Materials

Bentonite-Sand Mix
- Sensitivity to Salinity
- Sensitivity to Alkalinity

Low-Heat Concrete
- Sensitivity to Salinity

Bitumen-Sand-Lime Mix
- Sensitivity to Salinity

8 nwmo

Bentonite-Sand Durability

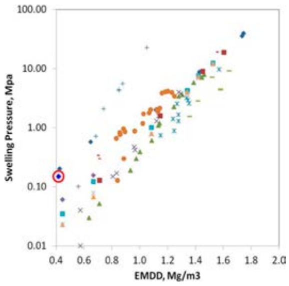
- Primary shaft-sealing material
- 70% MX-80 bentonite and 30% silica sand
- Bentonite provides sealing; sand improves mechanical properties

- High salinity is expected to affect swelling properties
- Mineral stability is expected to remain

Bentonite-Sand Durability

Cyprus Natural Analogue Project

- Perapedhi bentonites exposed to marine salinity ~ 90 Ma
- Comparison with various industrial bentonites indicates no significant impact



Bentonite-Sand Durability

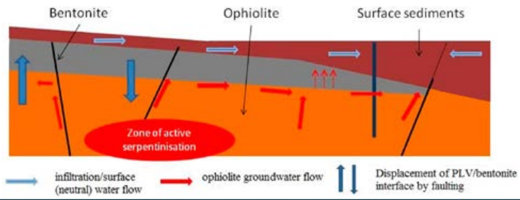
- Bentonite is unstable under high pH
- Concrete can produce alkaline conditions locally
- Concrete bulkheads can affect adjacent bentonite seals

- Low-Heat Concrete leachates are less alkaline than Ordinary Portland Cement (OPC)
- Slow kinetics and persistent metastable phases motivate natural analogue studies

Bentonite-Sand Durability

Cyprus and Philippines Natural Analogue Projects

- Natural alkali groundwaters contact the natural bentonite base
- Analogous to concrete leachates and bentonite-sand mix



Bentonite-Sand Durability

- Reaction is limited where groundwater flow is slow
- Natural analogues show reactions restricted to interface
- Philippines NA (pH~11), maximum 5 cm reaction zone
- Cyprus NA (pH~10-11), less than 1% reaction over 100ka
- Sufficient swelling pressure remains for pore throat reduction to minimise further reaction

Long-Term Durability of Shaft Sealing Materials

Bentonite-Sand Mix

- Sensitivity to Salinity
- Sensitivity to Alkalinity

Low-Heat Concrete

- Sensitivity to Salinity

Bitumen-Sand-Lime Mix

- Sensitivity to Salinity

Low-Heat Concrete Durability

- Concrete provides structural support for the shaft seal column
- Counters swelling of bentonite seals to maintain positions
- Low-Heat Concrete designed to limit effects on adjacent clay

- Natural analogues reported to date are more like OPC
- Very durable: Maqarin ~ 2 Ma; Northern Ireland ~ 58 Ma
- Natural concrete around Puteoli, Italy may include Low-Heat Concrete exposed to marine conditions, possibly brines

Low-Heat Concrete Durability

- To date, only archaeological analogues
- Roman pozzolanic concrete (~200 BCE) is essentially Low-Heat Concrete
- Exposure to marine salinities for ~2,000 years
- High degree of preservation of the bulk concrete

Long-Term Durability of Shaft Sealing Materials

Bentonite-Sand Mix
- Sensitivity to Salinity
- Sensitivity to Alkalinity

Low-Heat Concrete
- Sensitivity to Salinity

Bitumen-Sand-Lime Mix
- Sensitivity to Salinity

17 nwmo

Bitumen-Sand-Lime Durability

- Asphalt provides a low-permeability shaft seal
- Ability to flow and make good contact with host rock
- Effective barrier to water immediately upon placement

- Natural analogues of bitumen-sand-lime have not been reported
- Asphalt-impregnated sandstone and limestone are found in Athabasca (Canada), Utah (USA), Val de Travers (Switzerland), Hannover (Germany)

18 nwmo

Bitumen-Sand-Lime Durability

- Athabasca bitumen has remained stable for over 10 Ma
- Utah sandstone bitumen stable for ~ 30 – 70 Ma, likely formed under marine conditions
- Asphalt floating in Dead Sea could indicate durability under highly saline conditions (Egyptians mummies already a testament)

19 nwmo

Summary

Bentonite-Sand Mix
- Natural bentonite can be a reasonable analogue for Bentonite-Sand
- Cyprus bentonite immersed in marine waters for 90 Ma retains swelling pressure
- Cyrus and Philippine bentonite exposed to alkali groundwaters show reactions limited to interface

20 nwmo

Summary

Low-Heat Concrete
- OPC very durable: Maqarin ~ 2 Ma; Northern Ireland ~ 58 Ma
- Roman pozzolanic concrete analogue: well-preserved after 2,000 years under marine conditions

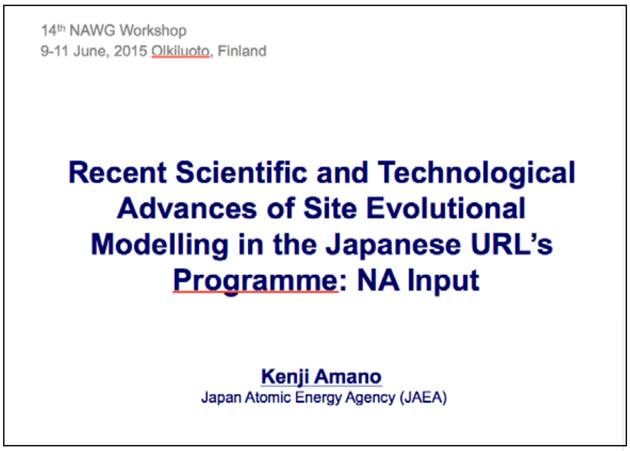
Bitumen-Sand-Lime Mix
- Utah sandstone bitumen likely formed under marine conditions, stable for ~ 30 – 70 Ma

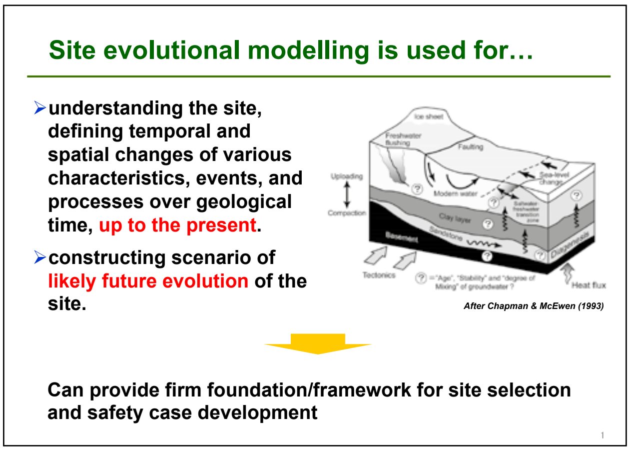
21 nwmo

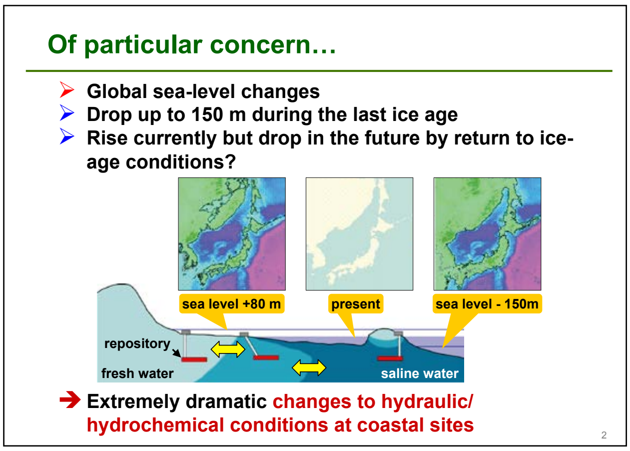
Summary

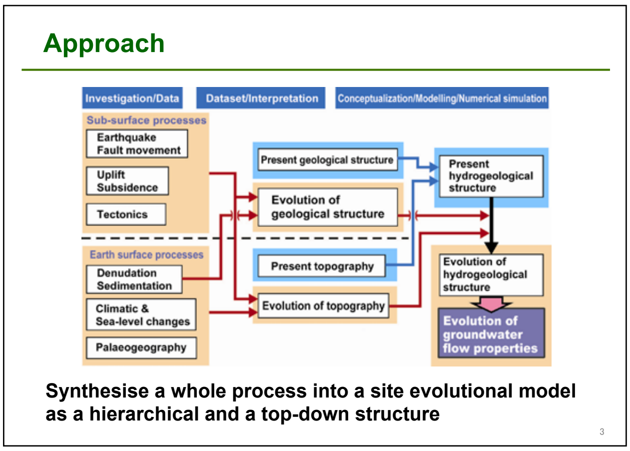
Potential Study – Bentonite Durability under Highly Saline Conditions
- Sample smectite from existing rock cores from the Sedimentary Formations in the Michigan Basin (self-analogue)
- Field sampling of smectite from the Dead Sea coast
- Analyse physico-chemical properties of the smectites to define evolution of properties with palaeohydrological conditions

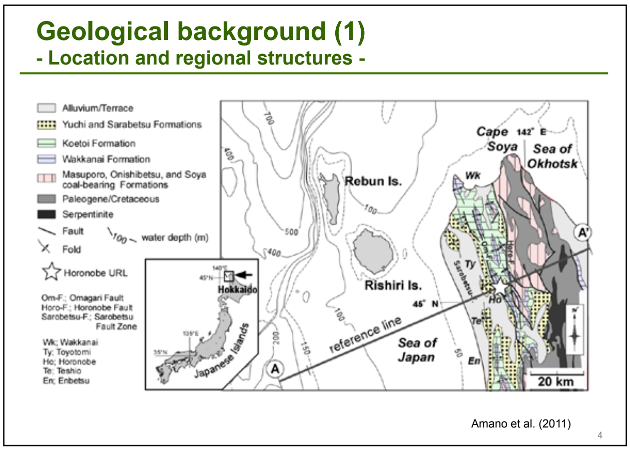
22 nwmo

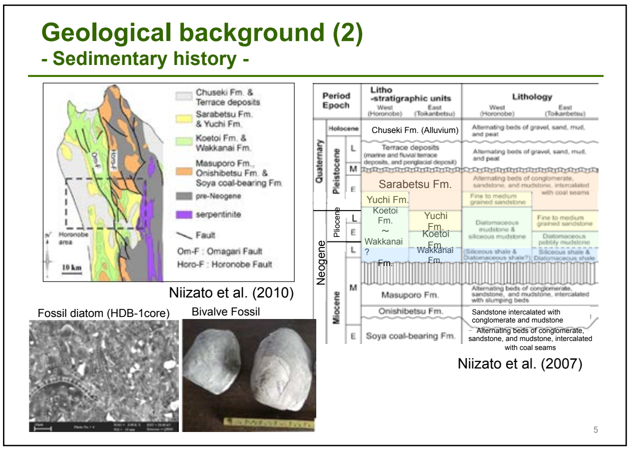

14th NAWG Workshop
9-11 June, 2015 Olkiluoto, Finland
Recent Scientific and Technological Advances of Site Evolutional Modelling in the Japanese URL's Programme: NA Input
Kenji Amano
Japan Atomic Energy Agency (JAEA)

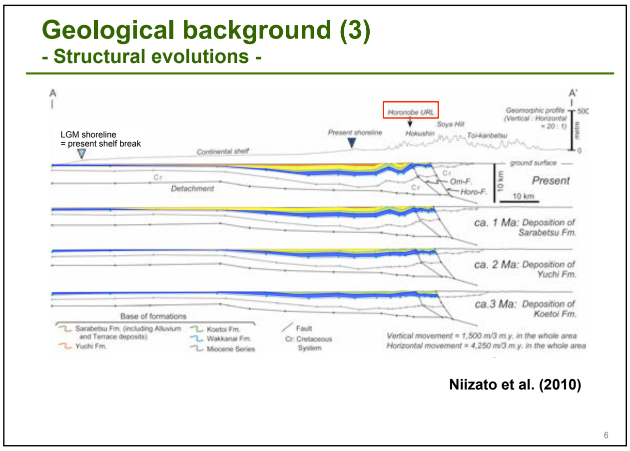

Site evolutional modelling is used for…
understanding the site, defining temporal and spatial changes of various characteristics, events, and processes over geological time, up to the present.
constructing scenario of likely future evolution of the site.
After Chapman & McEwen (1993)
Can provide firm foundation/framework for site selection and safety case development
1

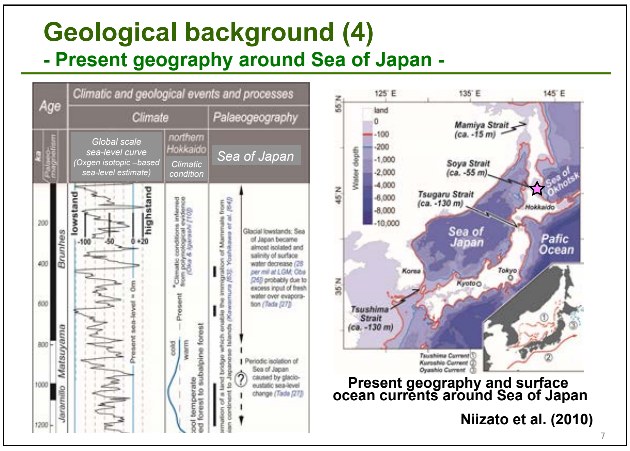

Of particular concern…
Global sea-level changes
Drop up to 150 m during the last ice age
Rise currently but drop in the future by return to ice-age conditions?
sea level +80 m
present
sea level - 150m
repository
fresh water
saline water
Extremely dramatic changes to hydraulic/ hydrochemical conditions at coastal sites
2


Approach
Investigation/Data
Dataset/Interpretation
Conceptualization/Modelling/Numerical simulation
Sub-surface processes
Earthquake Fault movement
Uplift Subsidence
Tectonics
Present geological structure
Evolution of geological structure
Present hydrogeological structure
Earth surface processes
Denudation Sedimentation
Climatic & Sea-level changes
Palaeogeography
Present topography
Evolution of topography
Evolution of hydrogeological structure
Evolution of groundwater flow properties
Synthesise a whole process into a site evolutional model as a hierarchical and a top-down structure
3

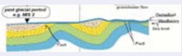
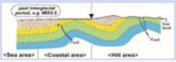

Geological background (1)
- Location and regional structures -
Alluvium/Terrace
Yuchi and Sarabetsu Formations
Koetoi Formation
Wakkanai Formation
Masuporo, Onishibetsu, and Soya coal-bearing Formations
Paleogene/Cretaceous
Serpentinite
Fault
water depth (m)
Fold
Horonobe URL
Cape Soya
Sea of Okhotsk
Rebun Is.
Rishiri Is.
Sea of Japan
reference line
20 km
Amano et al. (2011)
4


Geological background (2)
- Sedimentary history -
Niizato et al. (2010)
Fossil diatom (HDB-1core)
Bivalve Fossil
Niizato et al. (2007)
5


Geological background (3)
- Structural evolutions -
Present
ca. 1 Ma: Deposition of Sarabetsu Fm.
ca. 2 Ma: Deposition of Yuchi Fm.
ca.3 Ma: Deposition of Koetoi Fm.
Niizato et al. (2010)
6


Geological background (4)
- Present geography around Sea of Japan -
Climatic and geological events and processes
Climate
Palaeogeography
Sea of Japan
Mamiya Strait (ca. -15 m)
Soya Strait (ca. -55 m)
Tsugaru Strait (ca. -130 m)
Tsushima Strait (ca. -130 m)
Sea of Japan
Pacific Ocean
Present geography and surface ocean currents around Sea of Japan
Niizato et al. (2010)
7

Geological background (6)
- Conceptualisation-

Age	Geology	Descriptions
Present		- Transgression by global-scale sea-level change - Deposition of alluvium with 40 to 80 m thickness in the Sarobetsu Lowland in the past 14ky (Sakai et al., 2011)
Last Glacial Maximum (1.8 ka)		- Regression caused by global-scale sea-level change (Niizato et al., 2007) - Development of permafrost with 20 m in thickness in land (no data for sub-seabed permafrost)
Last Interglacial (125 ka)		- Transgression caused by global-scale sea-level change (Niizato et al., 2007) - Initiation of Horonobe Hill due to differential erosion - Sea level: +7 m to the present (Oba et al., 2006)
ca. 2000 ka		- Initiation of the fault movement of the Sarobetsu Fault Zone in the present coastal area and of the Omagari Fault in the vicinity of the present URL area (Ito, 1999; Ishii et al., 2008). Such fault movement result in the formation of fold structure in the Horonobe area (Ishii et al., 2008) - Deposition of sand (the Yuchi Formation) and burial of siliceous rocks (the Koetoi and Wakkanai Formations in descending order)

8

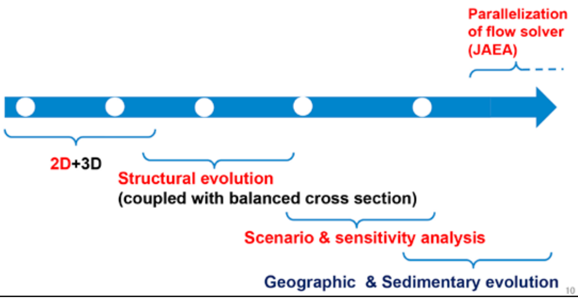
Geological background (6)
- Uncertainty clarification -

Natural events & processes	Descriptions of uncertainty
Tectonics	Inhomogeneous nature of spatial distribution of uplift and subsidence
	Variations in the rate of uplift and subsidence
Topography	Height of mountain range in the past period
Hydrogeology	Variations of recharge rate through the Glacial-Interglacial cycles
	Changes in porosity and hydraulic conductivity over geological time
	Uncertainties about sea-levels in the Sea of Japan
	Uncertainties about location of former shoreline through the past glacial-interglacial cycles
Hydrogeochemistry	Variations in seawater chemistry (especially salinity and stable isotopes)
Hydrogeology & Hydrogeochemistry	Degree of coverage of the permafrost (i.e. just how discontinuous is discontinuous?) around the northern end of the Sea of Japan

9

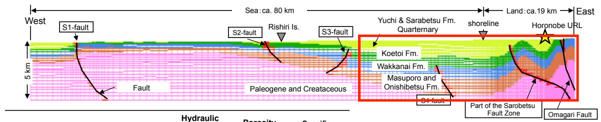
Site evolution modelling
- Tools development -

SMS (Sequential Modeling System of geo-environmental evolution impact on groundwater flow) by Hazama Co.

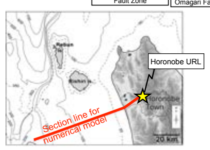


10

2D Modelling
- Modell setting (1)-



Units	Hydraulic conductivity (m/s)	Porosity (%)	Specific storage (1/m)
Surface deposits (ca. 10 m depth from surface)	1.0E-06	60	1.0E-05
Yuchi & Sarabetsu Fm. and Quaternary	*Depth_Yt* *	60	1.0E-05
Koetoi Formation	*Depth_Kt* *	60	1.0E-05
Wakkanai Formation	*Depth_Wk* *	40	1.0E-05
Masuporo & Onishibetsu Fm.	5.0E-10	30	1.0E-05
Paleogene & Cretaceous	1.0E-11	20	1.0E-05
Faults (Omagari Fault, part of the Sarobetsu Fault Zone, and S1- to S4-faults)	1.0E-07	50	1.0E-05



* *Depth_Yt*, *_Kt*, and *_Wk* are hydraulic conductivities of each formation as a function of depth. K: hydraulic conductivity (m/s), Z: depth (m)

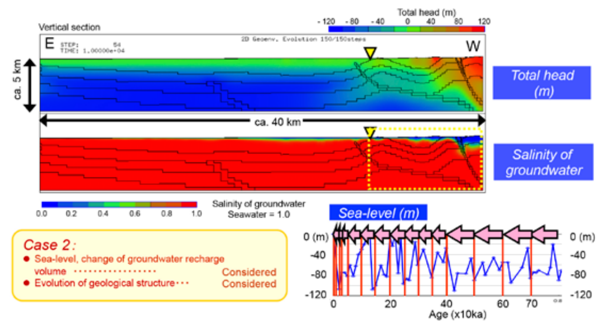
Depth_Yt : $\log_{10}(k) = -0.0034Z - 8.3665$ [upper limit: 1×10^{-8} m/s, lower limit: 1×10^{-11} m/s]

Depth_Kt : $\log_{10}(k) = -0.0039Z - 7.5935$ [upper limit: 1×10^{-7} m/s, lower limit: 1×10^{-11} m/s]

Depth_Wk : $\log_{10}(k) = -0.0061Z - 5.5626$ [upper limit: 1×10^{-6} m/s, lower limit: 1×10^{-11} m/s]

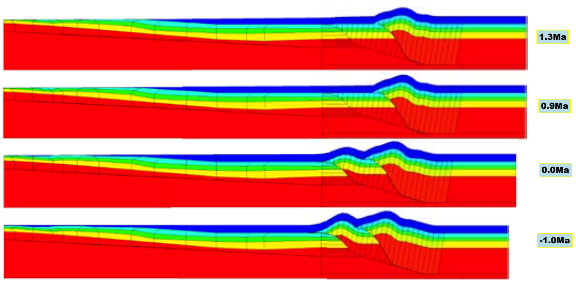
11

2D Modelling
- Modelling results -



13

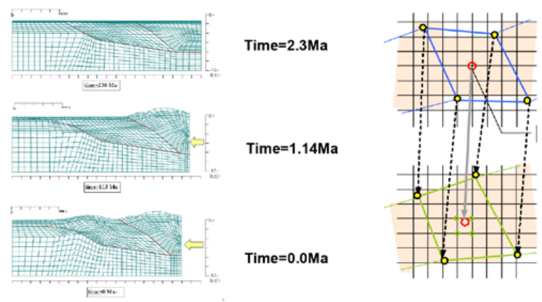
2D Modelling
- Structural evolution (balanced cross section method) -



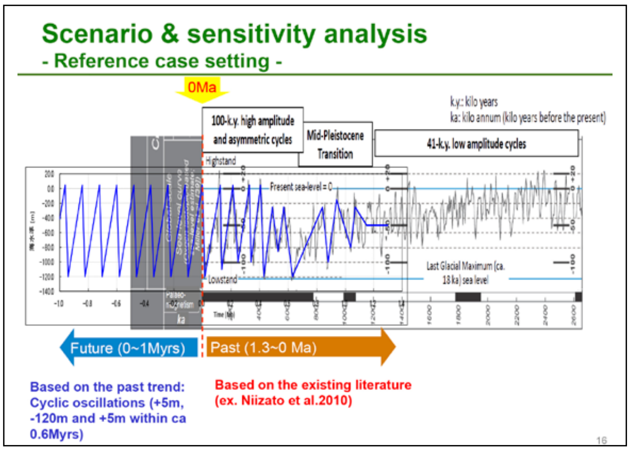
14

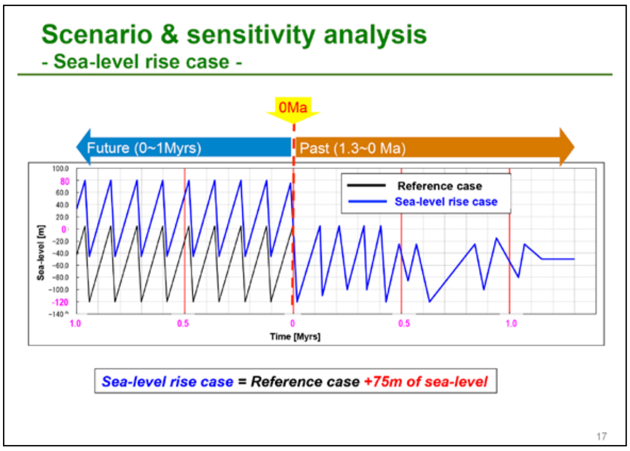
2D Modelling
- Structural evolution (Balance cross section method) -

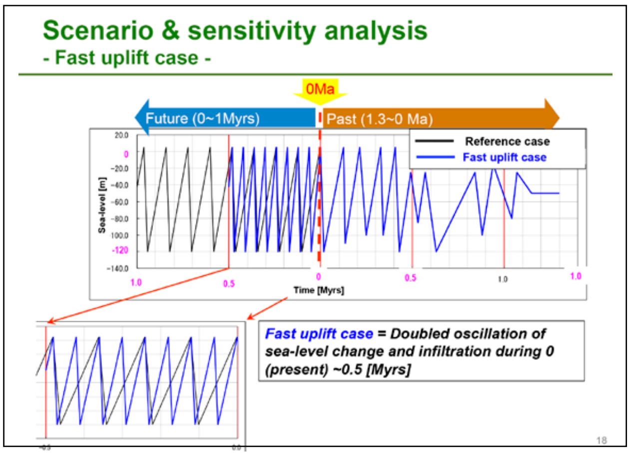
Deformed mesh systems with balanced cross sections

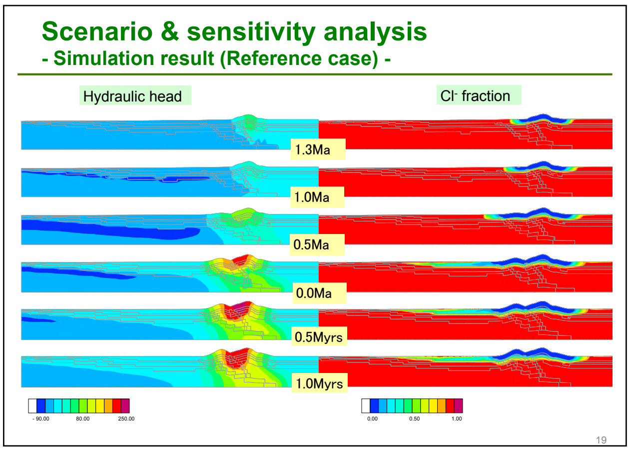


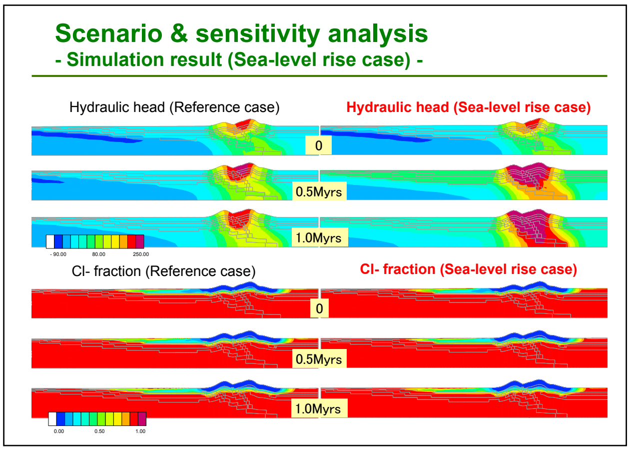
8. Amano: Recent scientific and technological advances of site evolutional modelling in the Japanese URL's programme: NA input.

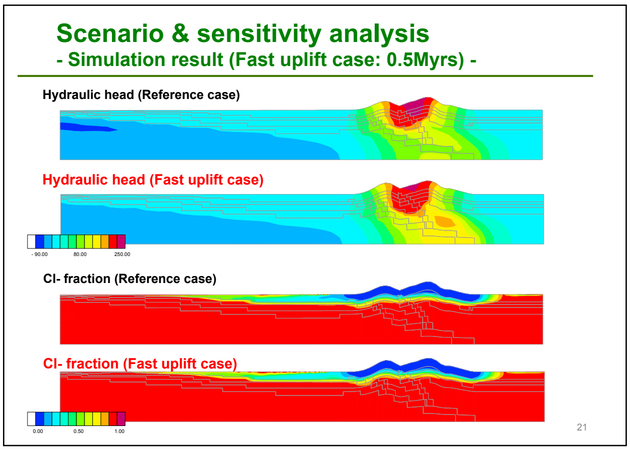


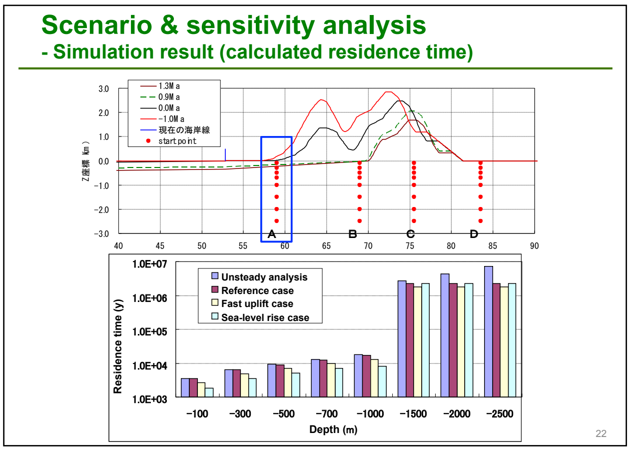


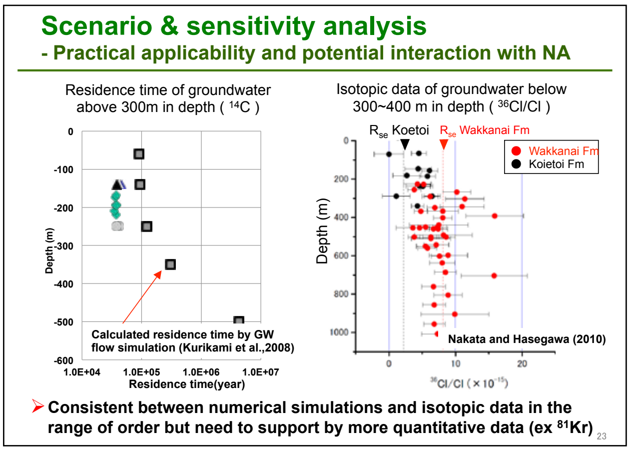










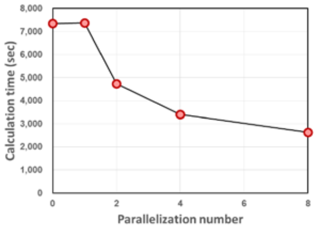


8. Amano: Recent scientific and technological advances of site evolutional modelling in the Japanese URL's programme: NA input.

Parallelization of flow solver

- For further improvement of SEM

Parallelization number	Calculation time (sec)
0 (Original code)	7344.7
1	7369.9
2	4731.0
4	3398.7
8	2625.1



Super Computing system in JAEA (200TFlops)

- Can accelerate time-consuming simulations with large spatial-temporal scales
- Difficulties while area partitioning in larger parallelization number due to causing overlapped sections

24

Summary

- SEM must be an fundamental basis for safety case development during whole phases of geological disposal processes, from site selection to final closure.
- Current SEM methodology can handle major geological, hydrogeological and hydrochemical processes conjointly in the geologic time scale.
- Some technical issues (uncertainty management, verification by quantitative field data, acceleration of the code etc.) are left to be improved.

25

Uranium occurrences in Finland as indicators of long-term geochemical stability

Maikki Siitari-Kauppi
Laboratory of Radiochemistry, University of Helsinki
Karl-Heinz Hellmuth, David Read, Nuria Marcos

NAWG 9.6.2015

OUTLINE

- Background
- Rock matrix structure
- Revisiting Palmottu / Uranium phases on fracture surfaces and in matrix
- Dating on pure mineral phases
- Conclusions

BACKGROUND

- Palmottu EU-project from 1996 until 2000
- Palmottu work continued as IAEA's CRP project by Karl-Heinz Hellmuth from STUK 2000-2002
- Revisiting Palmottu by a group of obstinate researchers 2003-2005
- Publicly funded project coordinated by Technical University of Finland Prof Kirsti Loukola-Ruskeeniemi 2006-2008

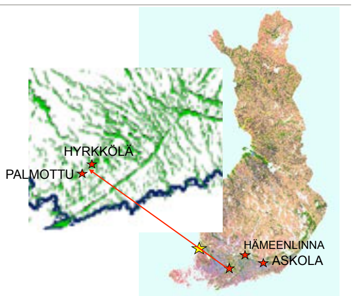
WHY IS IT IMPORTANT?

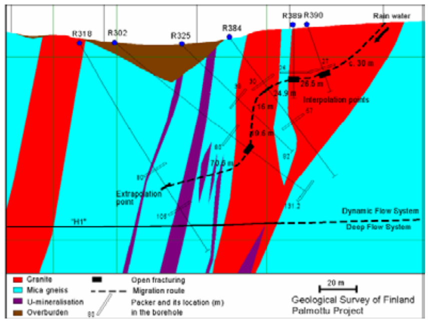
- Long-term safety linked to comparison with natural fluxes (Finnish legistlation).
- Glaciation regarded as the greatest natural threat to repository integrity (Posiva).
- Verifies UO_2 alteration and uranium migration processes in geochemically relevant settings.
- U series provides direct means of estimating timescales of past events.

URANIUM STUDIES IN FINLAND

- Salpausselät I-III
- Sampling places

Palmottu
Hyrkkölä
Askola
Hämeenlinna

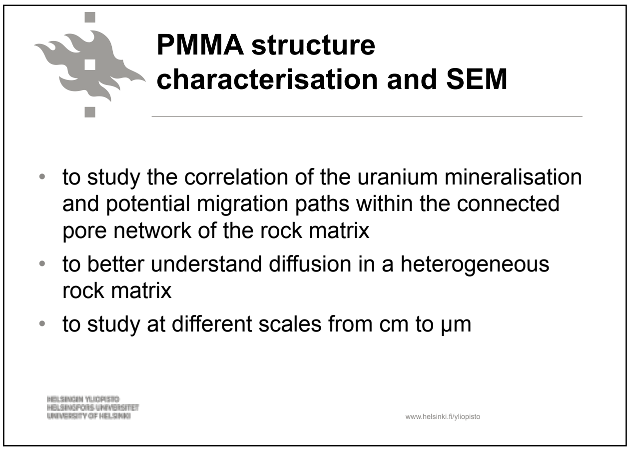


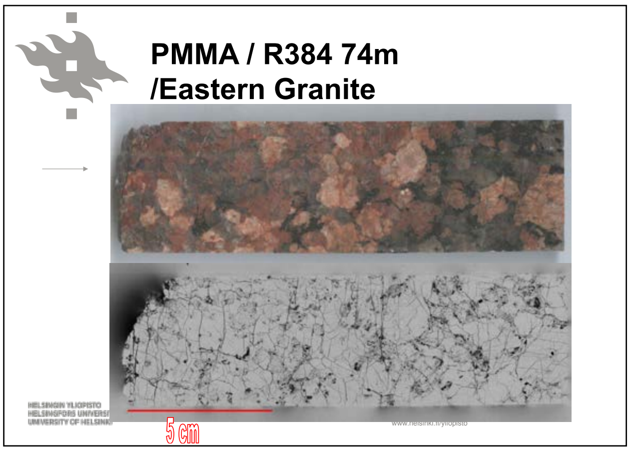


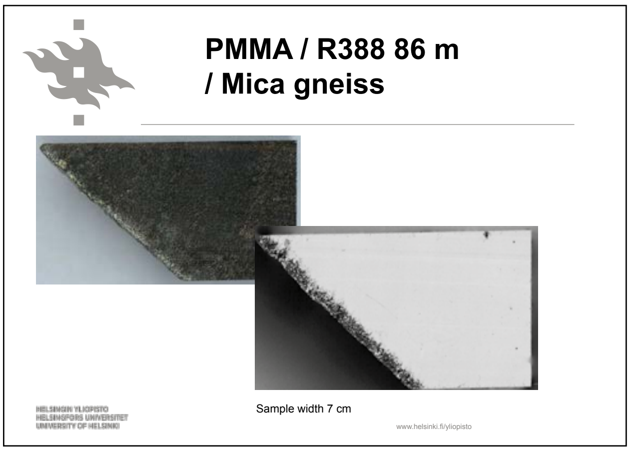
U mineralization at Palmottu (study area) indicating groundwater flow path)

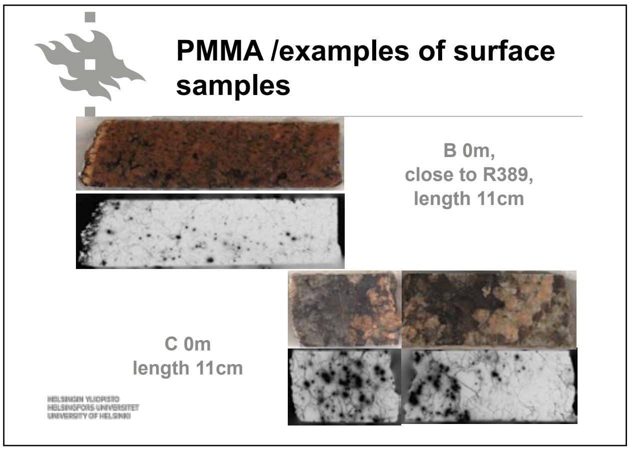
Surface samples, October 2003: drillholes B and C and a boulder D

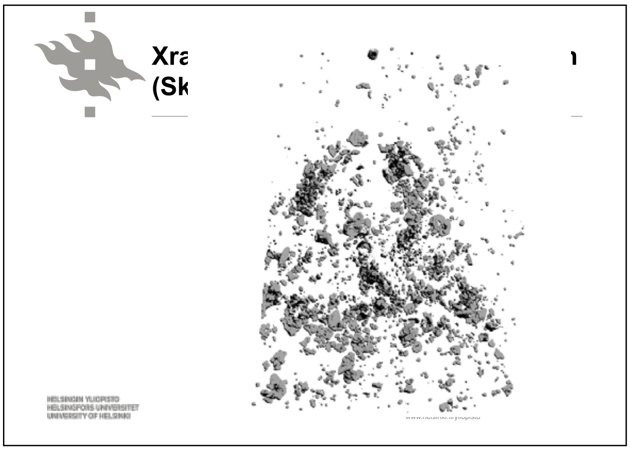

Drillcore samples
R302,R373,R384,R385,R388,
R389,R390
HELSINGIN YLIOPISTO
HELSINGFORS UNIVERSITET
UNIVERSITY OF HELSINKI
www.helsinki.fi/yliopisto


PMMA structure characterisation and SEM
• to study the correlation of the uranium mineralisation and potential migration paths within the connected pore network of the rock matrix
• to better understand diffusion in a heterogeneous rock matrix
• to study at different scales from cm to µm
HELSINGIN YLIOPISTO
HELSINGFORS UNIVERSITET
UNIVERSITY OF HELSINKI
www.helsinki.fi/yliopisto


PMMA / R384 74m
/Eastern Granite
5 cm


PMMA / R388 86 m
/ Mica gneiss
Sample width 7 cm
HELSINGIN YLIOPISTO
HELSINGFORS UNIVERSITET
UNIVERSITY OF HELSINKI
www.helsinki.fi/yliopisto


PMMA /examples of surface samples
B 0m,
close to R389,
length 11cm
C 0m
length 11cm
HELSINGIN YLIOPISTO
HELSINGFORS UNIVERSITET
UNIVERSITY OF HELSINKI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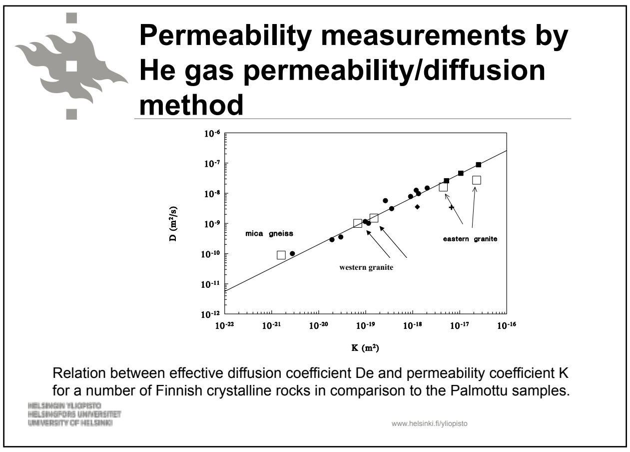
HELSINGIN YLIOPISTO
HELSINGFORS UNIVERSITET
UNIVERSITY OF HELSINKI


Permeability measurements by He gas permeability/diffusion method
D (m²/s)
K (m²)
mica gneiss
western granite
eastern granite
Relation between effective diffusion coefficient De and permeability coefficient K for a number of Finnish crystalline rocks in comparison to the Palmottu samples.
HELSINGIN YLIOPISTO
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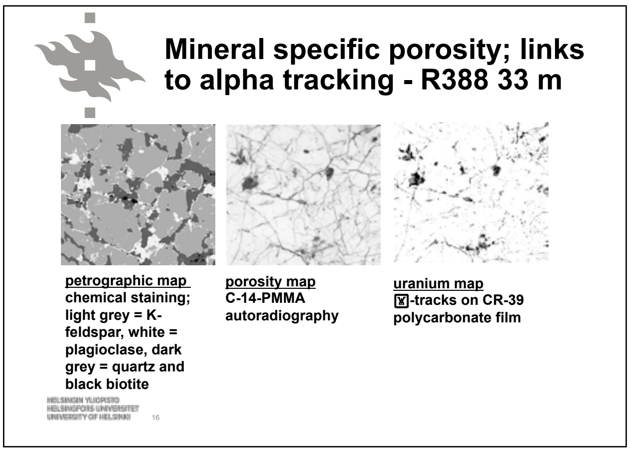
Mineral specific porosity; links to alpha tracking - R388 33 m
petrographic map
chemical staining; light grey = K-feldspar, white = plagioclase, dark grey = quartz and black biotite
porosity map
C-14-PMMA autoradiography
uranium map
-tracks on CR-39 polycarbonate film
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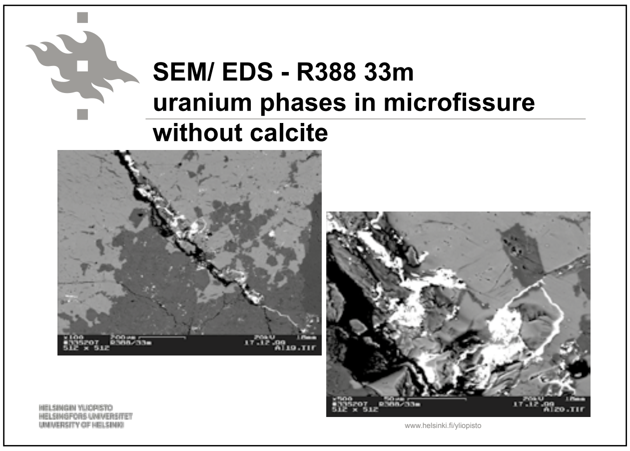
16


SEM/ EDS - R388 33m
uranium phases in microfissure
without calcite
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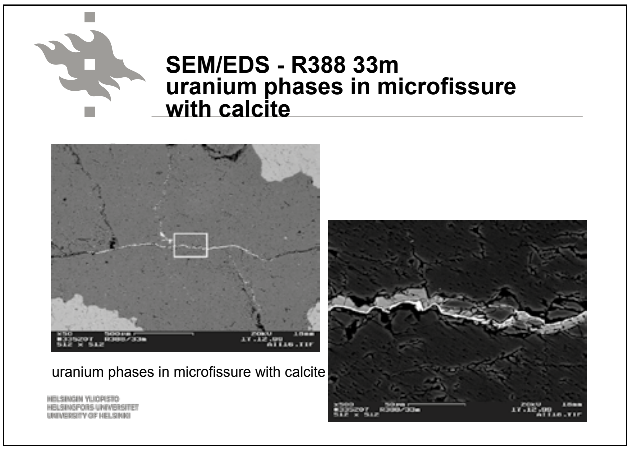
SEM/EDS - R388 33m
uranium phases in microfissure
with calcite
uranium phases in microfissure with calcite
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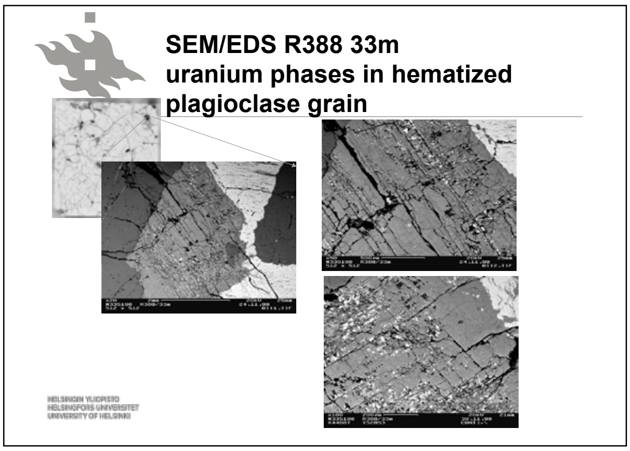
SEM/EDS R388 33m
uranium phases in hematized
plagioclase grain
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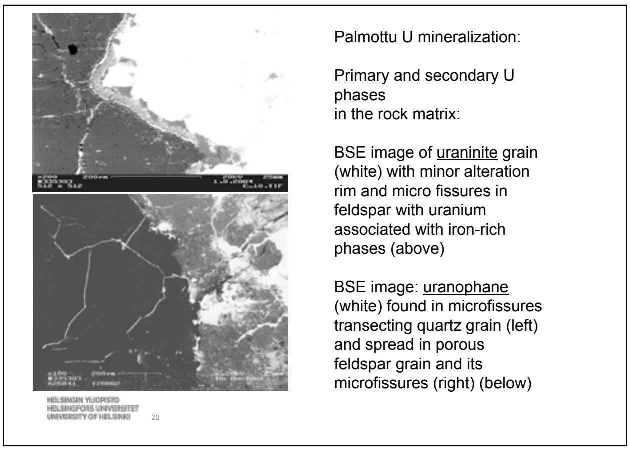
Palmottu U mineralization:
Primary and secondary U phases in the rock matrix:
BSE image of uraninite grain (white) with minor alteration rim and micro fissures in feldspar with uranium associated with iron-rich phases (above)
BSE image: uranophane (white) found in microfissures transecting quartz grain (left) and spread in porous feldspar grain and its microfissures (right) (below)
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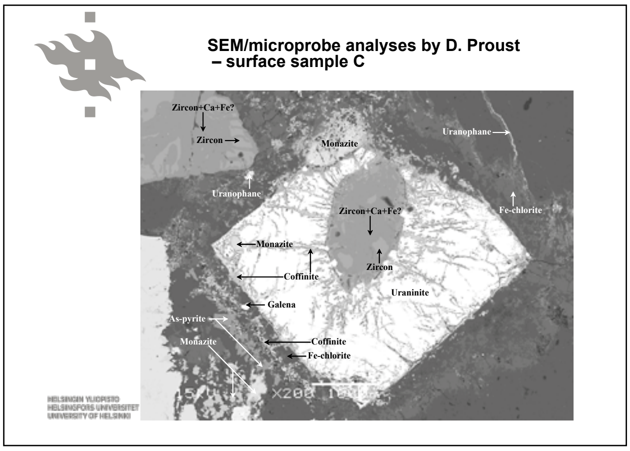
20


SEM/microprobe analyses by D. Proust
– surface sample C
Zircon+Ca+Fe?
Zircon
Monazite
Uranophane
Uranophane
Fe-chlorite
Zircon+Ca+Fe?
Monazite
Coffinite
Zircon
Uraninite
Galena
As-pyrite
Monazite
Coffinite
Fe-chlorite
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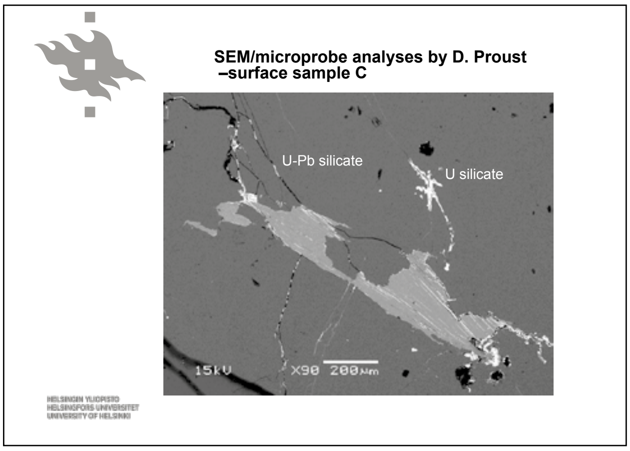
SEM/microprobe analyses by D. Proust
–surface sample C
U-Pb silicate
U silicate
15kV
X90 200µm
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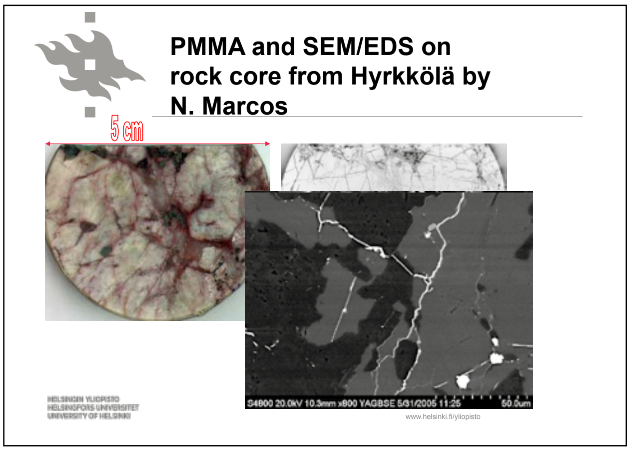
PMMA and SEM/EDS on
rock core from Hyrkkölä by
N. Marcos
5 cm
S4800 20.0kV 10.3mm x800 YAGBSE 5/31/2005 11:25
50.0um
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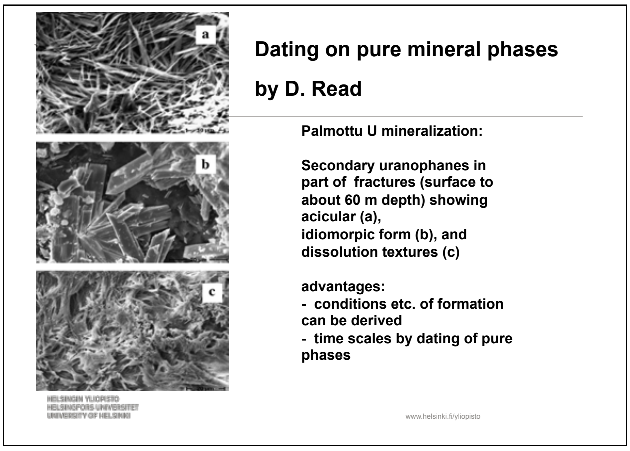
a
b
c
Dating on pure mineral phases
by D. Read
Palmottu U mineralization:
Secondary uranophanes in part of fractures (surface to about 60 m depth) showing acicular (a), idiomorpic form (b), and dissolution textures (c)
advantages:
- conditions etc. of formation can be derived
- time scales by dating of pure phases
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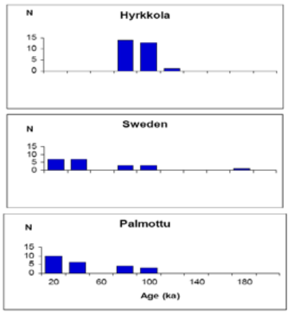
Mineral Ages in Fennoscandian shield by D. Read

U-Th ages for uranium silicates from Hyrkkölä, Palmottu and Sweden indicating consistently distinct glacial periods

(no samples from greater depth and near surface available from Hyrkkölä)



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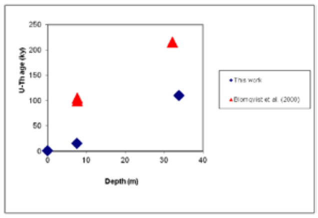
D. Read et al. (2008) Secondary uranium mineralization in southern Finland and its relationship to recent glacial events. Global&Planetary Change 60 235-249

25

Dating on pure mineral phases versus on bulk rock



Comparison of uranophane U series ages with earlier work:
- pure phase analysis by mechanical micro-separation
- bulk samples or selective leaching leading to artefacts

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Conclusions (I)

- Well developed network of migration pathways found througout the rock cores in Eastern granite and Western granite
- Permeabilities of rock cores varied from 10^{-17} m^2 - 10^{-21} m^2
- Microfissures apertures from 1µm to a few tens of microns; filled partly with calcites
- Secondary uranium phases were found on fracture surfaces in microfissures + within altered plagioclase grains in Eastern granites and Hyrkkölä samples
- Deepest uranophane was found from Hyrkkölä at a depth of about 60 m 🗵 depth of U oxidation

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Conclusions (II)

- Mobilisation and immobilisation processes of uranium are active under present climatic conditions
- Buffer capacity of the rock matrix - yes
- Long term stability of geochemical conditions – yes
- Indication of matrix diffusion of RN from flow ?

- A unique potential to describe the glacial water intrusion processes and the relevant chemical buffering reactions by the adjacent bedrock minerals, as well as the associated uranium mobilization and immobilization processes in a quantitative fashion.

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K-H. Hellmuth, D. Read, S. Black, T. Buckby, S.+V. Oziabkin, N. Marcos, M. Markovaara-Koivisto, M. Siitari-Kauppi, M. Kelokaski, T. Proust, P. Vaganov, D. Traber, T. Tarvainen, B. Backman, T. Hatakka, H. Savolainen, J. Kaija, R. Blomqvist, K. Rasilainen, P. Pitkänen, J. Löfman, A. Luukkunen, S. Partamies, Geochemical fluxes in the geosphere: quantitative understanding by identification and verification of processes. in: 2nd Annual Workshop Proceedings of the Integrated Project "Fundamental Processes of Radionuclide Migration" – 6th EC FP IP FUNMIG Edited by G. Buckau, B. Kienzler, L. Duro, V. Montoya SKB Technical report TR-07-05 (2007) p. 140-147

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Thanks to all co-authors

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N. Marcos, M. Markovaara-Koivisto (Technical University Helsinki),
M. Siitari-Kauppi, M. Kelokaski, L. Jokelainen, (University Helsinki),
T. Proust (University Poitiers)
T. Tarvainen, B. Backman J. Kaija, R. Blomqvist, A. Lindberg (Geological Survey of Finland, Espoo)

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Thank you for your attention!

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NUMO

Natural system evidence underpinning the NUMO 2015 Safety Case

Kunio Ota

Nuclear Waste Management Organization of Japan (NUMO)

NAWG 14th Workshop, Olkiluoto, Finland, 9-10 June 2015

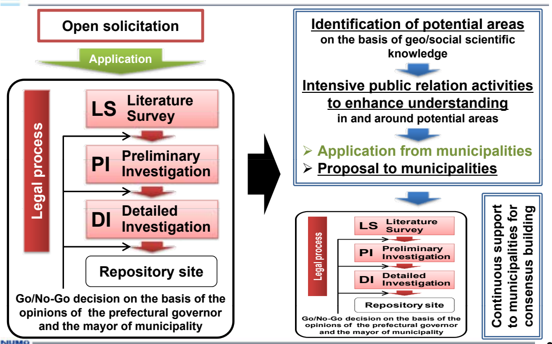
Recent News

NEW: Amendment to "Basic Policy" on 22 May 2015

- Government to play a leading role for final disposal
- Government to select potential areas where geo/social scientific suitability is ensured and to propose acceptance of site investigations to municipalities
- Government and NUMO to develop alternative options that ensure reversibility and retrievability in final disposal
- Government, NUMO and Utilities to implement final disposal as a responsibility of the current generation so as not to put this on the future generation
- Government, NUMO and Utilities to give support to interested municipalities for consensus building and sustainable growth
- Government, NUMO and Utilities to encourage the feeling of respect and gratitude among Japanese people to volunteer municipalities
- Japan Atomic Energy Commission to evaluate regularly the progress of technical development and site selection

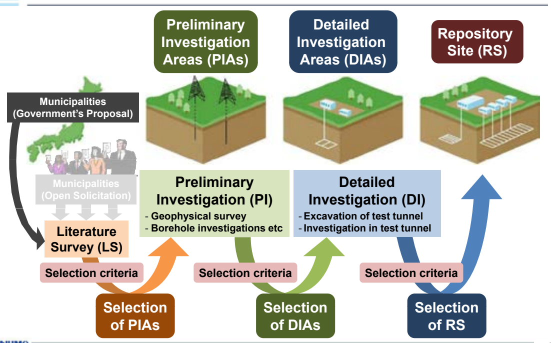
NUMO 2

NEW: Government leading siting process



NUMO 3

NEW: Staged identification of suitable repository site



NUMO 4

NUMO 2015 Safety Case

Quick look at "NUMO 2015 Safety Case"

- Whilst at the present generic stage there has been to date no choice of host rock or site, NUMO has been developing a generic safety case with the main aims of…
  - ◆ demonstrating the technical feasibility and long-term safety of geological disposal in Japan on the basis of state-of-the-art scientific and technological knowledge
  - ◆ providing the basic safety case structure applicable to any selected site, as the siting process is at a new stage
- Although the NUMO 2015 SC extends the H12 Report with reference to the international safety case concept, major progress is highlighted, for example…
  - ◆ potential host rock models, feasibility of retrievability, risk informed approach, pre-closure safety assessment

(Fujihara et al., 2015)

NUMO 6

Contents of "NUMO 2015 Safety Case" and NA input

1. Background and purpose
2. Safety strategy
3. Geological characterisation and synthesis ← NA input
4. Repository design and engineering ← NA input
5. Repository construction, operation and closure
6. Assessment of pre-closure safety
7. Assessment of post-closure long-term safety ← NA input
8. Confidence in the technical feasibility of geological disposal in Japan ← NA input
9. Conclusions

NUMO 7

Top-level statement of "geo" chapter

- Selecting a suitable repository site in Japan where the key safety functions of the host geological environment will persist for a long period of time would be feasible
  - Prevalence of the favourable THMC conditions in deep geological environments can be demonstrated in Japan
  - Potentially significant impacts of natural disruptive events/processes on the geological environment can be precluded
  - Stable geological environments can be identified through characterisation of 4D site evolution

→ Providing multiple lines of arguments and geoscientific evidence for the reliance that can be placed on the long-term geosphere stability in Japan

(Ota et al., 2015)

NUMO 8

Prevalence of favourable THMC conditions (1)

	Requirements redefined by *METI (2014)*	Prevailing conditions
T	Low geothermal gradients for prolonged periods	➢ Geothermal gradients 3~5°C/100 m in non-volcanic regions
H	Slow groundwater movements resulted from low hydraulic gradients and/or low rock hydraulic conductivities	➢ Hydraulic gradients 0.001~0.1 ➢ Hydraulic conductivities 10^{-12}~10^{-6} m/s ➢ High hydraulic gradient associated with low hydraulic conductivities and vice versa ➢ Very long groundwater residence time 2~10 Myr in low permeable sedimentary formations

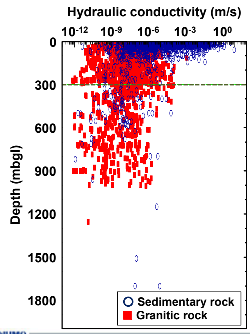
NUMO 9

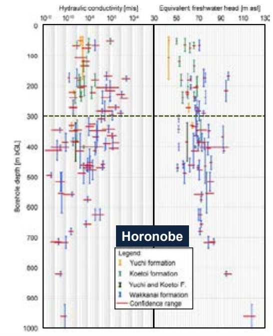
Prevalence of favourable THMC conditions (2)

	Requirements redefined by *METI (2014)*	Prevailing conditions
M	Small rock deformation Including rock creep	➢ In situ stress anisotropy <1:2 ➢ Sufficient mechanical strength against the magnitude of initial stress
C	Groundwater with near neutral pH, reducing conditions and DIC concentrations <0.5 mol/L	➢ Groundwater pH generally 6~9 ➢ Groundwater Eh more negative with depth ➢ Groundwater DIC concentrations <0.1 mol/L ➢ Long-term 1~10 Myr maintenance of pH-Eh conditions by a water-mineral-microbe system

NUMO 10

H: Low hydraulic conductivities and overpressure





NUMO 11

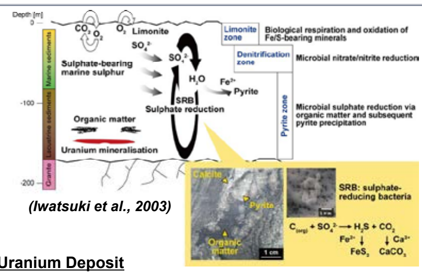
H: Groundwater ages

Location	Host rock	Depth (mbgl)	Age (ky)	Methods	Reference
Horonobe, Hokkaido	Miocene sedimentary rock	700	10,000~	^{4}He, ^{36}Cl	*Nakata & Hasegawa, 2010*
Yokosuka, Kanagawa	Miocene sedimentary rock	210~500	7,000~	^{4}He, ^{14}C, ^{36}Cl	*Hasegawa et al., 2013*
Kushiro, Hokkaido	Cretaceous sedimentary rock	750	2,000~	^{36}Cl	*Mahara et al., 2006*
Kobe, Hyogo	Pliocene sedimentary rock	1,500	230~25	^{4}He	*Morikawa et al., 2005*
Rokkasho, Aomori	Miocene sedimentary rock	180	80~50	^{4}He	*Nakata & Hasegawa, 2011*
Mizunami, Gifu	Cretaceous granite	1,000	20~10	^{4}He, ^{14}C	*Hasegawa et al., 2010*

→ A convincing indication for slow groundwater movements

NUMO 12

C: Long-term maintenance of pH-Eh conditions



Tono Uranium Deposit

Favourable pH-Eh conditions maintained by carbonate equilibria, microbial sulphate reduction with oxidation of organic matter and precipitation of pyrite, subsequently U immobilised

NUMO 13

Safety-affecting factors and basic solutions

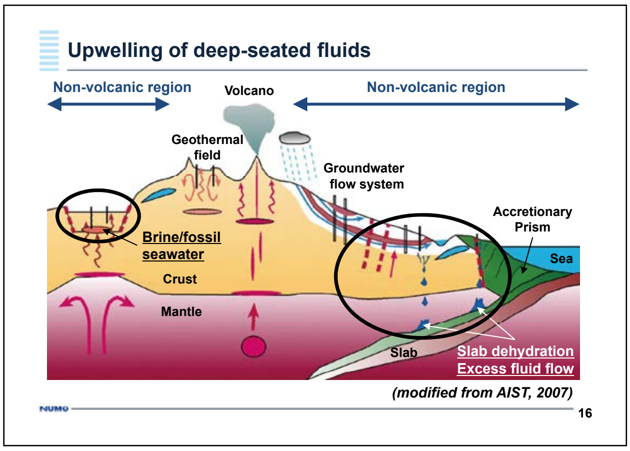
Phase	Safety functions & conditions	Safety-affecting factors	Basic solution strategy	Time [y]	Scale (Regional / Repository / Block)
Pre closure	Ease of construction	Thick Quaternary un-consolidated deposits	Exclude areas	10^{0} ~ 10^{2}	LS, PI
Pre closure	Low likelihood of natural disasters	Earthquake, tsunami, pyroclastic flow etc	Take engineering measures		LS, PI, DI
Post closure	Isolation	Exploration or mining of mineral resources	Exclude areas		LS, PI
Post closure	Isolation / Containment	Disruptive igneous activity, fault movement, uplift/erosion etc	Preclude significant impacts	10^{2} ~ 10^{6}	LS, LS, PI
Post closure	Containment	Gradual uplift/erosion, sea-level changes etc	Evaluate long-term evolution and take engineering measures		LS, PI, DI

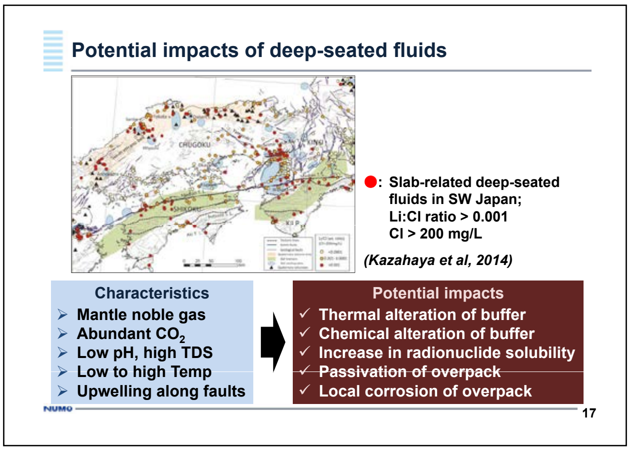
NUMO 14

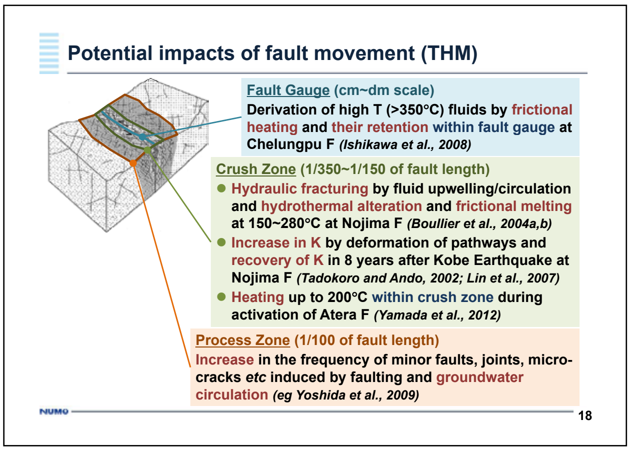
Disruptive events/processes and potential impacts

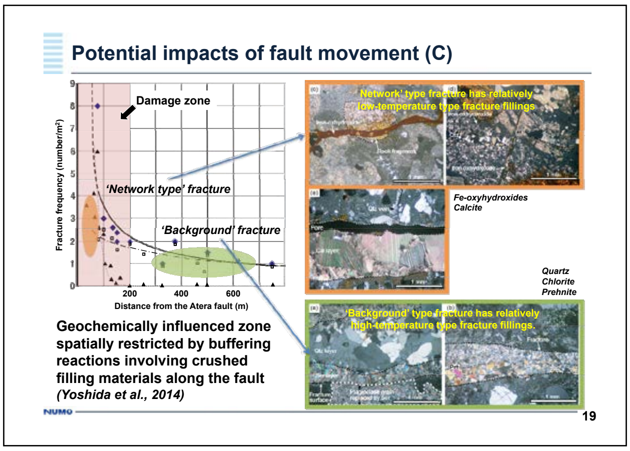
Major disruptive events/processes		Impacts		Areas to be excluded
Igneous activity	Magma intrusion into and eruption through repository	Loss of isolation		Within 15 km radius circle around a Quaternary volcanic centre
	Elevated heat flow and generation of heat sources	Impairment of containment	T	
	Penetration of volcanic thermal fluids		C	Where low pH, high DIC or high T fluids occurs
Non-volcanic or deep-seated fluids	Elevated heat flow and generation of heat sources		T	
	Penetration of non-volcanic thermal or deep-seated fluids		C	
Fault movement	High crustal displacement at repository		M	Transected by active faults and of a fault process zone having width of 1/100 of fault length
	Increase in hydraulic conductivity in/around fault		H	
	Penetration of oxidising surface water		C	
Uplift/ erosion	Significant reduction in thickness of geological barrier	Loss of isolation		Where uplift/erosion amounting to >300 m during the last 10^{5} yr

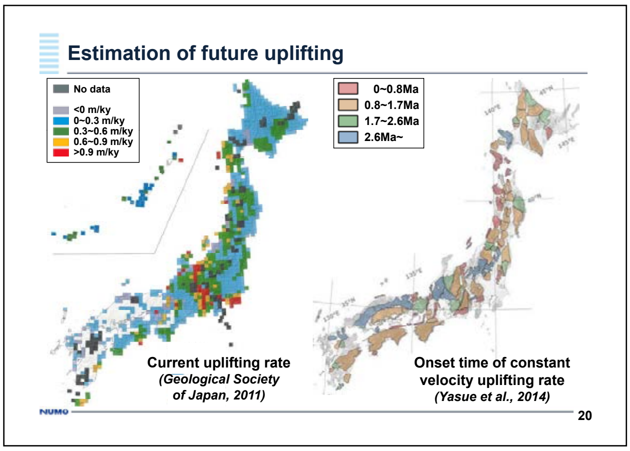
NUMO 15

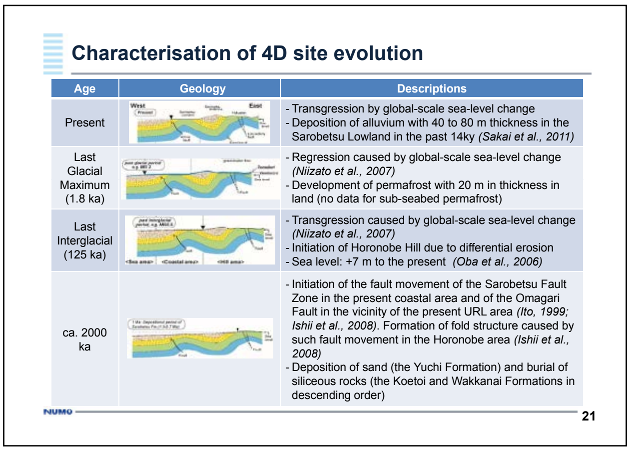




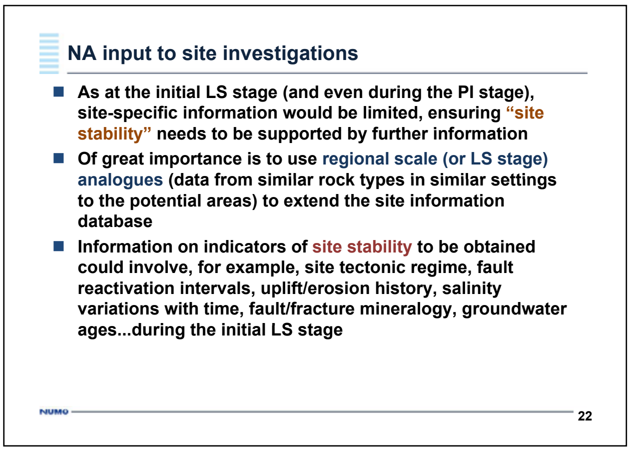


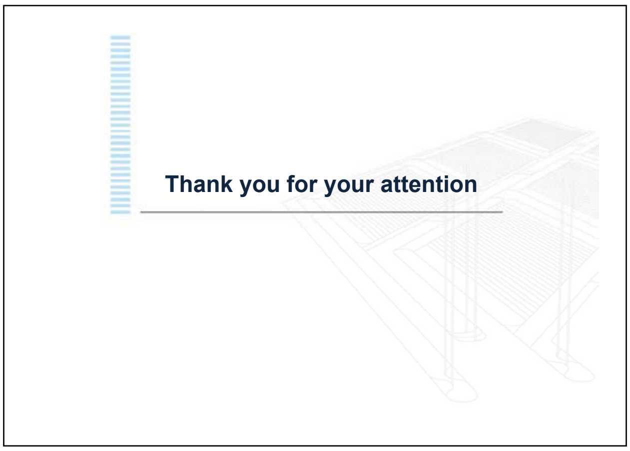






Age	Geology	Descriptions
Present		- Transgression by global-scale sea-level change - Deposition of alluvium with 40 to 80 m thickness in the Sarobetsu Lowland in the past 14ky (Sakai et al., 2011)
Last Glacial Maximum (1.8 ka)		- Regression caused by global-scale sea-level change (Niizato et al., 2007) - Development of permafrost with 20 m in thickness in land (no data for sub-seabed permafrost)
Last Interglacial (125 ka)		- Transgression caused by global-scale sea-level change (Niizato et al., 2007) - Initiation of Horonobe Hill due to differential erosion - Sea level: +7 m to the present (Oba et al., 2006)
ca. 2000 ka		- Initiation of the fault movement of the Sarobetsu Fault Zone in the present coastal area and of the Omagari Fault in the vicinity of the present URL area (Ito, 1999; Ishii et al., 2008). Formation of fold structure caused by such fault movement in the Horonobe area (Ishii et al., 2008) - Deposition of sand (the Yuchi Formation) and burial of siliceous rocks (the Koetoi and Wakkanai Formations in descending order)





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NUMO 24

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NUMO 28

11. Klein, McKinley, Hardie, Tachi, Shibata, Amano & Takase: Use of natural analogues to develop and test models of radionuclide release and transport in uplift/erosion scenarios.

Use of natural analogues to develop and test models of radionuclide release and transport in uplift / erosion scenarios

Liza Klein, Ian McKinley, Susie Hardie, Yukio Tachi, Masahiro Shibata, Yuki Amano, Hiroyasu Takase

mcm Quintessa JAEA

Introduction: Importance

- Normally uplift is a special perturbation scenario, but due to the Japanese tectonic setting, the impact of uplift and erosion must be considered as part of "likely" repository evolution. Technical & communication issues.
- To compare potential sites and disposal concepts, the treatment of uplift / erosion must be as realistic as possible.
- Even in Japan, significant impacts will occur only in the far future (after hundreds of ky) and develop very slowly.
- Inevitably, there are great uncertainties with extrapolations of laboratory studies to investigate the processes involved and therefore natural analogues have great potential here.

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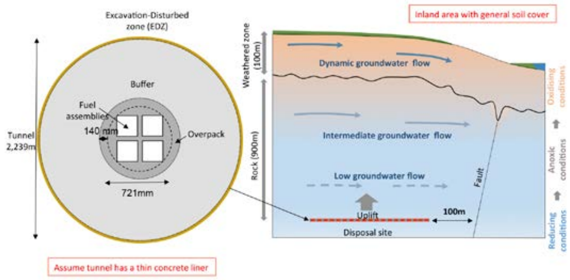
Introduction: Starting point (EDZ)



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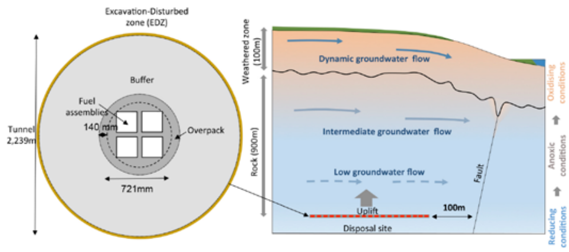
If uplift is negligible



If little uplift over the assessment time (< 100 m in 3 My, say):
•Far-field can be considered steady-state (no net change in properties)
•Biosphere requires consideration of different, site-specific variants as a result of climate change,...
•Gradual evolution of the EBS needs to be explicitly considered

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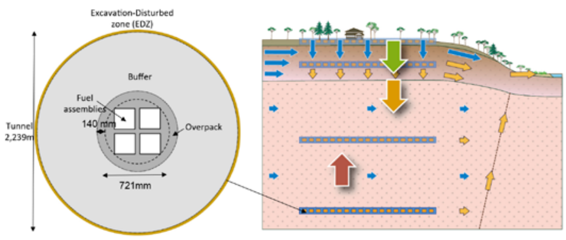
If uplift is significant



For period until uplift > 100 m (> 300 ky, say), scenario storyboard as for the negligible uplift case. Thereafter need to consider
•Uplift > 100 m but < 700 m (> 2 My on average, but variability over repository footprint to be expected)
•Uplift into oxidised / weathered zone (> 2.5 My on average)
•Erosion of the near field (> ≈ 3 My)

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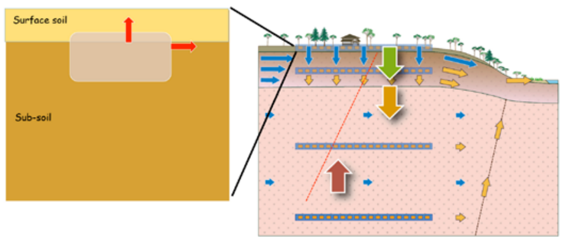
Erosion



- EBS completely degraded – represented only as region with high Fe content and possible high U (maybe also Np & Cs) associated irreversibly with oxidised phases
- Release of activity into oxidising water as a result of biochemical reactions in soil or as direct erosion / uptake into food chain
- Dose also from external gamma exposure
- Assessed by special biosphere model

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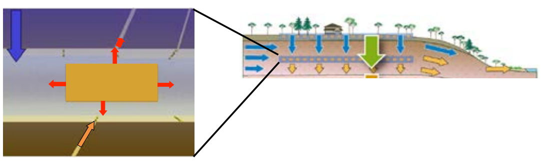
Challenges associated with near surface



- When repository at depths > 100 m or so, geochemical conditions fairly constant and effects of uplift mainly physical (fracturing, increased water flow), which are relatively easy to model
- As conditions gradually become more oxidising, it is assumed that remaining long-lived RN in the near-field are dispersed due to increasing solubilities.
- Although this is conservative for GW scenarios, this is non-conservative for erosion scenarios: evidence is thus required to demonstrate that the degraded near field cannot contain high activity levels when it is uplifted into the soil zone or that a pulse release of RN does not occur when the environment becomes oxidising
- This is possibly a minor concern for HLW, but could be serious for SF (giving doses >> background in Japan)

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Questions

- How long is it before uplift causes significant alteration of performance?
- REALISTICALLY – how is performance modified when...
  - The geosphere barrier is significantly modified?
  - The repository nears the surface?
    - Geochemical conditions slowly evolve
    - Gradual and heterogeneous weathering occurs
    - RN may be released (desorbed/dissolved) or immobilised in solid phases

The first question and the first part of the second one are relevant for all countries, the last one – for countries with high uplift/erosion rates

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To answer these questions

- The following quantitative models / data are available:
  - EBS material degradation
  - RN release & transport in the EBS
  - Modification of barrier properties of FF
  - RN transport in the FF
- How can models & data be validated – with a special focus on RN release & transport?
- What data is already available from NAs for this purpose?

The aim would be to find a natural system in which massive iron oxides within a compact clay are being uplifted into the soil zone: ideally these would be associated with U mineralisation

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What do we want from our NA?

- Analogue of a repository (relevant geochemical anomaly – massive iron, U or other relevant mineralisation)
- Active tectonic area with high uplift and erosion rates
- Possibility to assess changes in solubility and sorption / transport of key RN resulting from:
  - the perturbation of the EBS (near-field)
  - modification of the far-field

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Examples of NAs with data that could mined

- U ore body being intercepted by redox front: e.g. Poços de Caldas

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Advantage – plenty of data available

Comprehensive and timely production of a range of documentation

- 15 volume technical report series (with rigorous internal peer review)
- special volume of peer-reviewed papers (also published as book)
- many other papers in Journals, conference proceedings, magazines, ...
- special edition of Nagra Bulletin
- Ph.D. thesis, Uni Bern

+ possibility getting new data if necessary

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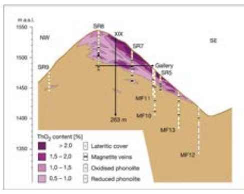
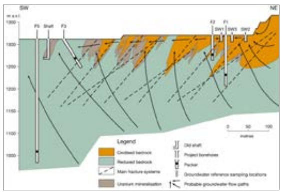
Morro do Ferro — Osamu Utsumi





- Ore body distribution mapped
- Hydrogeology defined
- Key locations for sampling water chemistry and examining colloid transport identified

- U mineralisation and redox fronts mapped
- Hydrogeology defined under both undisturbed conditions and after mining
- Key sample locations defined

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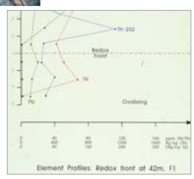
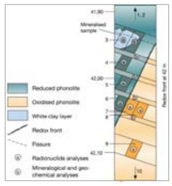
Redox front processes

Regional evaluation and detailed study of fine structure:

- Geometry controlled by fracture zones, preferential flow paths
- Natural series profiles and topographic analysis consistent with very slowly moving fronts
- Trace elements indicate active chemistry around fronts
- Strong indications of microbial catalysis of reactions at both sides of front





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Redox front processes: data to use

- Estimation of rate of redox front moving
  - Simple: supply rate of oxidants + simple chemical reactions
  - More sophisticated: + preferential transport of oxidants along fissures to predict the evolution of the form of the front – possibly oversophisticated for general uplift and erosion scenario, site dependent
- Kinetics of the chemical reactions at the front
  - Equilibrium model + solute transport
  - Transport models: do not explain (only predicts) variation between different redox fronts and trace-element distribution between them - probable issue for the future investigations
  - Other possible data (maybe missing): colloids and redox front

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Possible data from Poços de Caldas

Validation of models taking into account oxidising conditions:

- time sequence of alteration of iron oxides and host rock, together with associated U / trace elements (e.g. Cs, Se,...)
- RN transport (diffusion/solubility) under these conditions
- Role of microbiology in RN transport (especially relevent for the soil-zone)
- Role of colloids in RN transport

First, check data available, then identify main knowledge gaps that require further investigation

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One more time: What do we want from our NA?

- Analogue of repository (relevant geochemical anomaly – massive iron, U or other relevant mineralisation)
- Active tectonic area with high uplift and erosion rates
- Possibility to assess changes in solubility and sorption / transport of key RN resulting from:
  - the perturbation of the EBS (near-field)
  - modification of the far-field

Any further ideas for NA sites are welcomed!

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Conclusions

- Uplift/erosion scenarios important
  - Technical
  - PR
- NA study important for safety case / communication
- Lots of data available, the next step will be to analyse relevance and identify gaps (e.g. alteration time for the overpack, backfill and host rock; RN transport in changing conditions including microbiology, colloids, etc.)
- Good potential for international collaboration between programmes where such scenarios are relevant (Japan, Korea, Taiwan,...)

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Thank you for your attention!

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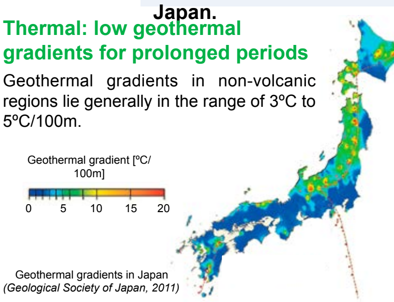
Nation-wide geoscientific information to identify "stable" geological environments

NUMO 原子力発電環境整備機構
Saori Yamada
Nuclear Waste Management Organization of Japan

NUMO has assembled the latest geoscientific information nation-wide and updated the NUMO geoscientific database, which would be used to identify "stable" deep geological environments in Japan.

Thermal: low geothermal gradients for prolonged periods

Geothermal gradients in non-volcanic regions lie generally in the range of 3ºC to 5ºC/100m.



Geothermal gradients in Japan
(Geological Society of Japan, 2011)

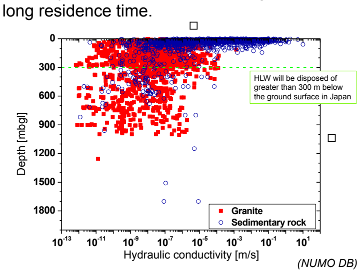
Groundwater ages

Place	Rock type	Depth (mbgl)	Groundwater age (ky)	Methods
Horonobe (Hokkaido)	Miocene sedimentary rock	700	10,000	$^{4}He,^{36}Cl$
Yokosuka (Kanagawa)	Miocene sedimentary rock	210-500	7,000	$^{14}C,^{4}He,^{36}Cl$
Kushiro (Hokkaido)	Cretaceous sedimentary rock	750	2,000	^{36}Cl
Kobe (Hyogo)	Pliocene sedimentary rock	1,500	230~25	^{4}He
Rokkasho (Aomori)	Miocene sedimentary rock	180	80~50	^{4}He
Mizunami (Gifu)	Cretaceous granite	1,000	20~10	$^{14}C,^{4}He$

(Morikawa et al., 2005; Mahara et al., 2006; Hasegawa et al., 2010, 2013; Nakata and Hasegawa, 2010, 2011)

Hydrological: slow groundwater movements due to low hydraulic conductivities and/or low hydraulic gradients

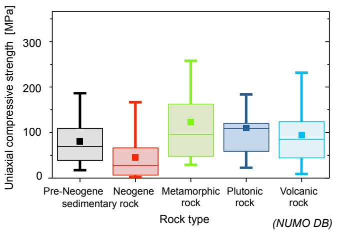
High hydraulic gradients normally associated with low hydraulic conductivities and *vice versa* indicate the occurrence of slow groundwater movements, which is supported by the evidence that such groundwater has a very long residence time.



(NUMO DB)

Mechanical: small rock deformation

Rock mechanical databases updated to date show that deep subsurface rock formations have sufficient mechanical strength against the magnitude of initial stress and generally small stress anisotropy.

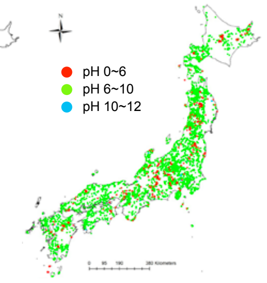


(NUMO DB)

Favourable THMC conditions in deep geological environments

The latest geoscientific knowledge demonstrates that the favourable THMC conditions most likely prevail in deep geological environments in Japan.

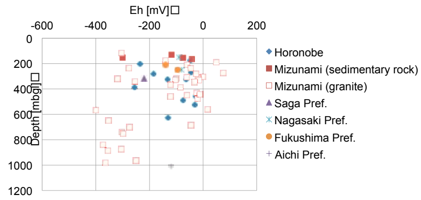
Stress anisotropy
(Cho et al., 2009; Nakamura et al., 2011; The Research Group for Crustal Stress in western Japan, 2011; Sato et al., 2013)



Distribution of groundwater pH
(AIST, 2009)

Geochemical: groundwater with near neutral pH, reducing conditions and DIC concentrations less than 0.5 mol/dm³

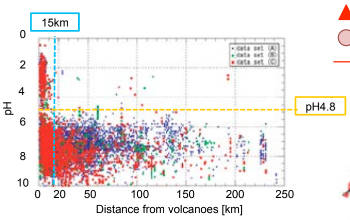
Groundwater with pH values between 6 and 9, more negative Eh values and DIC concentrations of smaller than 0.1 mol/dm³ commonly occurs in deep geological environments.



(JNC, 2003, 2004; Ioka et al., 2007; Iwatsuki et al., 2009; Amano et al., 2012; JAEA DB)

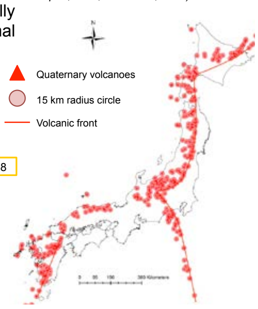
Igneous activity

Elevated heat flow and generation of heat sources by geothermal activity and mixing with acid groundwater by penetration of volcanic thermal fluids would have potentially disruptive impacts on the favourable thermal and geochemical conditions respectively.

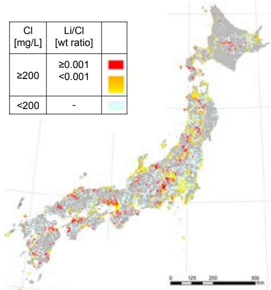


(Asamori et al., 2002)

Exclusion of areas within a 15 km radius circle around the centre of Quaternary volcanoes



Exclusion areas around Quaternary volcanoes
(AIST, 2013)



Distribution of groundwater Li/Cl ratios
(AIST Homepage)

Non-volcanic thermal or deep-seated fluids

Mixing with low pH or high DIC groundwater by penetration of non-volcanic thermal or deep-seated fluids could have potentially adverse impacts on the favourable geochemical conditions. Groundwater Li/Cl ratios could be used as an indicator of the occurrence of deep-seated fluids.

Exclusion of areas where low pH (<4.8), high DIC (>0.5 mol/dm³) or high T(>100ºC) fluids occur

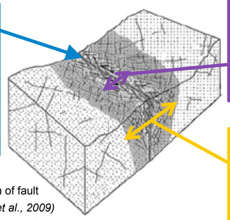
Exclusion criteria

The potentially significant impacts of natural disruptive events/processes that would appear likely to perturb the favourable THMC conditions in deep geological environments are to be precluded, in principle, during initial screening and literature survey.

Earthquake and fault movement

High crustal displacement at a disposal depth, changes in groundwater flow paths and penetration of oxidising surface water by increase in hydraulic conductivity in and around a fault would have potentially significant impacts on the favourable mechanical, hydrological and geochemical conditions respectively.

Fault gauge
Derivation of high T fluids >350ºC by earthquake related frictional heating and their retention within fault gauge at Chelungpu Fault
(Ishikawa et al., 2008)



Schematic illustration of fault
(modified from Yoshida et al., 2009)

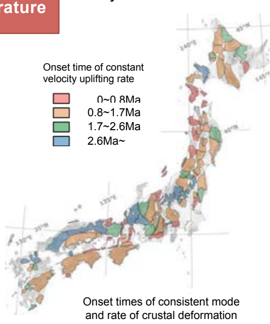
Crush zone (1/350~1/150 of fault length)
Hydraulic fracturing by fluid upwelling and circulation; hydrothermal alteration and frictional melting at 150~280ºC within a crush zone at Nojima Fault
(Boullier et al., 2004a,b)

Process zone (1/100 of fault length)
Increases in the frequency of joints, minor faults and microcracks induced by faulting and groundwater circulation
(Kanaori, 2001; Yoshida et al., 2009)

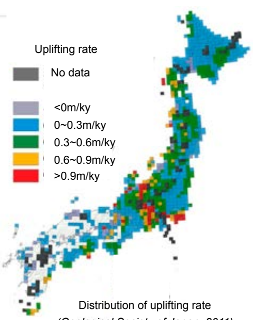
Exclusion of areas transected by active faults and also fault process zone that has a width of about 1/100 of the fault trace length in the vicinity of a fault

Uplift/erosion

Significant uplift/erosion would significantly reduce the thickness of the geological barrier. However, as the potentially accumulating impacts of slowly developing uplift/erosion cannot be precluded, it is necessary to ensure that the extent over which the THMC conditions vary with time would be insignificant.



Onset times of consistent mode and rate of crustal deformation
(Yasue et al., 2014)



Distribution of uplifting rate
(Geological Society of Japan, 2011)

Exclusion of areas where uplift/erosion amounts to greater than 300 m during the last 100,000 years

Nation-wide geoscientific information to identify "stable" geological environments

NUMO 原子力発電環境整備機構

Saori Yamada

Nuclear Waste Management Organization of Japan

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Bentonite analogues for geological disposal of radioactive waste – current status and future outlook

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Introduction

Numerous studies on bentonites, focussing on bentonite interaction with other components of the engineered barrier system (EBS) and a range of host rock environments, are present in the literature. In this article (Reijonen & Alexander, 2015a), recent bentonite NA studies were briefly reviewed and gaps in the current literature identified. The aim was to:

1) suggest where relevant new information could be obtained by data mining published bentonite NA studies with a new focus of current safety case requirements
2) collect relevant information by revisiting known bentonite analogue sites and conducting investigations with more appropriate (and up-to-date) analytical techniques
3) identify novel study sites where, for example, bentonite longevity in very dilute to highly saline groundwater conditions can be studied (cf. Reijonen & Marcos, 2015).

It must be noted that the use of NAs in safety case development is likely to be site and repository design-specific in nature and thus emphasis is placed on the appropriate use of relevant NA data on bentonite longevity (see also discussion in Alexander et al., 2014; 2015a; Reijonen et al., 2015).

Conclusions

This brief overview of several recent NA studies has shown that, although most processes relevant to the long-term behaviour of EBS bentonite under repository conditions could be addressed by NA studies (Table 1), *few have been done to an appropriate level*. Either the boundary conditions have not been fully defined or the environments studied were not as relevant as hoped or the studies simply were not focussed enough on supplying data for the safety case (i.e. bottom-up vs. top-down studies; cf. Alexander et al. 2014). This is reflected in the strength of the qualitative discussion and strong process understanding regarding engineered clay barriers and their longevity in general but, on the other hand, in the lack of usable quantitative data.

Nevertheless, it is clear from Table 1 that many NAs of relevance do exist and much information of use in the safety case, both quantitative and qualitative, could be provided. These data could be produced by a mixture of means, including:

- obtaining relevant information by data mining published NA studies with a new focus of current safety case requirements (cf. McKinley & Alexander, 2007; Alexander et al., 2007)
- obtaining relevant information by revisiting known bentonite analogue sites and conducting investigations with more appropriate analytical techniques (cf. Milodowski et al., 2015)
- identifying novel study sites where, for example, long-term stability of bentonite in very low salinity groundwaters can be studied (cf. Reijonen & Marcos, 2015)

Table 1: *An overview of the main processes of relevance to the long-term behavior of bentonite in the repository EBS, backfill and seals with recommendations for focussed NA studies.*

Process studied	Future recommendations regarding NA studies
Thermal effects on bentonite have been studied through NAs, but no information is available on the relevant temperature regime (Posiva 2012a) and will need to be carefully considered in the design and the development of the safety case (Wilson et al. 2011). Wyoming bentonites hold potential for both thermal and geochemical studies (Laine & Karttunen 2010).	Return to more relevant sites (e.g. Busachi in Sardinia; cf. Laine & Karttunen, 2010) and apply a range of more relevant analytical techniques to better understand the site history and temperature profiles (see discussion in section 3). The Clay Spur bed (Wyoming) should be studied in order to better constrain the thermal and hydrogeological boundary conditions preserving the deposit.
Bentonite resaturation: large-scale experiments suggest that the theory and laboratory-based understanding of bentonite resaturation may not be a good description of the process at full scale. Resaturation times may be significantly longer than previously predicted, and full resaturation may not occur in some cases (Wilson et al. 2011).	Boundary conditions for dry and wet thermal reactions can be obtained via new NA studies at existing sites (such as Busachi in Sardinia).
The evidence of bentonite plasticity is interesting, but this topic is not treated quantitatively enough to warrant use in safety assessments (Posiva 2012a).	The approach of Keto (1999) should be repeated plus the process should be included into current URL experiments (e.g. FEBEX; Gens et al., 2002). In addition, try to find a site where canister sinking could be studied by means of boulder fall on bentonite. See also discussions in Reijonen & Alexander (2015b) and Alexander et al. (2015b)
In the case of iron-bentonite interactions, useful NA data are very limited (cf. Marcos, 2003; Wersin et al., 2007). Steel/iron corrosion product could react with bentonite to produce reduced swelling or non-swelling minerals, thereby causing changes in swelling pressure, bentonite cementation, and loss of sorption sites. The kinetics of smectite alteration to iron-rich minerals is not well understood (Wilson et al., 2011). Identified need for future studies (e.g. Pelayo et al., 2011 type of NAs; Morron de Mateo).	Existing NA studies are not relevant enough for KBS-3 or other designs, although general observations from natural systems and the NF-PRO URL experiment (Kickmaier et al., 2005) suggest uptake of iron could reduce swelling pressures. These data need to be integrated with a more appropriate NA study.
Cu-bentonite interaction: only limited information available (Posiva, 2012a)	From Kronan cannon, qualitative data are available, which should be more rigorously integrated with existing/new laboratory data.
Effect on bentonite on waste glass in designs with no container, or very thin container, is not understood. Glass dissolution may increase in contact with some clays, but mechanisms are poorly understood (Wilson et al., 2011).	Sites where natural glasses are in contact with bentonites exist. However, the differences between natural glasses and vitrified radioactive waste should not be forgotten (cf. Miller et al., 2000)
Bentonite freezing and thawing processes: only general considerations are available (Posiva, 2012a).	A NA study in glaciated terrain could provide more reliable tools for extrapolating the experimental evidence to safety case time scales.
Chemical erosion of bentonite: There is no NA study to support or discount the process (dilute conditions during glacial melt water intrusion; Posiva, 2012a). Further research and development work is likely to be needed on erosion of bentonite, especially for specific conditions (Wilson et al., 2011). New work has just been initiated by Posiva (see Reijonen & Marcos, this workshop)	Montmorillonite has been observed to occur as fracture filling mineral e.g. at repository sites such as Olkiluoto in Finland and Forsmark in Sweden providing potential in situ NA sites for montmorillonite stability under varying geochemical conditions (see discussion in section 4 of Reijonen & Alexander, 2015a) (also many other sites for similar studies have been identified). Also studies related to the erosional histories of known bentonite deposits under the effects of the last glaciation or by meteoric waters could be of interest.
Effect of saline groundwater on bentonite: saline conditions may need to be further considered (Wilson et al. 2011). The impact of temperature gradients on the accumulation and dissolution of salts in the vicinity of canisters embedded in clay buffer also needs to be assessed. Experimental/modelling/NA work may be required to increase understanding if bentonite is to be exposed to unusually high Mg concentrations (i.e. Mg-rich brine). See also Milodowski et al. (2015).	Approach proposed in section 5 is worthwhile for bentonite-saline groundwater interaction and understanding the impact of Mg-rich brines. Temperature gradient impact on salt accumulation could be addressed at Busachi (see above).
OPC cement-bentonite interaction: experimental work to reduce uncertainties concerning smectite alteration rates and bentonite degradation rates under highly alkaline conditions needs to be extended to include integrated NA studies (Sidborn et al., 2014)	Little hard evidence, but could be addressed at the Khushaym Matruk site in Jordan (cf. Pitty & Alexander, 2011; Alexander, 2012). At Cyprus there is a site where pH 12 is observed (see Alexander et al., 2012).
Low alkali cement-bentonite interaction: NA studies at Cyprus are finalised (Alexander et al., 2012; Alexander & Milodowski, 2015; Milodowski et al., 2015) and ongoing in the Philippines.	Results (Cyprus) clearly show limited reaction; additional information from the Philippines study can contribute to confidence on the process understanding.
Coupled processes: a key point to note is that, although for simplicity the reactions and processes are discussed as individual topics, many of the processes of relevance to the long-term behaviour of bentonite will be strongly coupled and cannot be considered, or optimised, in isolation (Wilson et al., 2011).	Focussing on coupled processes should be introduced more strongly in NA research in general. Potential sites exist e.g. for cement-bentonite alteration coupled with thermal alteration and these should be studied.
Assessment of clay materials with respect to coupled THMCB behaviour: (including gas conductivity and ion diffusivity) needs to be made in order to obtain a basis for optimal selection of one specific candidate material.	Despite recent advances through the EU projects DECOVALEX (DECOVALEX, 2011), FORGE (FORGE, 2013) and PEBS (Schäfers & Fahland, 2014), there remains a general need to increase understanding of coupled processes, as seen in the latest stage of DECOVALEX (DECOVALEX, 2014).
Performance of bentonite – sand/rock mixtures: these are being considered more as repository designs are optimised	In reality, the physical and mineralogical properties of many natural bentonites are more akin to bentonite-sand or bentonite-rock mixtures (Alexander & Milodowski, 2015), allowing the long-term behaviour of these mixtures to be assessed quantitatively. To date, only a tentative assessment has been carried out, but this has shown the potential for further, detailed studies.
Consideration of smectites other than montmorillonites: when considering other clays (e.g. saponite, Wilson et al., 2011 or Friedland clay, Posiva, 2012b), the first action should be a study of the source deposit and what boundary conditions can be justified based on this (compare e.g. Clay Spur studies in case of MX-80; Laine & Karttunen, 2010).	Alteration patterns for trioctahedral smectites like saponite are well known in nature, providing a lot of potential for NA studies in various geological environments.
Microbial processes: it has been generally felt that the bentonite buffer would minimise microbial populations due to the restriction of groundwater flow, but more recent studies (e.g. Stroes-Gascoyne, 2010) of several bentonite-backfill and sealing materials used in large-scale experiments at AECL's Whiteshell URL, have demonstrated the presence of culturable populations in all bulk materials.	These few, URL-based examples should be checked by examining natural bentonites for *in situ* microbial populations. Could most easily be carried out in conjunction with studies on other aspects of bentonite (e.g. saline reaction). Certainly, there is a suggestion of microbial processes in the bentonites studied in the Philippines (Alexander et al., 2008).

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14. Ruskeeniemi, Aaltonen, Käpyaho, Lindberg, Mattila, Ojala, Palmu & Sutinen: A new approach to screen and investigate postglacial faults in Finland.

A new approach to screen and investigate postglacial faults in Finland

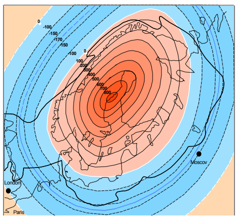
Timo Ruskeeniemi, Ismo Aaltonen, Asko Käpyaho, Antero Lindberg, Jussi Mattila, Antti Ojala, Jukka-Pekka Palmu and Raimo Sutinen

Outline

- Glaciations and postglacial faults (PGF)
- LiDAR – new tool for PGF work
- Project PGSdyn
- Examples of the conducted work

Fennoscandian ice ages

- During the Quaternary (2.6 Ma- present) at least 20 stages with substancial ice sheets (Ehlers & Gibbard 2011; Gibbard & Cohen 2008)
- Early, Middle and Late Weichselian glaciations (115-10 ka)
  - Late Weichselian (c. 28 000 – 10 000 a)
    - Max bedrock depression 800-900 m
    - Current uplift rate 3-9 mm/a
    - Remaining uplift 100-150 m



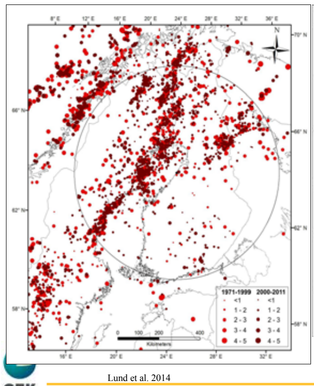
Postglacial faults (Muir Wood 1993; Olesen et al. 2004)

Criteria for the identification of late- and postglacial fault

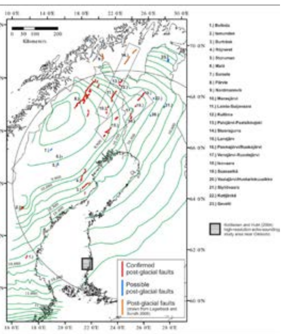
1) Offset of an originally continuous surface or sedimentary sequence of postglacial or late-glacial age.
2) Reasonably consistent direction and amount of slip along the length of the fault.
3) The ratio of displacement to overall length of the feature should be less than 1/1,000. For most faults this ratio is between 1/1,000 and 1/10,000.
4) Exclusion of gravity-induced sliding as the driving mechanism of faults in areas of moderate to high relief.
5) No signs of glacial modification (such as striation or ice-plucking) of fault scarps, especially those controlled by banding, bedding or schistosity.
6) Exclusion of mechanisms such as glaciotectonics (ice push features), collapse due to ice melting and differential compaction or deposition over a pre-existing erosional scarp being the cause of an apparent offset in overburden.

Reliability classification of late- and postglacial faults

(A) Almost certainly neotectonics
(B) Probably neotectonics
(C) Possibly neotectonics
(D) Probably not neotectonics
(E) Very unlikely to be neotectonics



Lund et al. 2014

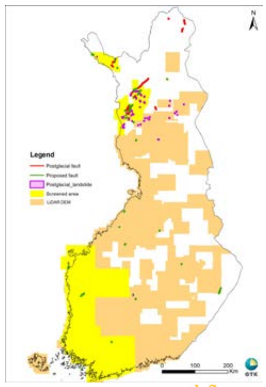


Sutinen et al. 2014c

Airborne LiDAR (Light Detection And Ranging)

High-resolution digital elevation model (2 m grid, vertical resolution ~ 30 cm)

Processing of cloud point data
-Multidirectional hillsiding and vertical exaggaration
-Slope (tilt) theme



Postglacial faults – why they are important?

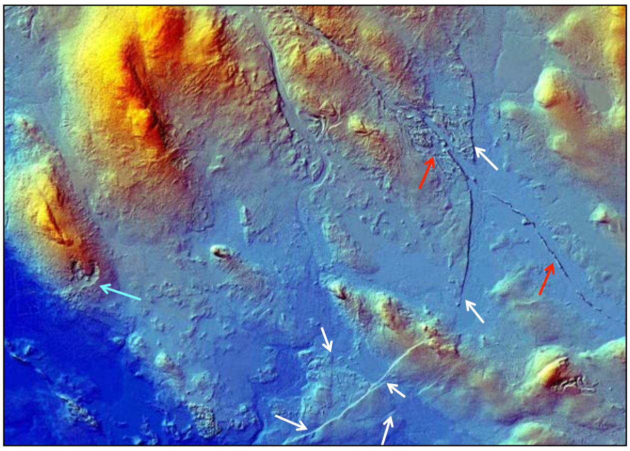
- Recent bedrock movements induced by an event expected to re-occur in 60 000 years and several times in 1 Ma?
- Potentially inducing large earthquakes
- Assumed to be reactivated old structures – is it really so?
- Is there a temporal reactivation pattern? Reloading period?
- Geometry of the faults – is the fault turning subhorizontal deeper down?
- Tectonics behind: is the real reason the forcing from Mid-Atlantic ridge?
- Impact zone around the actual fault?

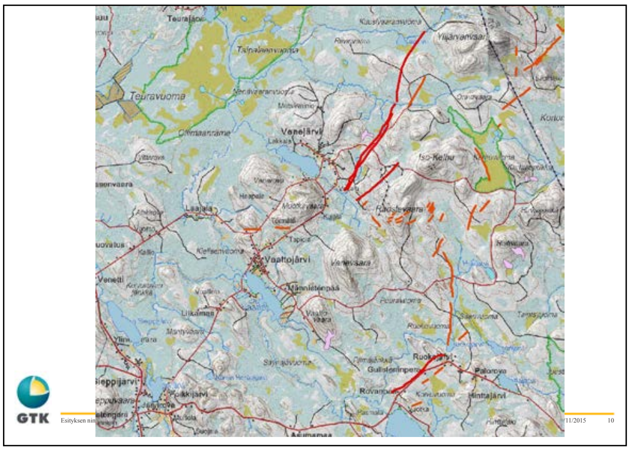
Postglacial faults and their dynamics (PGSdyn)

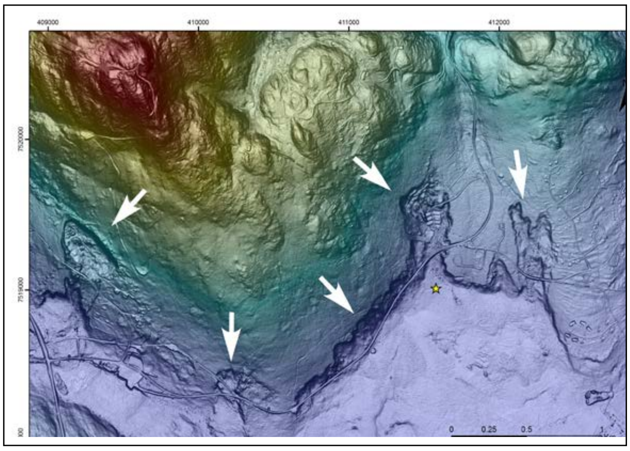
- Geol. Surv. of Finland & Posiva, 2014 -
- Aims:
  - Screening of the whole country using LiDAR based elevation model
    - Fault scarps, morphological features in Quaternary deposits possibly related to seismic activity
    - Location and characteristics (orientation, dimensions, geometry, date etc.)
  - Compilation and evaluation of historical data
  - Field investigations (geophysics, trenching, drilling….)
  - Storage in a specific database: Paleoseismic.gdb

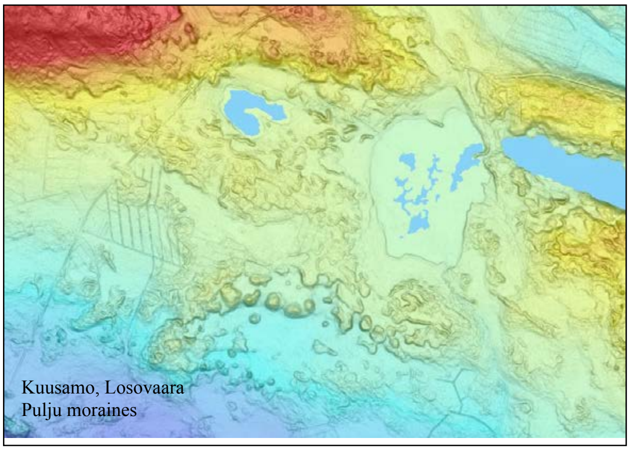
→ increased understanding of the reactivation mechanisms, internal geometry and properties of the faults
→ 'neotectonic' map of Finland
→ assessment of the stability of structures in Olkiluoto (NA aspect)

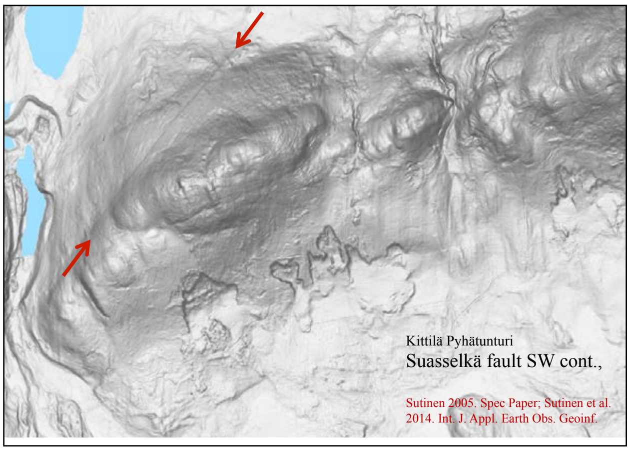
14. Ruskeeniemi, Aaltonen, Käpyaho, Lindberg, Mattila, Ojala, Palmu & Sutinen: A new approach to screen and investigate postglacial faults in Finland.

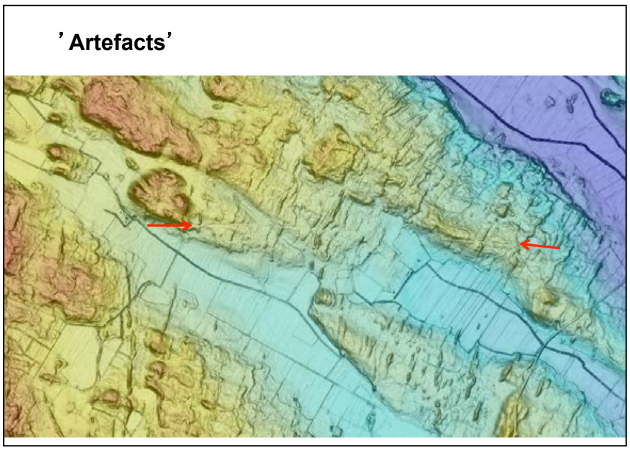












Confirmation – potential research tools

- Field observations
- Geophysics
  - Aiborne data, GPR, Seismics, Resistivity tomography
- Trenching
  - Quaternary geology: mapping, sampling, dating?....
  - Bedrock geology: lithologies, fracturing, dating?, fluid inclusions…
- Drilling
  - Logging, video survey, hydraulic testing
- Dating of landslides: 14C-dating of organic material
- Groundwater sampling

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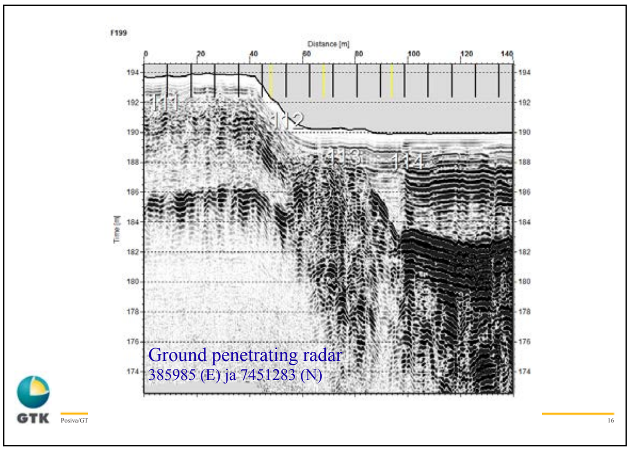

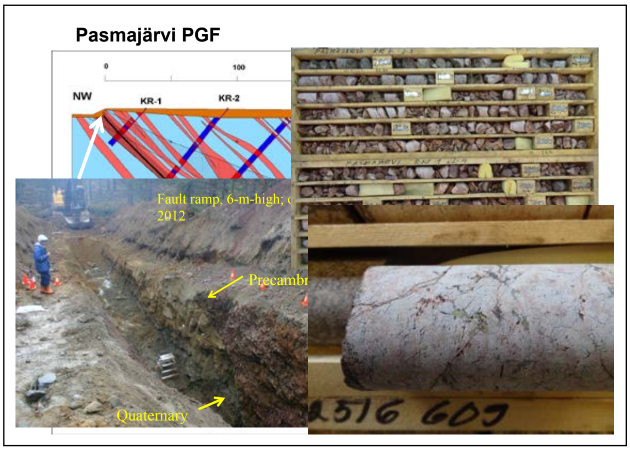
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KR-2
Fault ramp, 6-m-high;
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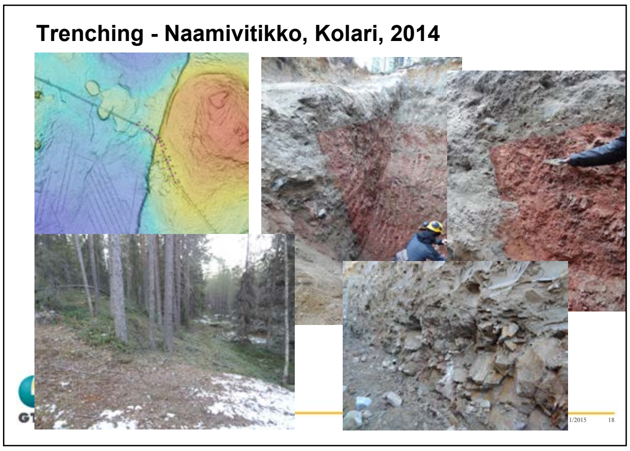
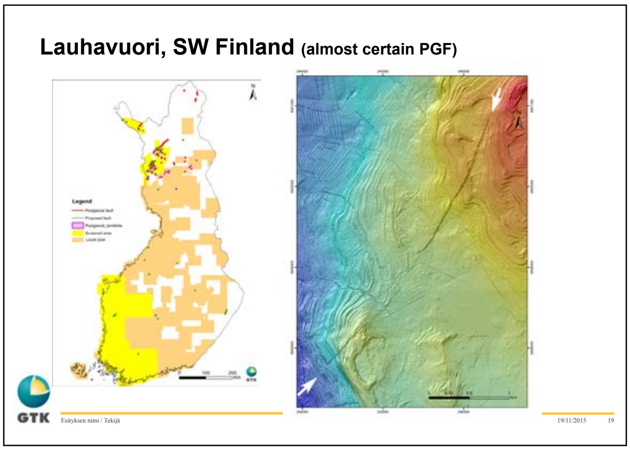
Quaternary


Trenching - Naamivitikko, Kolari, 2014


Lauhavuori, SW Finland (almost certain PGF)
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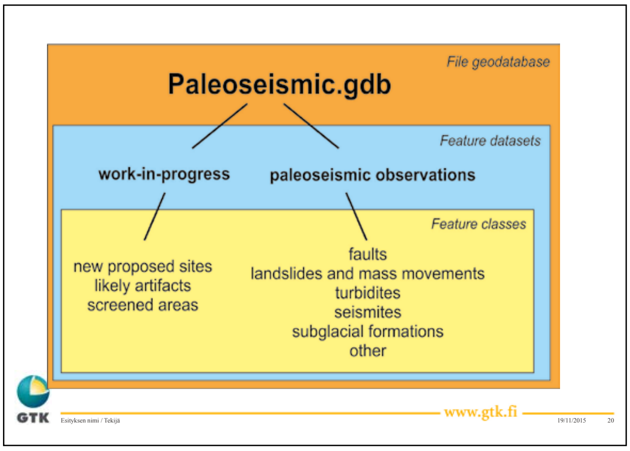
File geodatabase
Paleoseismic.gdb
Feature datasets
work-in-progress
paleoseismic observations
Feature classes
new proposed sites
likely artifacts
screened areas
faults
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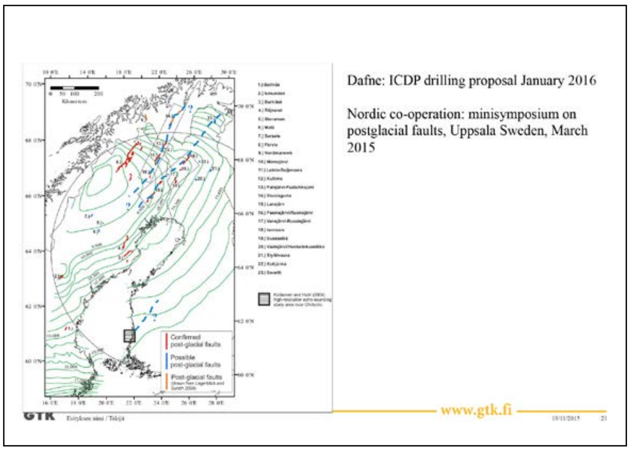
20


Dafne: ICDP drilling proposal January 2016
Nordic co-operation: minisymposium on postglacial faults, Uppsala Sweden, March 2015
Confirmed post-glacial faults
Possible post-glacial faults
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21


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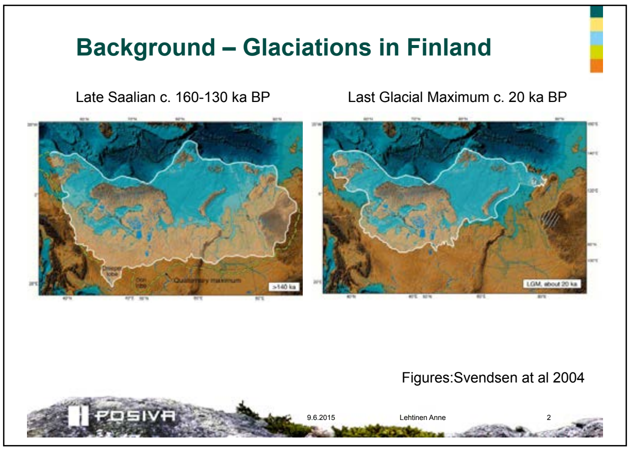
Thank you!
GTK
Esityksen nimi / Tekijä
www.gtk.fi
19/11/2015
22


PALEOHYDROGEOLOGICAL INVESTIGATIONS OF THE GLACIAL WATER EFFECTS IN THE VICINITY OF END MORAINE (PROJECT SAIMAA)
Anne Lehtinen
10th June 2015
POSIVA


Background – Glaciations in Finland
Late Saalian c. 160-130 ka BP
Last Glacial Maximum c. 20 ka BP
Figures:Svendsen at al 2004
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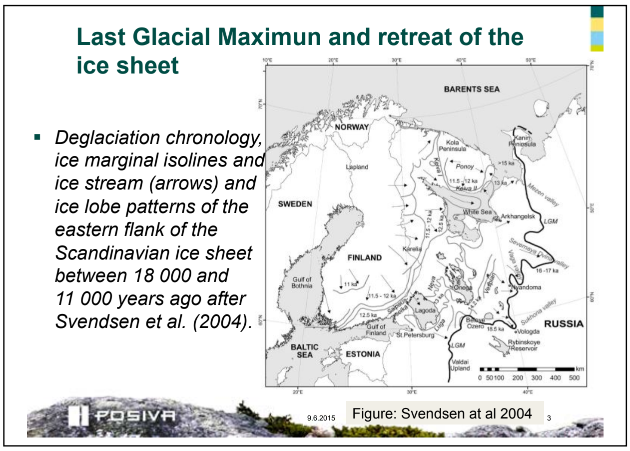
Last Glacial Maximun and retreat of the ice sheet
Deglaciation chronology, ice marginal isolines and ice stream (arrows) and ice lobe patterns of the eastern flank of the Scandinavian ice sheet between 18 000 and 11 000 years ago after Svendsen et al. (2004).
BARENTS SEA
NORWAY
SWEDEN
FINLAND
RUSSIA
BALTIC SEA
ESTONIA
Figure: Svendsen at al 2004
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3


Formation of the Salpausselkä end morains (SSI and SSII)
Salpausselkä I and II end moraines were deposited between ca. 12 800 to 11 500 years ago.
During the formation of Salpausselkä II the ice margin remained stationary ~700 years
Can this happen in Olkiluoto ?
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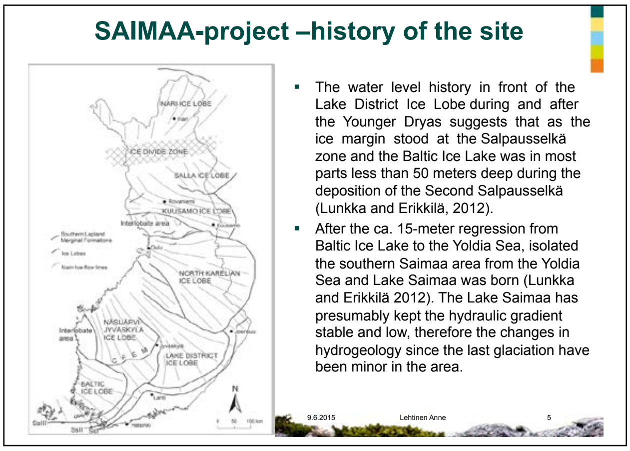
4


SAIMAA-project –history of the site
The water level history in front of the Lake District Ice Lobe during and after the Younger Dryas suggests that as the ice margin stood at the Salpausselkä zone and the Baltic Ice Lake was in most parts less than 50 meters deep during the deposition of the Second Salpausselkä (Lunkka and Erikkilä, 2012).
After the ca. 15-meter regression from Baltic Ice Lake to the Yoldia Sea, isolated the southern Saimaa area from the Yoldia Sea and Lake Saimaa was born (Lunkka and Erikkilä 2012). The Lake Saimaa has presumably kept the hydraulic gradient stable and low, therefore the changes in hydrogeology since the last glaciation have been minor in the area.
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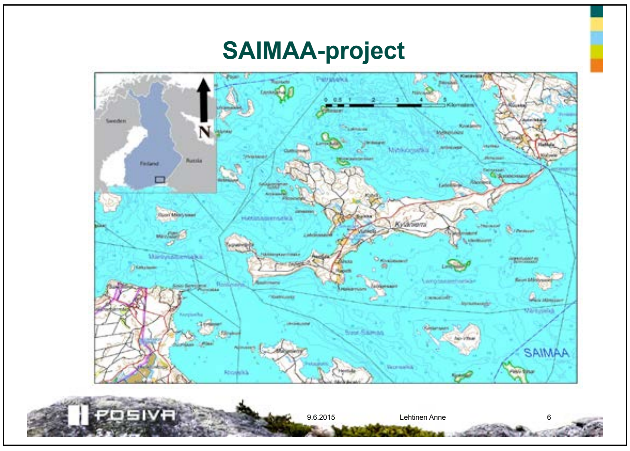
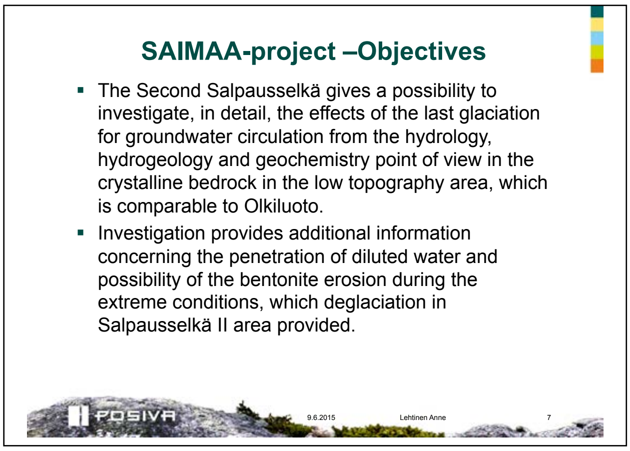
5


SAIMAA-project
SAIMAA
POSIVA
9.6.2015
Lehtinen Anne
6


SAIMAA-project –Objectives
The Second Salpausselkä gives a possibility to investigate, in detail, the effects of the last glaciation for groundwater circulation from the hydrology, hydrogeology and geochemistry point of view in the crystalline bedrock in the low topography area, which is comparable to Olkiluoto.
Investigation provides additional information concerning the penetration of diluted water and possibility of the bentonite erosion during the extreme conditions, which deglaciation in Salpausselkä II area provided.
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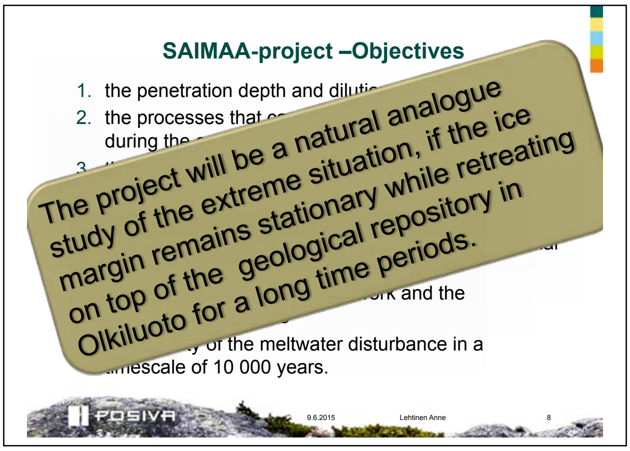
7


SAIMAA-project –Objectives
1. the penetration depth and dilutio
2. the processes that c
during the
The project will be a natural analogue study of the extreme situation, if the ice margin remains stationary while retreating on top of the geological repository in Olkiluoto for a long time periods.
rk and the
of the meltwater disturbance in a
timescale of 10 000 years.
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Lehtinen Anne
8

SAIMAA-project activities

9

SAIMAA-project schedule

- applying research permission from the Posiva board
- drilling
- matrixporewater sampling ?
- drillhole measurements
- multipacking the drillholes
- pressure, temperature, EC and pH monitoring
- watersampling
- hydrological and hydrogeochemical modelling
- implying the results for Posiva's safety assessment

POSIVA 9.6.2015 Lehtinen Anne 10

SAIMAA-preoject vs GAP

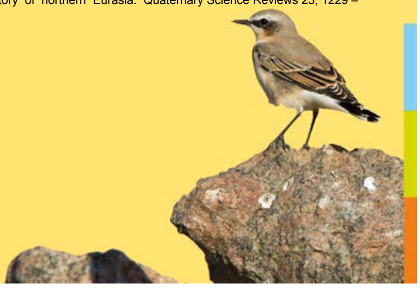
- GAP project has provided us new information about the effects of the ice sheet on deep hydrogeological systems
  - Results show that the hydrogeological system is connected to the ice sheet system, and the meltwater signature is seen in the repository depth.
- In the Lake Saimaa area, the hydraulic gradient has stayed stable and low, due to presence of the lake, since the last glaciation
  - The changes in hydrogeology have been minor in the area
- The project Saimaa concentrates on investigating the stability of the changes in a hydrogeological condition that a glaciation can effect in the hydrogeologically active area (in a timescale of 10 000 years) and provides the "worst case" scenario for the safety assessment for Olkiluoto.

POSIVA 9.6.2015 Lehtinen Anne 11

References:

Lunkka, J. P., Erikkilä, A. 2012. Behaviour of the Lake District Ice Lobe of the Scandinavian Ice Sheet During the Younger Dryas Chronozone 8ca. 12 800 – 11 500 years ago). POSIVA Working Report 2012-17. 54 pp.

Svendsen, J. I., Alexanderson, H., Astakhov, V. I., Demidov, I., Dowdeswell, J. A., Funder, S., Gataullin, V., Henriksen, M., Hjort, C., Houmark-Nielsen, M., Hubberten, H. W., Ingólfsson, Ó., Jakobsson, M., Kjær, K. H., Larsen, E., Lokrantz, H., Lunkka, J. P., Lyså, A., Mangerud, M., Matiouchkov, A., Murray, A., Möller, P., Niessen, F., Nikolskaya, O., Polyak, L., Saarnisto, M., Siegert, C., Siegert, M. J., Spielhagen, R., Stein, R. 2004. Late Quaternary ice sheet history of northern Eurasia. Quaternary Science Reviews 23, 1229 – 1271.



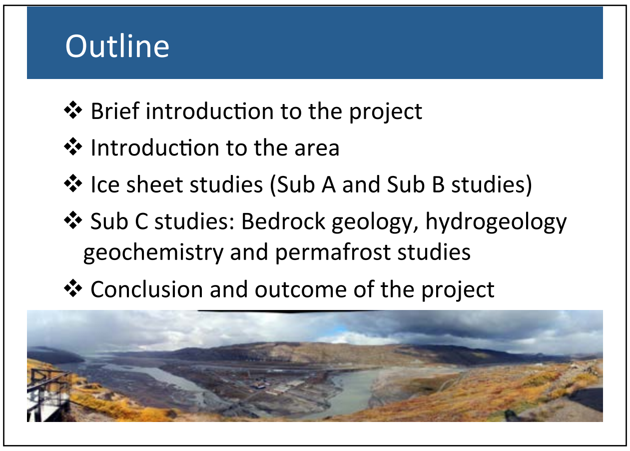
POSIVA

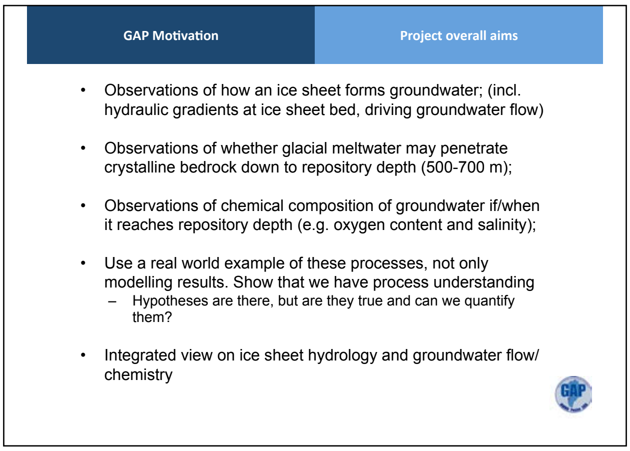
Kiitos

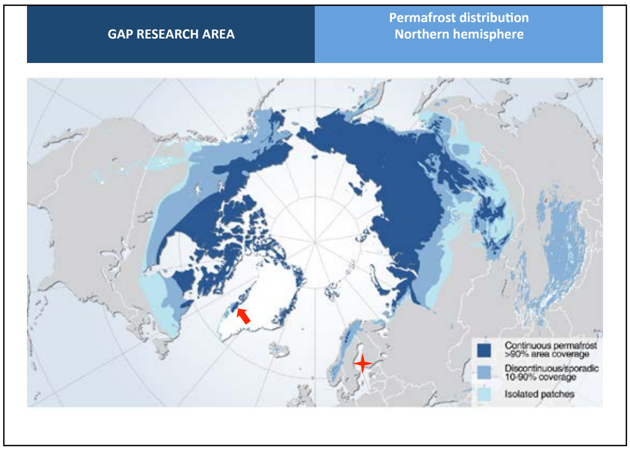
Thank you

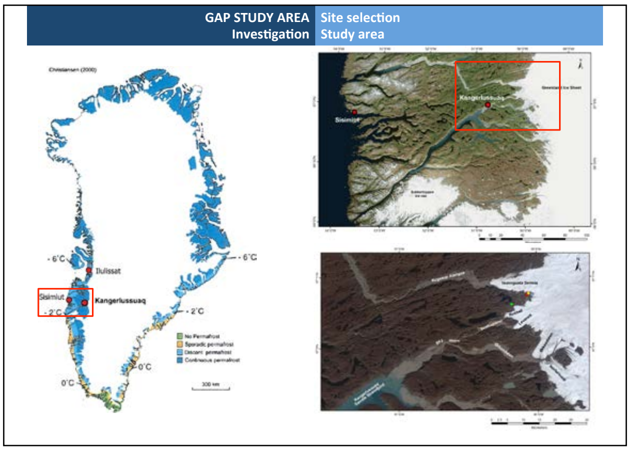
POSIVA

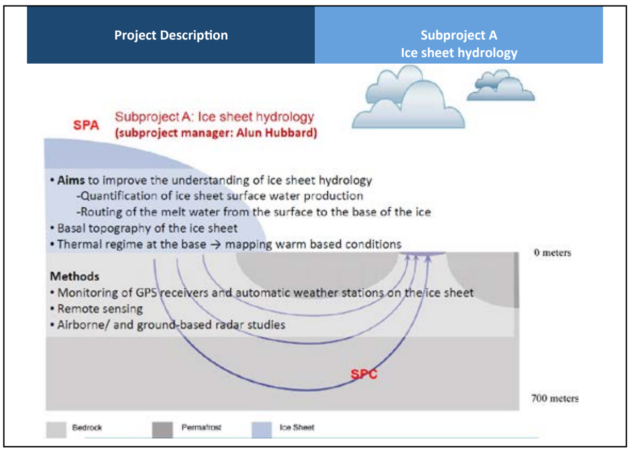
16. Engström, Lehtinen, Ruskeeniemi & Claesson Liljedahl: The Greenland analogue project – Project update.

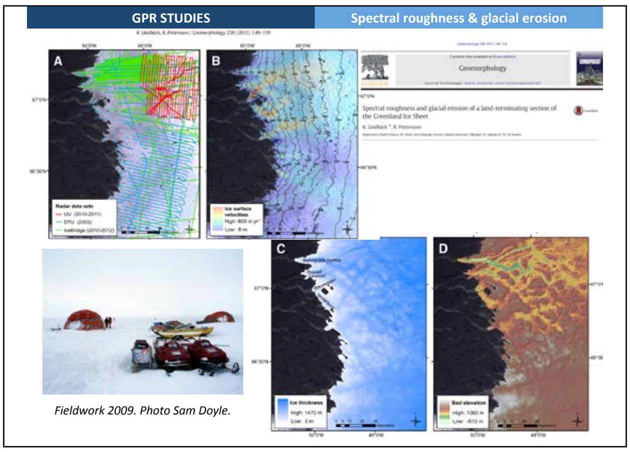


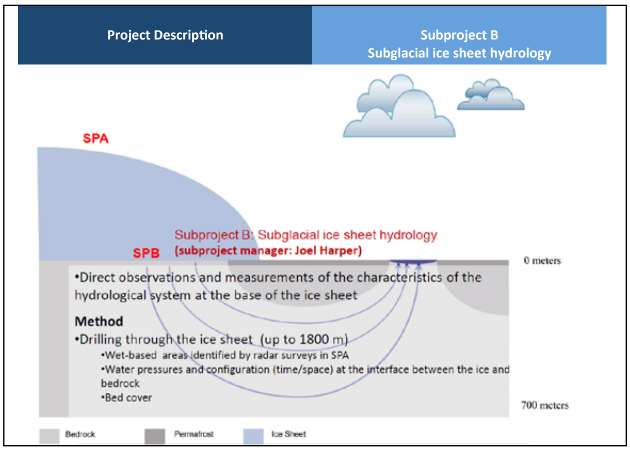




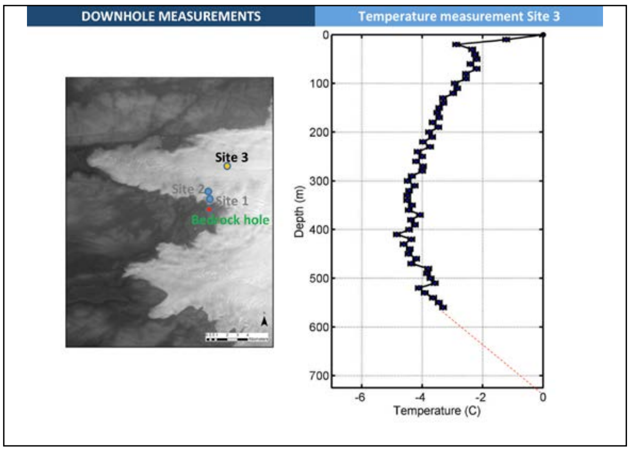


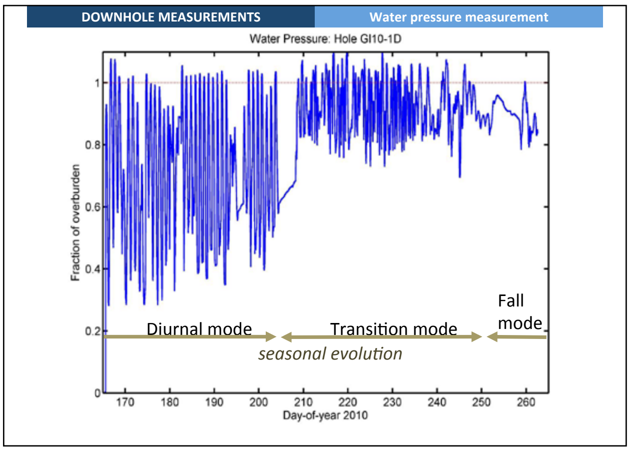


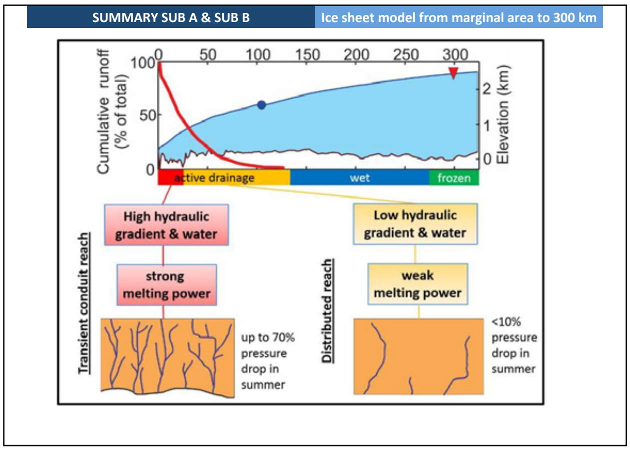


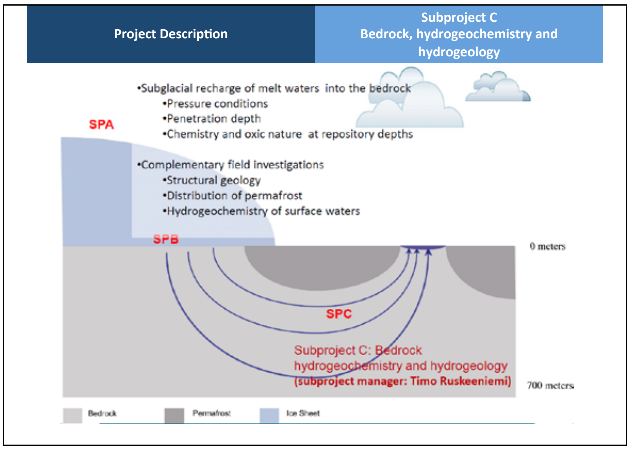

Project Description
Subproject B
Subglacial ice sheet hydrology
SPA
Subproject B: Subglacial ice sheet hydrology
SPB
(subproject manager: Joel Harper)
0 meters
•Direct observations and measurements of the characteristics of the hydrological system at the base of the ice sheet
Method
•Drilling through the ice sheet (up to 1800 m)
•Wet-based areas identified by radar surveys in SPA
•Water pressures and configuration (time/space) at the interface between the ice and bedrock
•Bed cover
700 meters
Bedrock
Permafrost
Ice Sheet

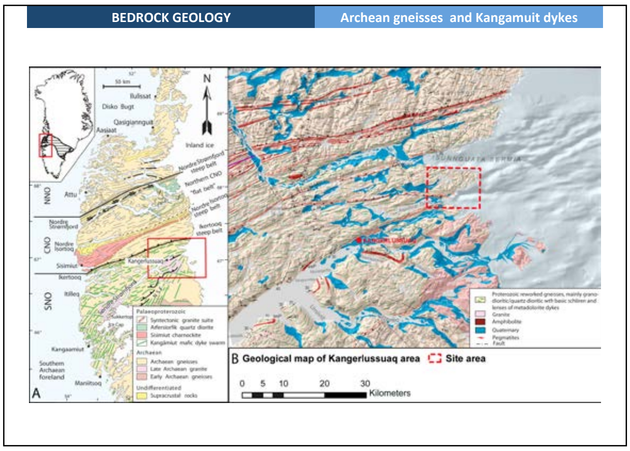

ICE DRILLING
Hot water ice drilling to the bed

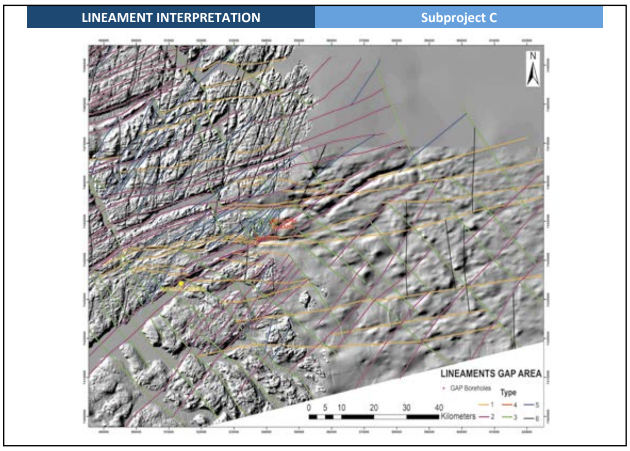

DOWNHOLE MEASUREMENTS
Temperature measurement Site 3
Site 3
Site 2
Site 1
Bedrock hole
Depth (m)
Temperature (C)


DOWNHOLE MEASUREMENTS
Water pressure measurement
Water Pressure: Hole GI10-1D
Fraction of overburden
Diurnal mode
Transition mode
Fall mode
seasonal evolution
Day-of-year 2010

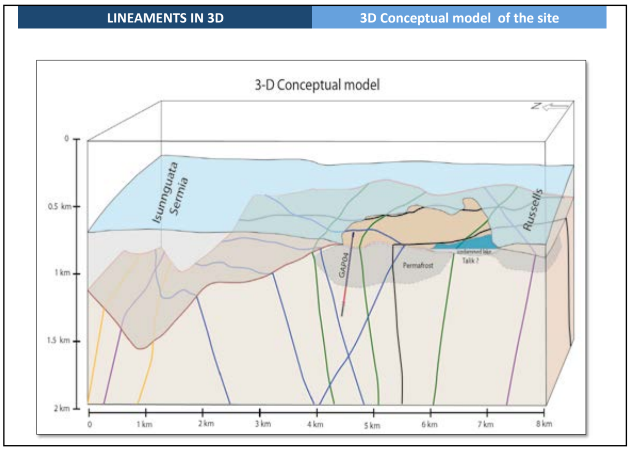

SUMMARY SUB A & SUB B
Ice sheet model from marginal area to 300 km
Cumulative runoff (% of total)
Elevation (km)
active drainage
wet
frozen
High hydraulic gradient & water
Low hydraulic gradient & water
strong melting power
weak melting power
Transient conduit reach
Distributed reach
up to 70% pressure drop in summer
<10% pressure drop in summer

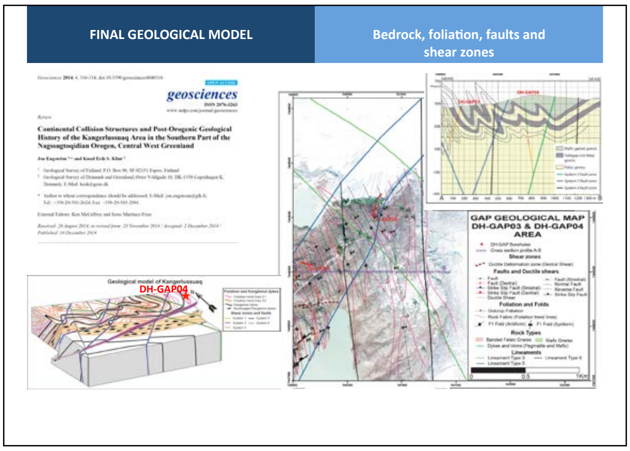

Project Description
Subproject C
Bedrock, hydrogeochemistry and hydrogeology
•Subglacial recharge of melt waters into the bedrock
•Pressure conditions
•Penetration depth
•Chemistry and oxic nature at repository depths
SPA
•Complementary field investigations
•Structural geology
•Distribution of permafrost
•Hydrogeochemistry of surface waters
SPB
0 meters
SPC
Subproject C: Bedrock hydrogeochemistry and hydrogeology
(subproject manager: Timo Ruskeeniemi)
700 meters
Bedrock
Permafrost
Ice Sheet

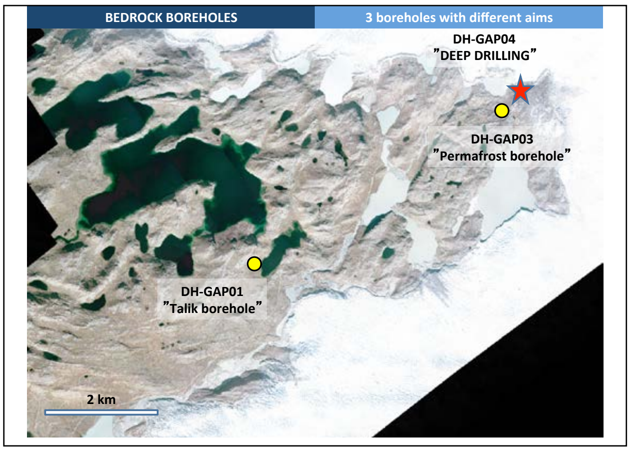

BEDROCK GEOLOGY
Archean gneisses and Kangamuit dykes
B Geological map of Kangerlussuaq area
Site area
Kilometers

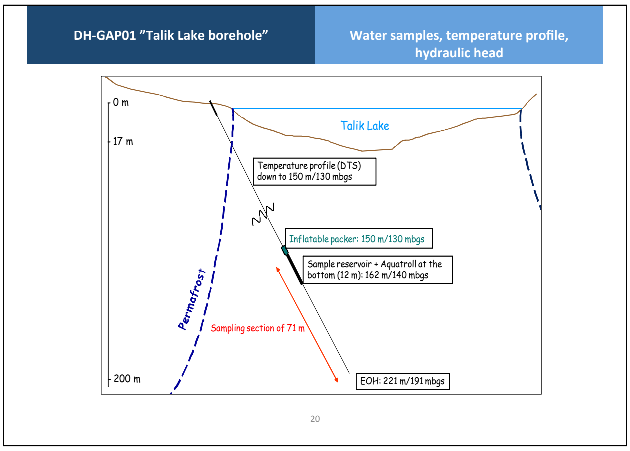

LINEAMENT INTERPRETATION
Subproject C
LINEAMENTS GAP AREA
Kilometers

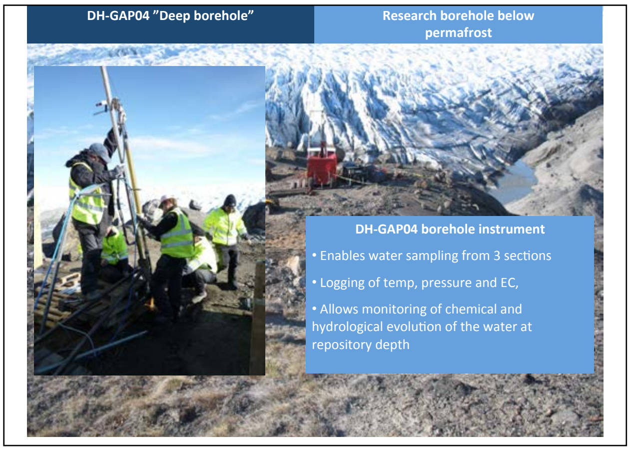
16. Engström, Lehtinen, Ruskeeniemi & Claesson Liljedahl: The Greenland analogue project – Project update.

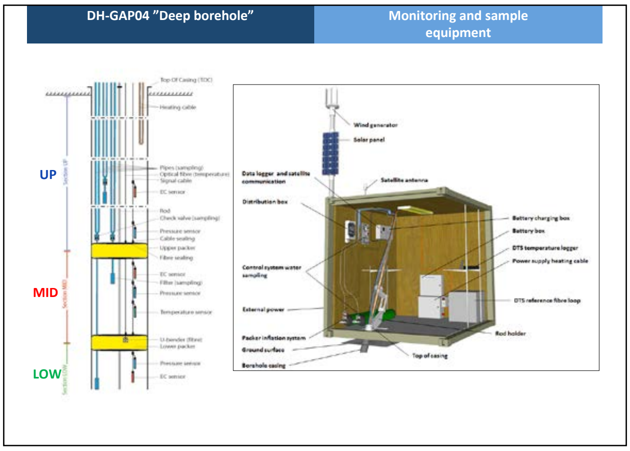


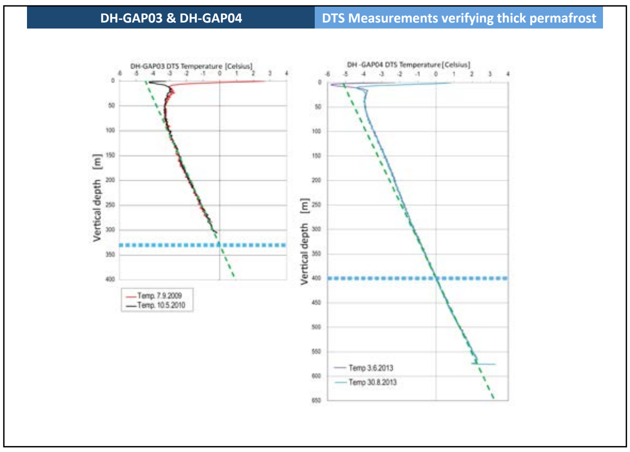


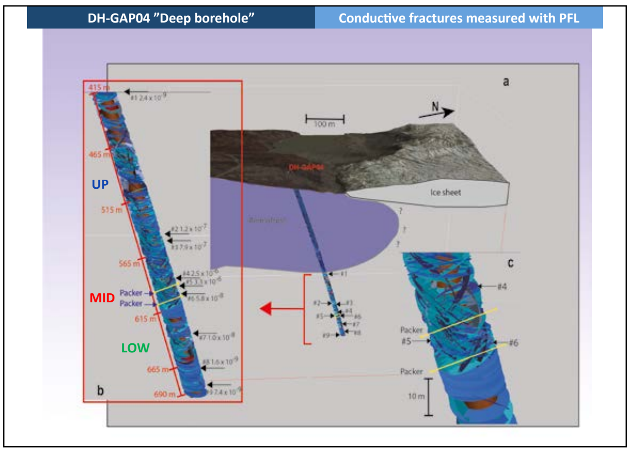






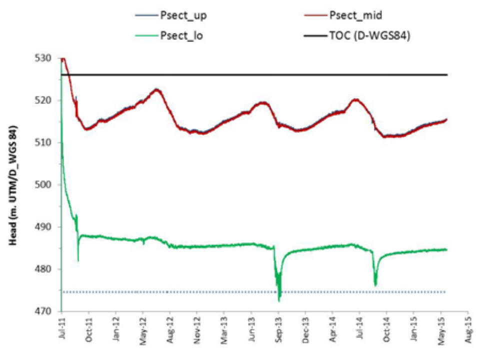






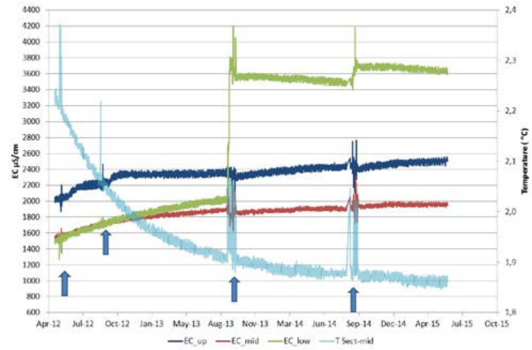
DH-GAP04 "Deep borehole" | Head measurements

Head measurement in upper middle and lower section compared with meltwater production from July 2011 to May 2015

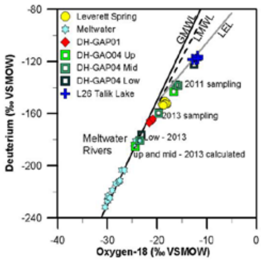


DH-GAP04 "Deep borehole" | EC and temp measurements

EC in upper, middle and lower section, and temperature measurement from April 2012 to May 2015



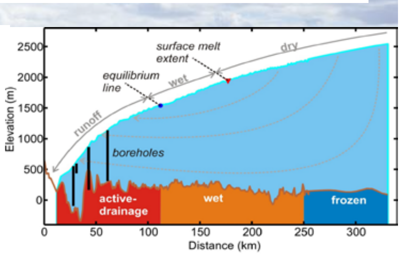
HYDROGEOCHEMISTRY | Hydrogeochemical characteristics



- Meltwater is very dilute, no specific chemical composition, light stable isotope signature is the characteristic feature
- Surface water have variable chemistries affected by local geology and evaporation etc.
- Groundwaters down to the monitoring depth are basically meltwaters (dilute: 180 mg/L Cl; Ca-Na-SO4-Cl type ; light stb isotope signature)
- Chemistry affected by dissolving fracture infilling gypsum ($CaSO_4$)
- Long residence times
- Extended glacial period in Greenland (16 Ma) probably strengthens the meltwater impact
- Hydrogeology is assumed to have a strong impact on the hydrogeochemistry
- Hydrogeochemical information only from 1-2 drillholes

Conclusions | GAP RESULTS

- Hydrologic conditions at the ice sheet bed were found to vary across. Between the ice divide and the margin, there is evidence for three different basal zones as defined by the amount and configuration of meltwater: ***the frozen zone, the wet bed zone, and the surface-drainage zone.***
- These zones result from surface, bed, and internal ice flow processes, and are likely representative of Northern Hemisphere ice sheets in a similar stage of development to the current GrIS.



Conclusions | GAP RESULTS

- The main part of the base of the ice sheet in the study area, including the marginal areas, has been shown to have basal melting conditions, with the exception of the central parts of the ice sheet.
- This, together with the glacial history of the region, suggests that permafrost does not exist under the major part of the large warm-based areas of the ice, with the exception of the ice margin, where a wedge of permafrost most likely extends underneath the ice for some distance.
- The existence of unfrozen areas through the entire permafrost thickness, so-called through taliks, in close proximity to a continental-scale ice sheet has been confirmed by scientific investigations conducted on the Talik lake and in borehole DH-GAP01.
- Several lakes in the investigation area are larger and deeper than the investigated lake, indicating that through taliks are common features in the hydrological system in the study area.

Conclusions | GAP RESULTS

- Below the permafrost (at depths greater than c. 350 m), reducing conditions are interpreted to prevail in the study area.
- Past penetration into the bedrock of dissolved oxygen in meltwaters has been limited in depth, as indicated by the presence of iron oxyhydroxides in fractures only in the upper parts of the rock (down to 60 m) with only two isolated occurrences of goethite down to 260 m depth.
- The stable water isotopic signatures (δ 2 H and δ 18 O) indicate that the groundwaters sampled in borehole DH-GAP04 (depth of at least 500m) are of glacial meltwater origin
- A preliminary interpretation of dissolved He concentrations from the upper section of DH-GAP04 suggests that the deep groundwaters may have residence times exceeding hundreds of thousands of years.

Conclusions | GAP RESULTS

Applicability to safety assessments for nuclear waste repositories

- Results provides important information on how the boundary conditions for groundwater flow modelling during glacial cycles should be defined, also in the case of the existence of deep permafrost at the ice sheet margin
- The evidence of anoxic groundwaters deep in the bedrock provides confidence in the quantification of geochemical parameters at depth, especially on the solubility of solids containing redox-sensitive radionuclides
- The evidence of the buffering capacity of the bedrock, not only for oxygen, but also pH, to maintain favourable chemical conditions at a repository at depth.

- **GAP Final reports published during autumn 2015**

The Greenland Analogue Project | Collaborators

POSIVA
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GEUS
GTK
University of Waterloo
UPPSALA UNIVERSITET
Stockholms Universitet
University of Wyoming
The University of Montana
PRIFYSGOL ABERYSTWYTH UNIVERSITY
The University of Edinburgh
InEarth
terrasolve
University of BRISTOL
UNIVERSITY OF CAMBRIDGE
Princeton University
NASA

16. Engström, Lehtinen, Ruskeeniemi & Claesson Liljedahl: The Greenland analogue project – Project update.

GRASP (Greenland Analouge Surface Project) Publications:
- Studies of the Talik Lake

Svensk Kärnbränslehantering AB

Johansson, E., Berglund, S., Lindborg, T., Petrone, J., van As, D., Gustafsson, L.-G., Näslund, J.-O., and Laudon, H.: Hydrological and meteorological investigations in a periglacial lake catchment near Kangerlussuaq, west Greenland – presentation of a new multi-parameter dataset, Earth Syst. Sci. Data Discuss., 7, 713-756, doi:10.5194/essdd-7-713-2014, 2014.

Data evaluation and numerical modeling of hydrological interactions between active layer, lake and talik in a permafrost catchment, Western Greenland
Johansson E. et al. 2015 Journal of Hydrology, Volume 527, August 2015, Pages 688–703.

17. Read: Industrial NORM and its relevance to nuclear waste disposal.

Industrial NORM & its Relevance to Nuclear Waste Disposal

David Read
Loughborough University

Rauma

June 2015

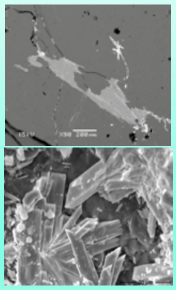
Outline

- Recap - Golden Age of U analogue studies
- Influence on PCSA
- Rationale for further N/A studies
- Natural laboratories: - re-visited (or what we should have done first time round)
- Behaviour of progeny – evidence from NORM
- Usefulness of other legacy sites

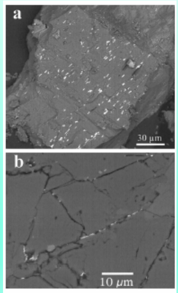
Uranium Fixation at N/A Sites

Poços de Caldas	Reduction & mineralisation
Koongarra	Secondary U(VI) mineralisation
Broubster	Organic complexation
Needle's Eye	Organic complexation, reduction, mineralisation
South Terras	Secondary U(VI) mineralisation
Steenkampskraal	Secondary U(VI) mineralisation
Palmottu	Secondary U(VI) mineralisation

Uranophane



Palmottu (Read et al., 2008)



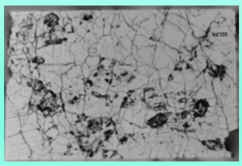
Hanford (Wang et al., 2004)

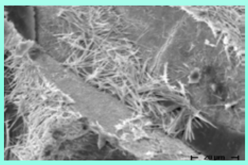
Mineralogy

- U occurs as essential constituent >160 minerals
  - Primary minerals (U(IV), Th)
  - Secondary minerals (U(VI))
- U (& Th) as major constituent (1-10%)
  - Monazite (LREE,Th,U)PO_4
  - Pyrochlore (Ca,Na)$_2Nb_2O_6$(OH,F)
- U as minor constituent (☒1%)
  - Apatite, rutile, baddeleyite, zircon etc.

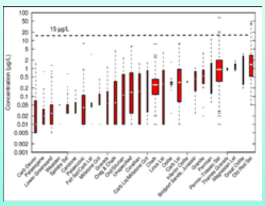
Natural U Fluxes

- U fluxes tend to be lower than theoretical estimates
- Re-concentration can produce secondary sources
- Significant inventory already present





Natural U concentrations in groundwaters



Smedley (2010)

	mol.dm^{-3}
$UO_{2(am)}$ solubility:	~$10^{-8.5}$
U ore deposits:	10^{-9} to 10^{-5}
Forsmark:	10^{-10} to $5x10^{-7}$
UK aquifers:	~10^{-10} to 10^{-7}
Sellafield BVG:	<$2x10^{-8}$
Sellafield SSG	<$3x10^{-8}$
Opalinus Clay:	~$2x10^{-9}$

- Natural U concentrations in some groundwaters exceed $UO_{2(am)}$ solubility
- U series nuclides already present in far field; those derived from GDF are incremental
- Disequilibrium of $^{234}U/^{238}U$ indicates that water-rock reaction occurs on short timescale, *i.e.* irreversible or kinetic processes other than reversible sorption

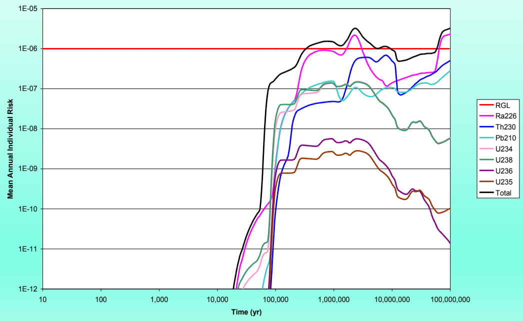
RWM's approach to far-field U transport

Generic DSSC (2010)

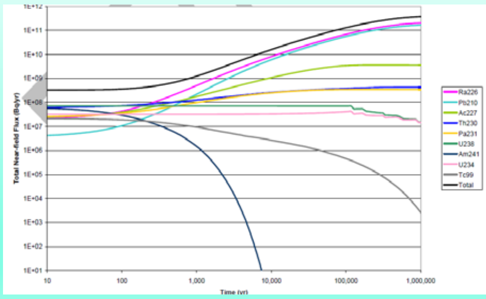
- Groundwater pathway modelled as q (specific discharge), T (advective travel time), F (mixing flux), A (discharge area), C (containment time) for sandstone far field
- K_ds for U(VI) & U(IV) are Nirex experimental data, used in GoldSim as log triangular PDFs for oxidising & reducing layers respectively
- K_ds for ^{230}Th, ^{226}Ra, ^{210}Pb are from various sources, applied in near-surface sandstone
- Initial condition: far field [rn] = 0

17. Read: Industrial NORM and its relevance to nuclear waste disposal.

Risk vs time for DNLEU in DSSC 2010



Flux near field to far field for DNLEU



Inventory Information - DNLEU

- Tails UF_6 to be converted to U_3O_8 post 2020
  - Ref case - compacted and cemented
- After conversion U_3O_8 will contain some UO_2F_2
  - Potential for container corrosion
- UO_3 from Thorp/Magnox rep. conversion to U_3O_8
- Unc. re composition/state of older uranic wastes
- Different chemical forms may be disposed of

Uranium Source Properties

	TDU	RDU	N	LE
Decay Products	X	X	√	X
Transuranics/FP	X	√	X	X
Accessories	(X)	(X)	√	(X)

Form - U_3O_8, UO_2, UO_3, metal, grouted, am/cr

Scenarios (U-IPT)

- Uranium disposed of in oxidised state and remains in this state during transport to the far field
- Uranium disposed of in oxidised state but reduced by interaction with components of EBS or host rock
- Uranium disposed of in reduced state and remains in this state during transport to the far field
- Uranium disposed of in reduced state but then oxidised by interaction with circulating groundwaters, components of the EBS or the host rock

Key Gaps

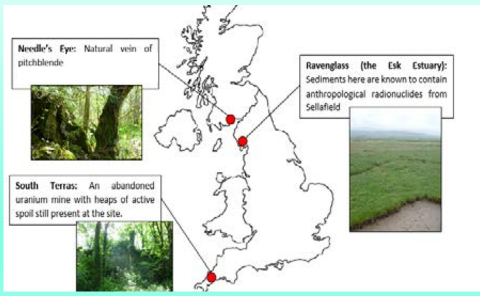
- Formation & stability of U phases in disposal settings
- Redox equilibria in alkaline conditions
- Dissolution rates as *f*(aq. chem, grain size etc.)
- Extent to which alteration products incorporate TRU & FP
- Factors that can be controlled (waste form, backfill etc) not yet specified ⌧ Increased uncertainty in processes & parameters
- Original N/A studies largely focussed on U

Estimation of Source Terms

Assuming adequate characterisation of source:

- theoretical/mechanistic methods
  - require detailed description of the inventory & t.d. data
- empirical methods
  - based on leach experiments
- analogue approaches
  - natural uraniferous deposits

Natural Laboratories: Re-visited



17. Read: Industrial NORM and its relevance to nuclear waste disposal.

Industrial Processing

"Nearly all materials contain trace amounts of natural radionuclides…. Where these materials are *processed*, *concentration* of these radionuclides may occur. The products or wastes from these processes are described as Naturally Occurring Radioactive Material, NORM." RASAG. *EA 2005*

NORM ≈ TENORM

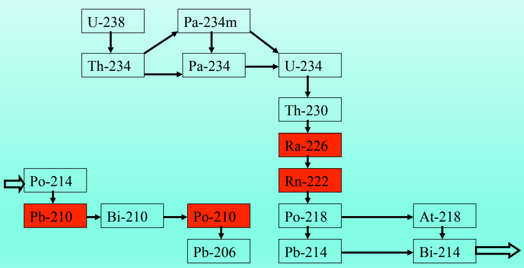
Radioisotopes in the environment owing to natural geological processes are not 'NORM', irrespective of their concentration

NORM Industries (EPR 2011)

Production of Th & Th compounds	Production of U & U compounds
Extraction, processing of REE & alloys	*Mining & processing of ores*
Oil and gas production	*Removal of scales & precipitates*
Utilising phosphate ore	*TiO_2 pigment production*
Extraction & refining of Zr & Zr compounds	*Production of Sn, Cu, Al, Zn, Pb, Fe & steel*
De-watering coal mines	*China Clay extraction*
Water treatment	*Remediation of Type 1 activity*

More generic than BSS but omissions *e.g.* cement, geothermal

The U-238 Decay Scheme

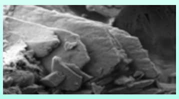


Waste Characteristics

- Scales, soils, building materials (e.g. concrete), sediment, dusts, sludge, effluent etc.
- Mineralogy dominated by:
  - Radium – Barite or bariocelestite ($(Ra)Ba\text{-}SrSO_4$)
  - Lead – Pb, PbO, amalgam ($HgPb_2$), PbS, $Pb_3(CO_3)_2(OH)_2$
  - U/Th – Silicates, phosphates (struvite)
- Co-contaminants – Heavy metals (Cr, Zn, Hg, Pb) As, C_{org}
- Often toxicological hazard > radiological
- Activities - exemption levels → ILW

Scales

$Ba\text{-}Sr(Ra)SO_4$
Carbonates
Fe oxides



Pb/PbO
Galena

Phosphates

Activities (Bq/g)

Source	^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po
Ore processing (non-metal)	10759	2400	7700	7400
Ore processing (metal)	>100000	8000	10000	10000
Oil & Gas (UK)	1200	260	11173	31769
Landfill (Exempt)	<5 (10) – HoC			
Landfill (SPB)	<200 – total activity			
LLWR (Drigg) limit	<4000 α <12000 β/γ - total			

Secular Disequilibrium

- Radium wastes
  - MOD sites, luminising works (^{226}Ra). ^{210}Pb by ingrowth
  - Oil & gas scales/slurry. (^{226}Ra & ^{228}Ra). Possible ^{210}Pb/ ^{210}Po excess
  - Mineral processing scales. As above + possible U

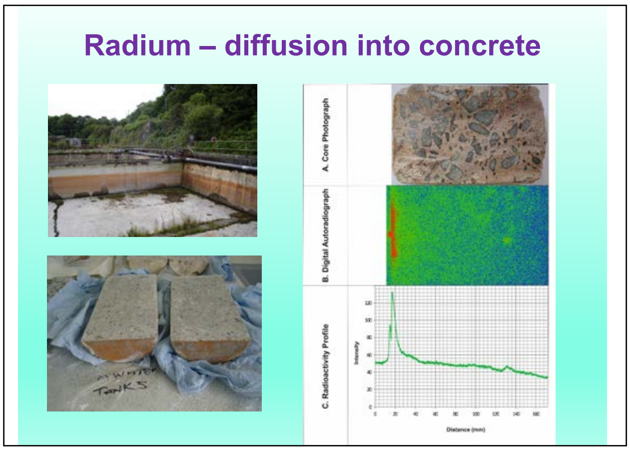
	Activity Concentration (Bq g⁻¹)					
	^{238}U	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th	^{228}Ra
MOD	<0.49	80.4	54.8	n.a.	n.a.	<0.34
CNS	0.98	42	95	110	n.a.	31
Mineral	<0.9	1906	2898	135.7	<1.2	676
SNS	<0.01	0.06	854	3973	<0.01	0.07

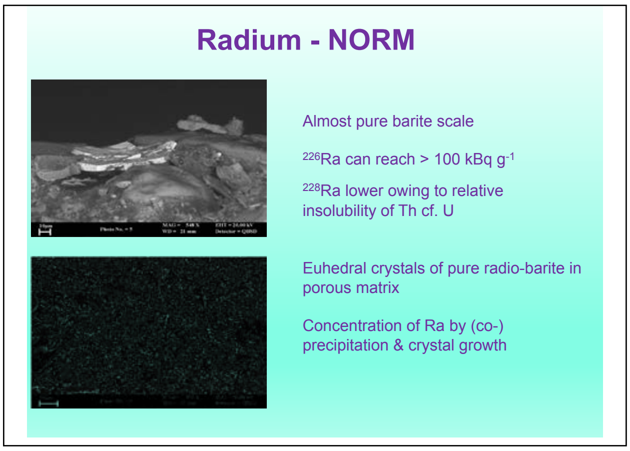
- Lead wastes
  - Oil & gas (SNS). Unsupported ^{210}Pb. Possible ^{210}Po excess
  - Iron & steel, cement, incinerators etc. As above

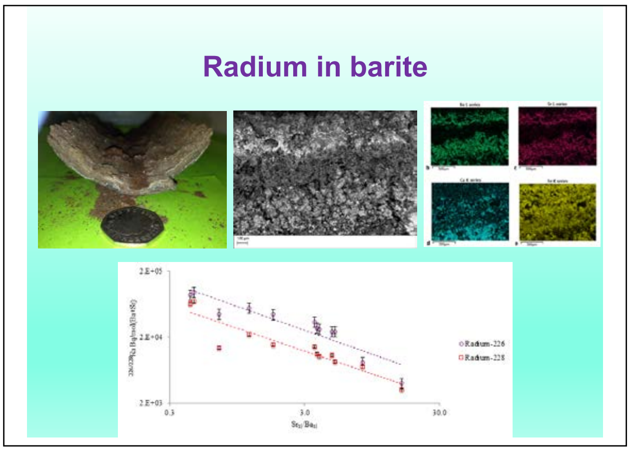
Fate of U/Th Series in NORM

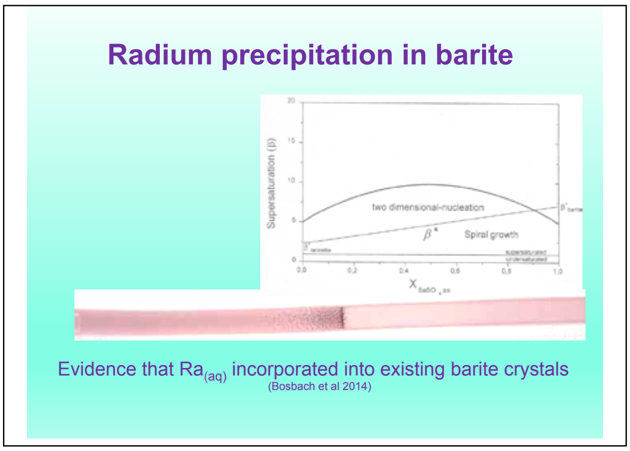
- Uranium
  - Mostly in products & discharges
  - Conc. e.g. in Fe deposits (tanks) & calciners
- Thorium
  - Rarely found, retained in source minerals
- Radium
  - Highly conc. in scales, dusts etc.
- Lead & Polonium
  - Conc. in driers, scrubbers, calciners, precipitators

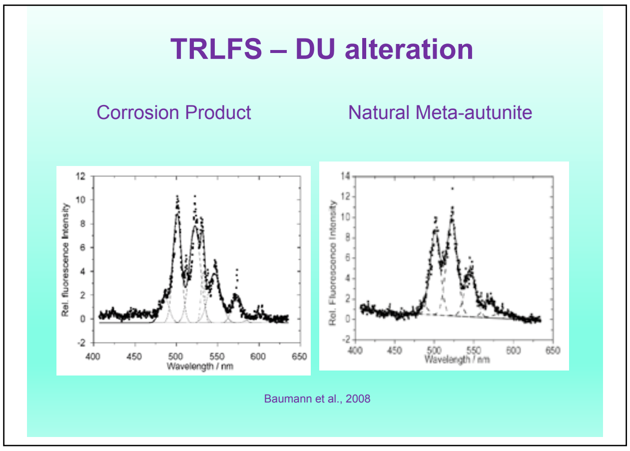
17. Read: Industrial NORM and its relevance to nuclear waste disposal.

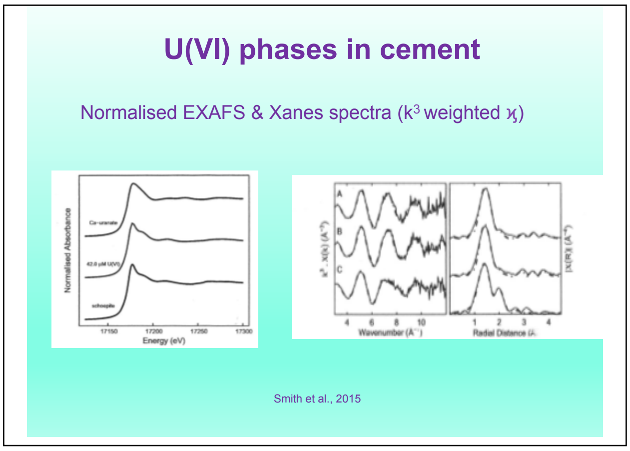


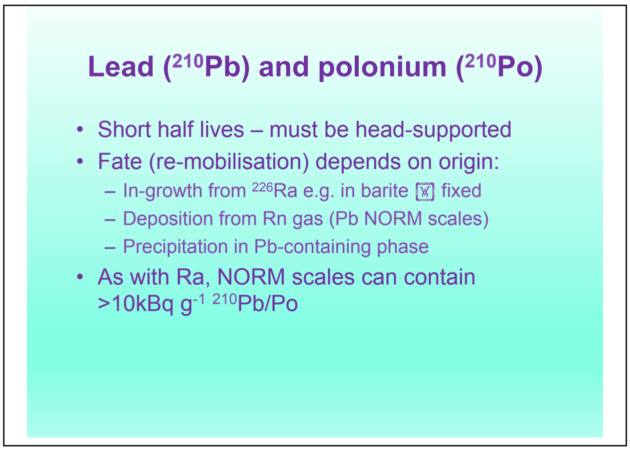


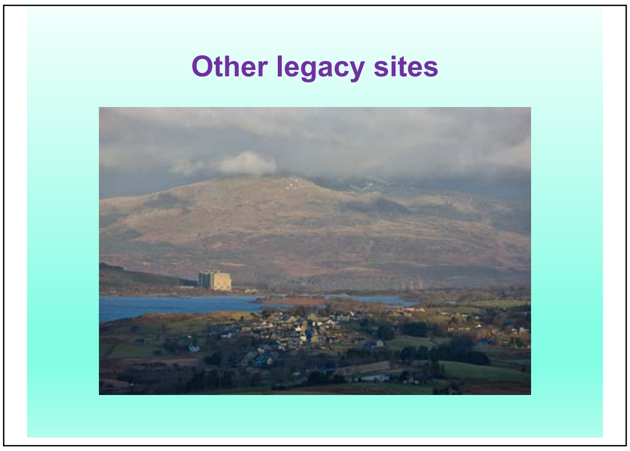








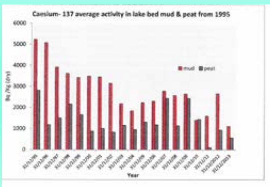




17. Read: Industrial NORM and its relevance to nuclear waste disposal.

Llyn Trawsfynydd

- Shallow lake (5 km^2) in North Wales
- Originally used for hydro-electric power
- Water used to cool NPP (1965 – 1994)
- Excellent record of discharges (^{3}H….^{241}Am)
- Data for range of isotopes in sediments etc.



Summary

- Previous NA studies focussed on U
- U phases not always identifiable
- Work on progeny limited (e.g. USD using Kd models)
- Opportunity to re-examine:
  - Old databases
  - Archived samples
- Some sites have changed – land use

Use of analogues of carbon to support disposal of spent HTR fuel

Fiona Neall, David Bennett
and Robert Winsley

Galson Sciences Ltd — TerraSalus Ltd — Radioactive Waste Management

A gallop through…

- Disposal of old experimental HTR fuel
- Potential problems with an ILW disposal route
- Developing a conceptual model for waste evolution
- Use of analogues to support the model

….and provide the solution (we hope)

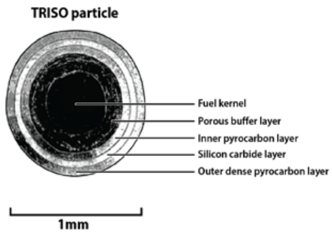
Dragon Reactor Project

- High-temperature, gas-cooled research reactor
- Ran 1965 – 1975
- OECD-funded project (→ 1000 reports)

The fuel – part I

Fissile & fertile material in kernels:
•U – mainly HEU
•Oxides & carbides
•C, Zr as diluents

Carbon and SiC coatings to retain fission products

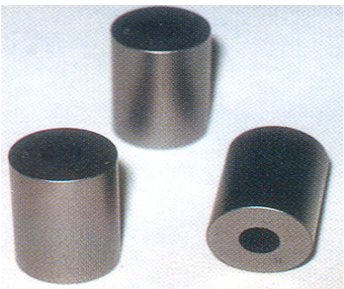


The fuel – part II

Particles moulded into compacts with graphite powder & resin binder

40 mm high
44 mm diameter

~10,000 particles per compact

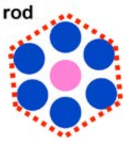


The fuel – part III

Compacts slid into graphite tubes:

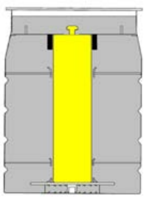
39 compacts per rod

6 driver rods per element + 1 experimental rod



Disposal concept

- Compacts unloaded into third-length containers (TLCs)
- ~250 TLCs
- One TLC per 500-l drum
- Grouted
- Disposal in ILW vaults (high pH environment in the post-closure period)



The problem

- Still a fair amount of fissile material in the fuel
- Criticality safety assessment → safe fissile mass (SFM) for single packages in post-closure period
- Complex burn-up histor… …out inventory in each waste package

→ Use the as-manufactured … …s in the CSA

Carbon in the fuel acts as a diluent and means a higher SFM can be assured

As long as it sticks around

? How do we show that the carbon will remain in situ ?

The (first) solution

- GoldSim model of the carbon evolution
- Elicit values for carbon loss rate
- Calculate how much carbon is lost in 1 My

And the answer is… ~~All of it in a short period of time~~

Do we believe the carbon would react on a 100 – 1000 year timescale?

→ Time for the analogues study

The (better) solution

- Define a conceptual model for carbon evolution over about 1 million years
- Assemble evidence to support the key arguments of the conceptual model

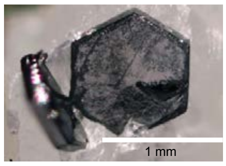
Conceptual model

- Carbon is basically unreactive (kinetics)
  - only corroded by oxygen (water is a catalyst)
  - usually requires high temperature (> 100°C)
- Limited oxygen in the post-closure GDF
- After oxygen is used up, radiolysis → oxidants
- Fission & activation products decay quickly (100s y)
- Alpha decay – low dose rate, no big supply of oxidants
- Products of carbon corrosion = mainly CO_2
- CO_2 in ILW vault (high pH, Ca) → $CaCO_3$ (calcite)
- Calcite still acts as a diluent

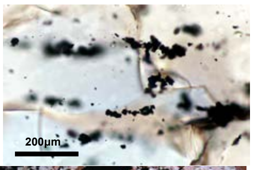
Evidence for carbon stability

- Kinetic constraints on carbon reaction – but how slow is the reaction with oxygen?
- Under geological conditions, carbon (graphite) is common and occurs in nearly all rock types:
  - common detrital mineral (clay fraction)
  - primary mineral in igneous rocks
  - vein graphites – commercial deposits
  - metamorphic rocks, including late-stage metasomatic alteration

© A. Jaszczak, Crestmore Quarries, Calif. US.

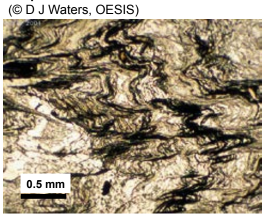


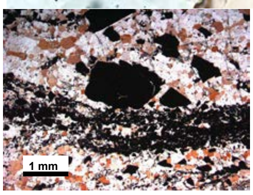
...in metamorphic rocks



Graphite in cherts of the Isua Supergroup, West Greenland (© Tokyo Institute of Technology)

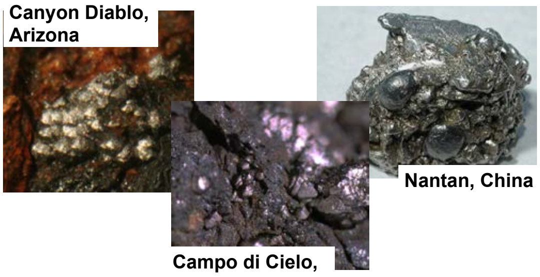
Graphite in schists (© D J Waters, OESIS)





Graphite in (Jurassic) schist, Fiddlers Flat, NZ (© U. Otago, NZ)

...in meteorites



Canyon Diablo, Arizona

Nantan, China

Campo di Cielo, Argentina

Industrial experience with graphite

Short timescales, but extreme (non-aqueous) conditions:

•Graphite is used in large quantities at very high temperatures in the steel and other industries

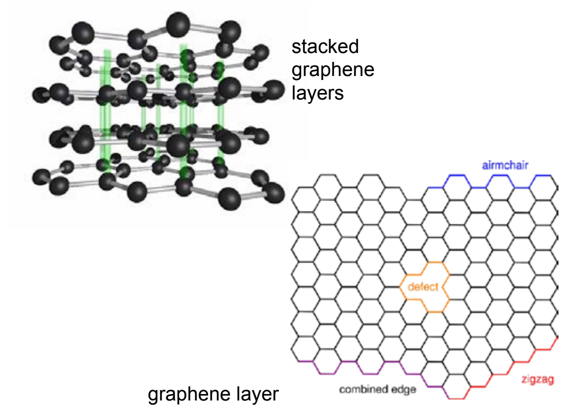
•T > 1800° C is required to start the carbon-carbon atomic diffusion process →graphitisation process

•In air, T >> 300° C is required before appreciable oxidation takes place and oxidation only becomes significant at temperatures around 400° C

•Air-cooled / graphite-moderated reactors: core operated in air at low T (100-200° C) operated for many years without significant thermal oxidation of the moderator

Use of graphite in catalysis

- Heterogeneous catalysis relies on the stability of the surface properties of graphite
- Known that oxygen from the air is sorbed on the graphite surfaces (surface oxidation)
- Depending on conditions, a variety of functional groups can occur at active sites
- Active sites at edges of graphene layers or associated with lattice defects
- Surface properties can be modified by use of different functional groups



Stability of graphite surface complexes

Experimental determination *in vacuo.*

Temperature range covers different carbon substrates

(Zacharia 2004)

Surface group	Product	Temperature range (°C)
Carboxylic acids	CO_2	180–300
Acid anhydrides	CO_2 and CO	400–650
Lactones	CO_2	137–650
Phenol	CO	600–700
Quinones	CO	800–900

Behaviour of graphite at high pH - I

Industrial experience

- **Experiments involving boiling graphite powder in conc. NaOH (2M) to reduce intercalated sulphuric acid demonstrate high degree of stability to high pH**
- **Use of carbon fibres, particulates and nanotubes in cementitious materials suggests expectation of minimal chemical interaction between high pH pore fluids and the carbon components**

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Behaviour of graphite at high pH - II

Natural occurrences

•Maqarin / Khushaym Matruck – fine-grained primary graphite persisted through thermal metamorphism and low T hydration and high pH alteration over some 100s ky

•Serpentinites – primary igneous and secondary graphite is observed (e.g California) to persist in ophiolite formations despite high pH, reducing conditions over My timescales during uplift and serpentinitsation.

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Summary of analogue evidence

- **Geological evidence suggests graphite stability under reducing conditions in the subsurface over My periods**
  - **No evidence to suggest microbial activity could significantly affect its stability under these conditions**
- **Geological and industrial evidence to support graphite stability under high pH conditions**
- **Understanding from catalysis science supports model of interaction of graphite with atmospheric oxygen → this could lead to some carbon loss until the available oxygen is used up**
- **Evidence from many sources supports model contention that CO_2 produced from surface reactions would react under high pH conditions → calcite**

How much carbon could be lost?

Use processes in the conceptual model to estimate loss

- **Initial oxygen sorbed on carbon surfaces: 0.15%**
- **Beta/gamma radiolysis: 2%**
- **Alpha radiolysis around failed particles: 7%**
  - **Assume all the particles fail over 1 My**
- **Over 1 My, all processes → < 10% carbon corroded**
- **< 10% loss if CO produced**
- **Expected that most of the corroded C would remain in close proximity as $CaCO_3$**

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AMPHOS 21

Potential long-term alteration of NSF and fate of minor radionuclides. Information from NNAA

Lara Duro, Mireia Grivé, Jordi Bruno

NAWG. Olkiluoto, June 2015

Background

- UK illustrative concept for demonstrating viability for GD of HLW and SF is based on the KBS-3V concept developed by SKB
- Radiological hazard from spent fuel will still be significant after long times (>100,000 years) have elapsed.
- How does a change of the stability of the UO_2 fuel matrix under reducing conditions decrease its capacity to contain relevant radionuclides?

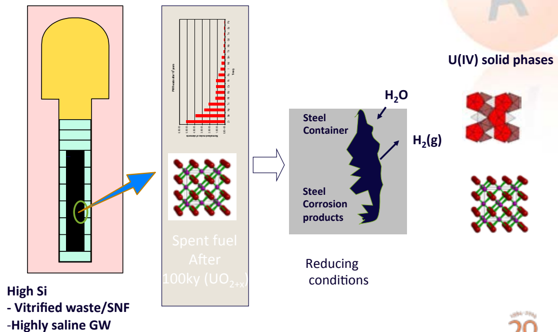
- Current plans for the UK Geological Disposal Facility (GDF)
  - vitrified HLW and ceramic SNF
  - intermediate level waste (ILW)

  potentially in the same installation (co-location)

  Different parameters affecting the behaviour of the waste

  - Si source
  - High alkalinity
  - Steel inserts in Cu canisters for SF

Possible evolution



Objective

- How able are secondary uranium phases formed under reducing conditions to retain minor components?
- Literature research of
  - Experimental laboratory data
    - Difficulty in keeping reducing conditions in laboratory experiments
    - U solubility under reducing conditions is low
    - Slow kinetics of the processes
    - Lack of quantitative data from laboratory experiments
  - Natural Analogues

In general:

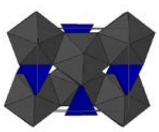
Main secondary U(IV) minerals:

- Secondary uraninites
- Coffinites
- Coexistence of both minerals observed

Coffinite. $USiO_4$

$USiO_4$ = ABO_4

SiO_4 tetrahedra and UO_8 dodecahedra



- It has been proven to retain
  - U, Th, Y, REE, Ca, Zr, Hf and Fe in the A-sites,
  - Si, P and S in the B-sites

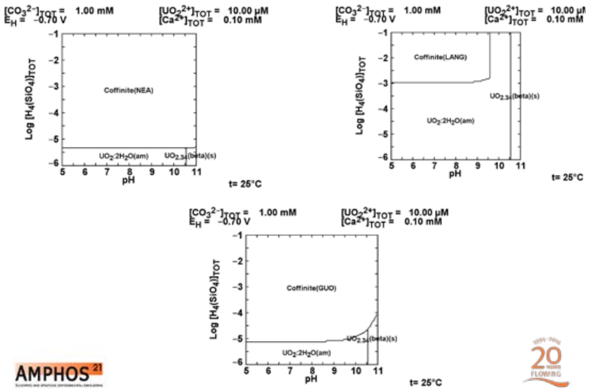
(Hansley and Fitzpatrick, 1989; Ewing, 1999,; Jensen et al., 2000; Jensen and Ewing, 2001),

---

Coffinite. $USiO_4$

- Composition and Stability not well resolved
- Grenthe et al., 1992; Guillaumont et al., 2003 (NEA selection): only derivation for crystalline $USiO_4$(cr)
- Langmuir (1978): estimation for $USiO_4$(am)
- Latest publication by Guo et al. (2015): DH_f by calorimetry

$USiO_4$ Stability



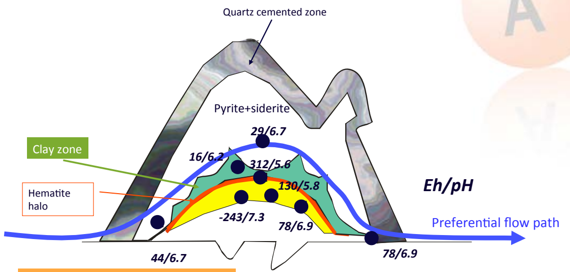
Laboratory studies on $USiO_4$ indicate:

- In laboratory, uranium silicate from UO_2 pellets observed at 180°C (Amme et al., 2005).
- A good review of synthesis of coffinite can be found in Deditius et al. (2008) and Pointeau et al., (2009).
- Recently Guo et al. (2015) have published new data on DH_f
- Preferent crystallization of coffinite over uraninite reported in the presence of sulphides
- No direct determination of coffinite structure. XRD and EDX: isostructural with thorite and zircon
- Difficulties in the synthesis due, among other reasons to:
  - Difficulty in maintaining reducing conditions
  - Very low solubility of coffinite
  - Metastability with regards UO_2+SiO_2 assemblages

Review of experimental data and natural analogue studies

- Cigar Lake (Canada). U deposit @ 450m depth.
- Oklo (Gabon). U natural reactors. Deep Okélobondo @ 350m depth.
- Palmottu (Finland). U mineralisation in granite down to 450m
- Tono (Japan). U deposit in sedimentary rocks @ 150 m depth
- Mina Fe (Spain). Schistose fractured rock @ca. 100 m depth
- Grants Region (USA). U-V ore in sandstones
- Liueryiqi (China). U deposit in granite @ 100-350 m depth

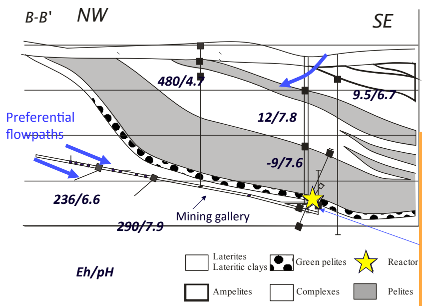
Cigar Lake, Canada



- Primary **uraninite & pitchblende** (1.3 Ga) Zr, Fe, Ca, REE, P
- **Coffinite** with Ca, REE, Al
- Pitchblende
- Coffinitisation observed

Janeczek and Ewing (1992): The replacement textures of the *uraninites* from Cigar Lake are indicative of *uraninite* dissolution and subsequent precipitation of clays, removal of Pb and coffinitization

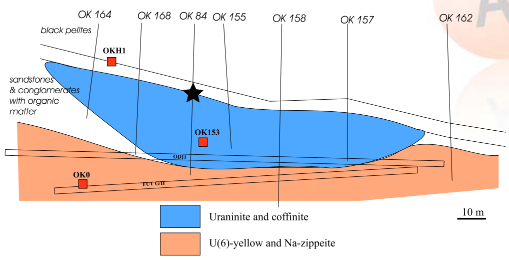
Oklo, Okélobondo-deep reactor, Gabon



Reactor zone:
Uraninite core covered by coffinite, containing: Pu, Th, Zr, Ru, Rh, Pd, Ba, Sr, Mo, Xe, Te, REE, Fe, Ti, Ca, Si, P

Coffinite and pitchblende in surrounding fractures, containing: Zr, Fe, Ti, Ca, Al REE,

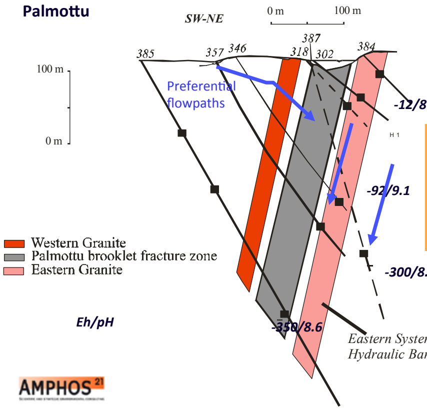
S-N



Formation of the silica-rich *uraninite* rims and partial coffinitization during and/or after the crystallisation of the phylosilicate matrix surrounding the massive reactor cores.

Extensive coffinitization also appears to have occurred with episodic Pb loss and *galena* formation in the reactor zones.

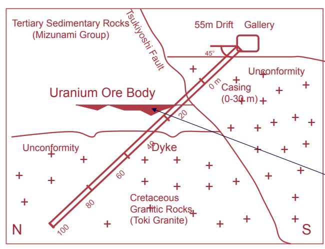
Palmottu



Oxidised zone: uranophane

Reducing zone:
- **Uraninite** containing Th, Low Y, Ca
- 2ry. Thin layer of **Coffinite** surrounding uraninite, containing: Zr, Fe, Ti, Ca, Al, REE,P

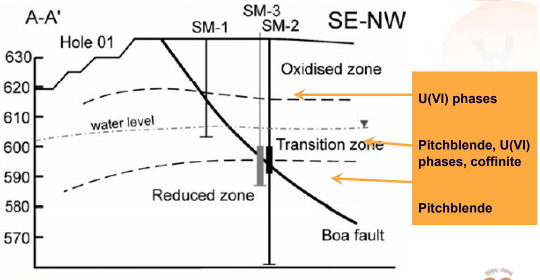
Tono, Japan.



Coffinite and pitchblende in the orebody

Oxidising groundwaters leached the U from the Toki Granite and transported it upwards into the lignite-bearing rocks, where it was precipitated or adsorbed (or both), under reducing conditions

Mina Fe, Spain

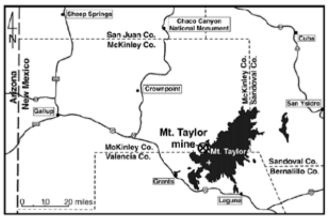


Highly fractured schistose rock

Grants Region, USA

- Uranium ore occurs mainly in *quartz-K-feldspar* sandstone
- Large crystals of ***coffinite*** (10mm)
- Rich OM environment:
  Warm fluids dissolved ore, reconcentrated when contacting OM, accelerated its maturation and dissolved silicates
- 3 coffinite generations
- Transformation of one coffinite into another took place under reducing conditions.

- $U^{4+} + Si^{4+} \Leftrightarrow U^{6+} + [2(OH)^{-}]$;
- $REE^{3+} + As^{5+} \Leftrightarrow U^{4+} + Si^{4+}$;
- $Ca^{2+} + (As,P)^{5+} \Leftrightarrow REE^{3+} + Si^{4+}$.



Liueryiqi granite (China)

- Main U-bearing minerals are *uraninite* and *coffinite*
- CO_2-rich fluids mobilised U from the sediments. During post-magmatic alteration of the host granites, U from the most refractory minerals was made more easily leachable by the hydrothermal solutions
- *coffinite* formed in a weakly alkaline reducing environment, with sulphide or selenide present in the fluids being the reductants for U

SITE	Primary minerals	Secondary U(IV) minerals
CIGAR LAKE	Uraninite, pitchblende, coffinite	Pitchblende, coffinite, uraninite recryst.
OKLO	Uraninite	Pitchblende, coffinite, uraninite recryst.
PALMOTTU	Uraninite	Coffinite
TONO	Pitchblende, coffinite	None, all secondary U(VI) minerals
MINA FE	Pitchblende	Coffinite
LIUERYIQI	Uraninite	Uraninite, Coffinite
GRANTS REGION	Coffinite	Different coffinite generation and secondary U(VI) phases

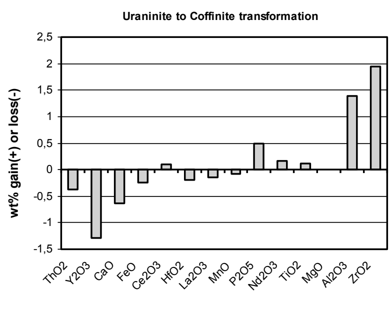
Some observations

- Coffinitization processes in nature reported at temperatures in the range 80°C to 300°C;
- It is normally associated with the presence of sulphides or organic matter (for example evidences from Cigar Lake, from Oklo and from the Liueryiqi deposit)
- Ages of some of the coffinitization events similar to the times of interest for the deep geological repository: the last coffinitization in Cigar Lake occurred ca. 0.8 Ma ago and in the Grants region 0.7 Ma ago;
- The maximum coffinitization thicknesses observed are in the range of 10mm;

Minor elements reported in coffinite

Coffinite		
Site	**Primary mineralisation**	**Secondary mineralisation**
Oklo	-	Zr, Fe, Ti, Ca, Al REE,P (esp. Bangombé)
Palmottu	-	Gain in Ce, Nd and Zr Loss of Th, Y, Hf and La
Cigar Lake	Ca, REE, Al	
Liueryiqi	-	High Ca Moderate Al Low Pb, Fe(II), K
Mina Fe	-	REE
Grants region	Ca, Y, REE, P, As	Ca, Y, REE, Zr, P, As

Transformation of Uraninite to Coffinite:



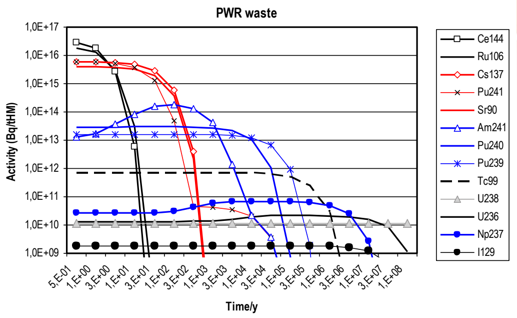
Data from Palmottu (Ruskeeniemi, 1998)

- So:

When SF is altered under reducing conditions may form secondary U(IV) solids:

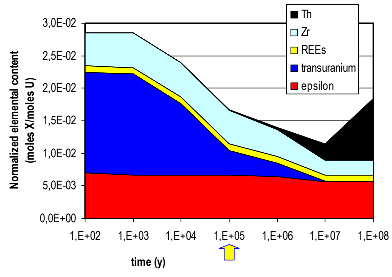
Can these solids host the minor radionuclides released when SF alters?

Evolution of PWR MOX 55 GWd/t



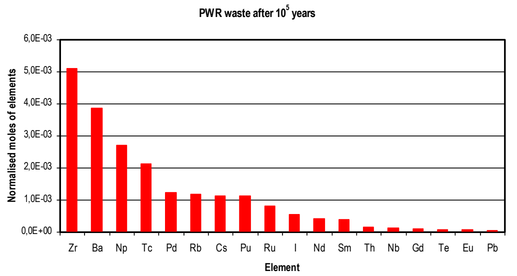
AMPHOS[21]

As elemental concentrations (not activities)



AMPHOS[21]

And as normalized concentrations at 10^5 years



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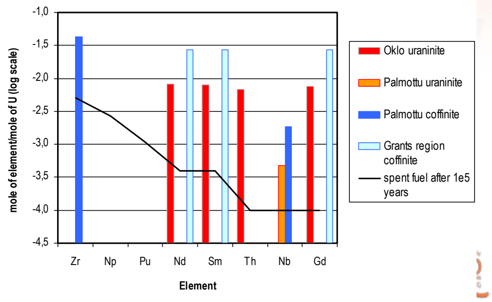
Element	Spent fuel after 10^5 years(1)	Uraninite: Oklo(2)	Uraninite: Palmottu(3)	Coffinite: Palmottu(3)	Coffinite: Grants region(4)
Zr	0.00510			*0.0426*	
Np	0.00270				
Pu	0.00110				
Nd	0.00040	0.0083			0.02733
Sm	0.00040	0.0079			0.02733
Th	0.00010	0.0068			
Nb	0.00010		0.00048	0.00185	
Gd	0.00010	0.0076			0.02733

AMPHOS[21]

Comparison of the molar fractions of different elements remaining in the fuel after 10^5 years and the observations from natural samples.



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- The results of the calculation of the evolution of the composition of the fuel with time indicate that, at the moment of the canister failure (considered to happen after 10^5 years):
  - the content in REEs, Th, Np, Pu and Zr is always lower than the content reported in secondary uraninites and coffinites,
  - Elements present in the fuel as metallic inclusions will not be so prone to be retained in these solid phases, but to form individual secondary solids, mainly sulphides, as evidenced by observations in nature

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So…

- Secondary U(IV) minerals can potentially host minor elements released from the fuel in adequate quantities under the expected conditions
- Many uncertainties: .....coffinite formation??
- More data on minor elements content in secondary mineralizations
- Plenty of natural coffinite samples exist: revisit samples and data

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  - NDA (UK) for partial financial support
  - Marcus Amme for technical discussions
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AMPHOS 21

Natural Analogue Study Ruprechtov – An Experience Report

R. Červinka, U. Noseck, V. Havlová, Th. Brasser, W. Brewitz, F. Woller

14[th] NAWG Workshop - 09.-11.June 2015, Rauma

Safety Case for a DGR

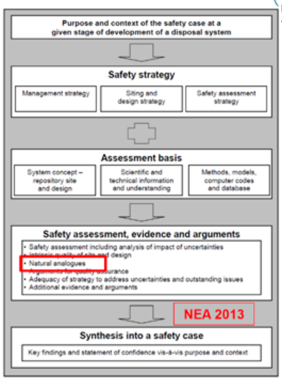
Compliance period of 1 million years in Czech Republic and Germany

- Remaining uncertainties from "short"-term lab experiments and field studies
- Uncertainties in predictive modelling

Comparison with processes in nature, may help in gaining greater confidence in

- data
- numerical models

Analogues as additional arguments for the assessment of repository safety



Analogue study Ruprechtov: Experience report

Natural analogue for

- Hydrogeochemical behaviour of uranium in clay materials / argillaceous sediments
- Sedimentary rocks / clay as a barrier for uranium migration

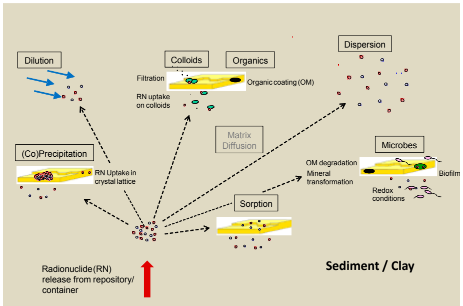
More than 15 years of co-operation

- CZ, D and several international project partners

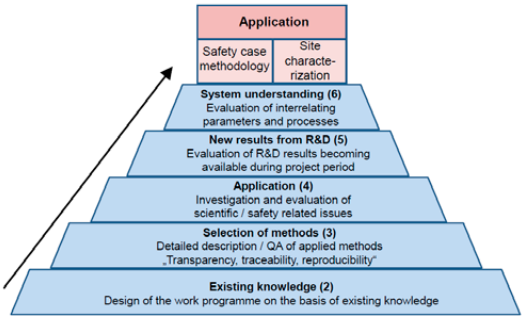
Main ideas for an Experience Report

- Compile / discuss decisions on selection of Ruprechtov site as a natural analogue
- classify the Ruprechtov site with regard to the type of uranium accumulation
- iterative steps, decisions and evolution of knowledge during site investigation
- experiences selection and application of experimental laboratory and field methods,
- outline the scheme by which these methods have contributed to understanding and characterizing the main features of the site,
- illustrate the main findings relevant for a Safety Case

Ruprechtov project: Processes of interest

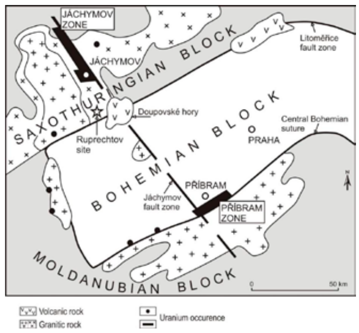


Ruprechtov Experience Report: Structure

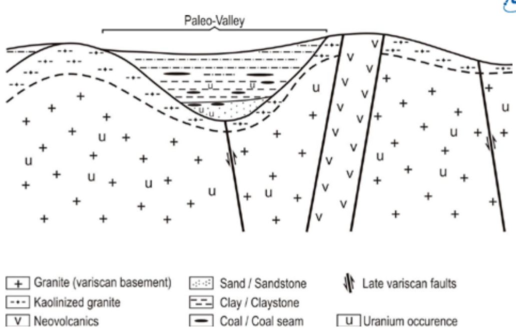


Ruprechtov site:

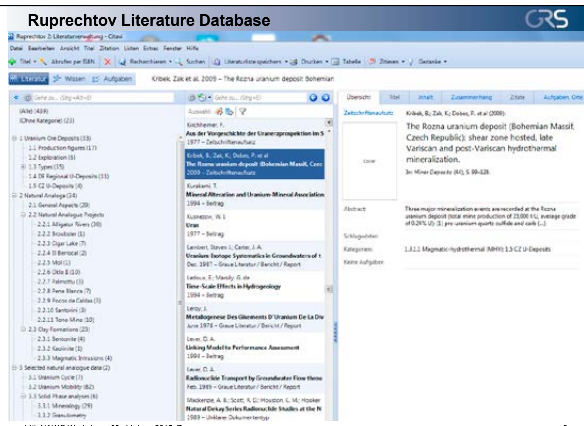
- Sediments representing characteristics of the overburden of potential host rocks
- Occurrence of organic rich material and uranium enrichment in the formation
- Occurrence and quality of bentonite
- Availability of geological information on uranium accumulation
- Occurrence of uranium mineralization within an app. 40 m depth (for practical purposes: no mining work required)
- Accessibility of the site in terms of ownership and mining regulations



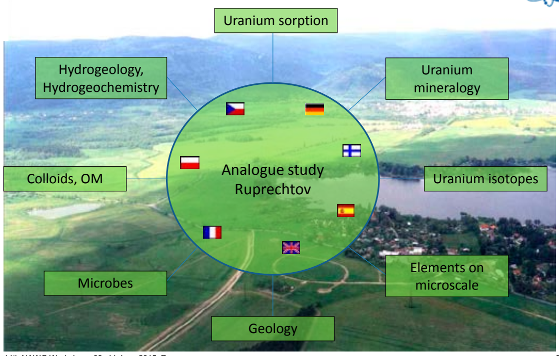
Ruprechtov site: Geological cross section



Ruprechtov Literature Database

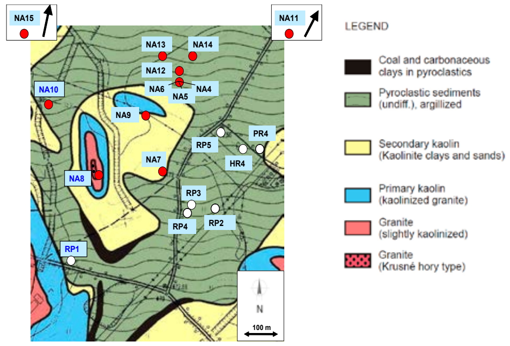


Ruprechtov: International Multidisciplinary Project



14th NAWG Workshop - 09.-11.June 2015, Rauma 9

Several Drilling Programmes

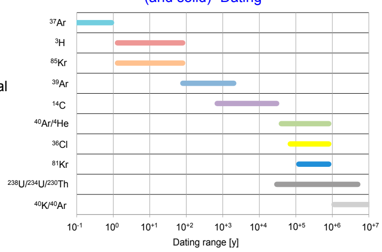


14th NAWG Workshop - 09.-11.June 2015, Rauma 10

Methods

- Drilling methods
- Drill core and GW-sampling methods
- Methods for characterizing geological parameters
- In-situ measurements
- Chemically based analytical methods
- Micro- to nanoscale analytical methods
- Isotope investigation methods
- Modelling tools

Example: Isotopes for Groundwater (and solid) -Dating



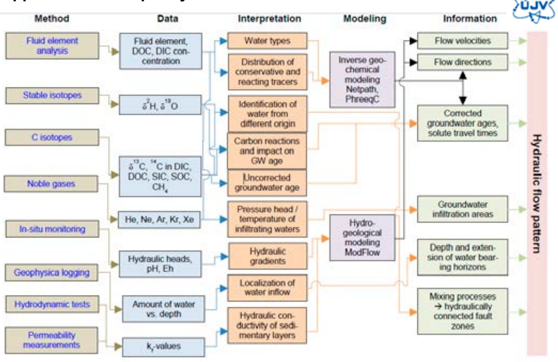
14th NAWG Workshop - 09.-11.June 2015, Rauma 11

Application

- Identification of (groundwater) flow patterns
- Geochemistry / geochemical milieu
- Microbiology
- Role of organic matter and carbon
- Uranium transport and uranium enrichment processes

14th NAWG Workshop - 09.-11.June 2015, Rauma 12

Application: Example Hydraulic Flow Pattern

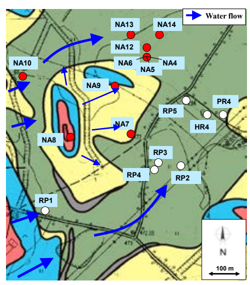


14th NAWG Workshop - 09.-11.June 2015, Rauma 13

Application

- Identification of (groundwater) flow patterns
- Geochemistry / geochemical milieu
- Microbiology
- Role of organic matter and carbon
- Uranium transport and uranium enrichment processes

Example: Local groundwater flow



14th NAWG Workshop - 09.-11.June 2015, Rauma 14

Short introduction into the report

14th NAWG Workshop - 09.-11.June 2015, Rauma 15

Relevance for the Safety Case

Uranium efficiently immobilised in a reducing environment

- No significant release of immobilised uranium over millions of years
- Long-term stable uranium bearing horizon (although near surface)

Identification of relevant features

- Impact of microbial activity on several processes
- Low impact of organic colloids on uranium mobility
- Strong isolation capacity of clays

Analytical methods applied and further developed

- Remote colloid sampling under undisturbed conditions,
- Modern analytics (μ-XRF, μ-XANES, U(4)/U(6) separation) for solid phase uranium characterization
- Combination of several isotope analyses helped to identify relevant processes in the field.

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Lessons learnt

Relevance of available knowledge from natural analogue studies for selecting sites with geological potential for the investigation of radionuclide retention / isolation

Development of a general strategy for field investigation
- Stepwise approach to maintain flexibility
- Thorough planning of further steps according to real needs and requirements

Application of drilling, sampling and site characterization methods and their optimization during the work progress
- Various technical experiences listed in formalized tables

Only possible by co-operation with international experts and working teams.
- knowledge and expertise in different disciplines (geology, mineralogy, hydrology,…)
- specific technical experience (e.g. isotope methods, state-of-the-art μ-/nano-analytics)

14th NAWG Workshop - 09.-11.June 2015, Rauma 17

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We really thank

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BMWi / PTKA (D)

for long-term support and interest in the project.

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PTKA Projektträger Karlsruhe

SÚRAO

Ruprechtov team and many others

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Thank you for your attention !

14th NAWG Workshop - 09.-11.June 2015, Rauma 19

Current natural analogue activities in Germany

Ulrich Noseck, Jens Wolf

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Current situation in Germany

Standortauswahlgesetz (site selection act), 23. July 2013

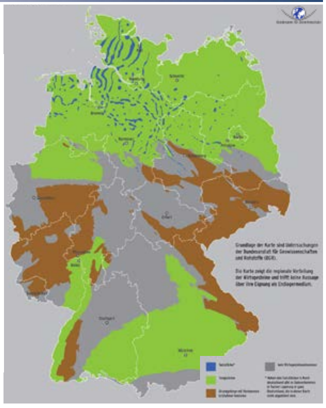
- Objective: Selection of a disposal site for the high-level waste produced in Germany within a scientific based and transparent process

Before start of the process 2 year work of a committee:

Committee: "Lagerung hochaktiver Abfallstoffe" ("Disposal of high level waste")

- Development of proposals for
  - Deep disposal or long-term interim storage
  - Exclusion criteria and minimum requirements
  - Involvement of the public / other stakeholders

Site selection process shall be completed in 2031



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Regulatory requirements for complementary considerations (analogues, BMU 2010)

"**Long-term statement on the integrity of the containment providing rock zone (CPRZ)** : For probable developments, evidence must be provided on the basis of a long-term geoscientific prognosis verifying that the integrity of the isolating rock zone is guaranteed throughout the reference period of 1 million years."

"For a numerical analysis of the final repository's long-term behaviour with respect to integrity of the isolating rock zone, radiological consequences, mobilisation of natural radionuclides, properties of containers and backfill, properties of the sealing structures, deterministic calculations should be based on the most realistic modelling possible ...
... Furthermore, where applicable, reference models (e.g. reference biospheres) shall be applied to periods with a high level of uncertainty regarding the input data and calculation models. Additionally, for such periods, **qualitative arguments** should also be consulted."

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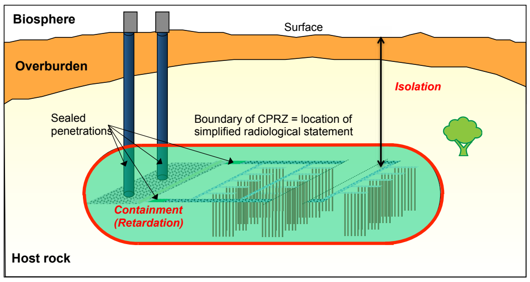
Concept of containment providing rock zone (CPRZ)



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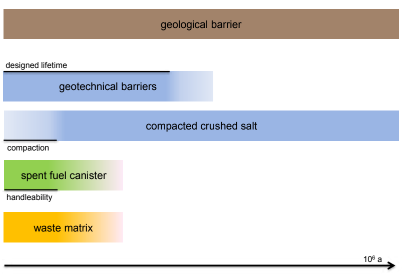
Types of analogues for the safety concept in salt



Safety function: containment

- Integrity of the geological barrier
- Integrity of geotechnical barriers

Safety function: retardation / containment

- Natural analogues for release scenarios

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Status at NAWG 13

Start of a systematic compilation and evaluation of how analogues can be used for the safety case of a HLW/SF repository in salt

Focus on the safety concept, i.e. the long-term safe containment by the assessment of the geological and geotechnical barriers

- Step 1: **Identification of NA**
  For which aspects can analogues contribute to the assessment of safety?
- Step 2: **Assessment of NA**
  What is the status of the identified aspects / analogues?
- Step 3: **Future Work**
  Identification of future work on aspects / analogues

Wolf & Noseck, 2015

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Example: Analogues for the integrity of the geological barrier

Aspect	Application	S	A	C
Existence of salt domes in Northern Germany	Long-term stability of salt domes	++	+	☼☼
		++	--	☼☼
		++	+	☼☼
		++	+	☼
		+	ø	☼☼
		++	++	☼
Chemical composition of fluid inclusions in salt formations	Interaction between salt formation and external solutions	++	+	☼
Chemical and isotope composition of gas inclusions in salt formations	Migration of gases in a salt dome	++	+	☼
Investigation of openings from salt mining	Behaviour of rock salt in the depth	ø	+	☼☼
Basalt intrusions in Fulda-Werra Series of	Sealing of fissures (Self sealing)	--	++	☼☼
		--	++	☼
		-	+	☼

++ Natural Analogon identified and well documented
\+ Natural Analogon identified but documentation insufficient
ø Natural Analogon identified but no documentation
\- Natural Analogon is not identified
-- Natural Analogon is (probably) not identifiable

☼ rather appropriate for communication with experts
☼☼ rather appropriate for communication with public

Step 1 → Step 2 →

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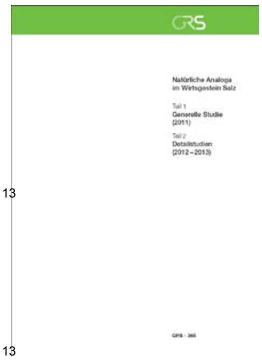
New report for NA in rock salt

Detailed reviews including
(i) state of knowledge of lab experiments and modelling and
(ii) potential for analogues
on

- Behaviour of Competent formations
- Compaction of crushed salt
- Composition of fluid inclusions
- Thermal stability of rock salt
- Mechanical stability of rock salt
- Impact of earthquakes on a rock salt formation / repository
- Qualified shaft and drift sealings
- Iron corrosion
- Microbial processes

NAWG 13 (Behaviour of Competent formations – Thermal stability of rock salt)
NAWG 13 (Microbial processes)



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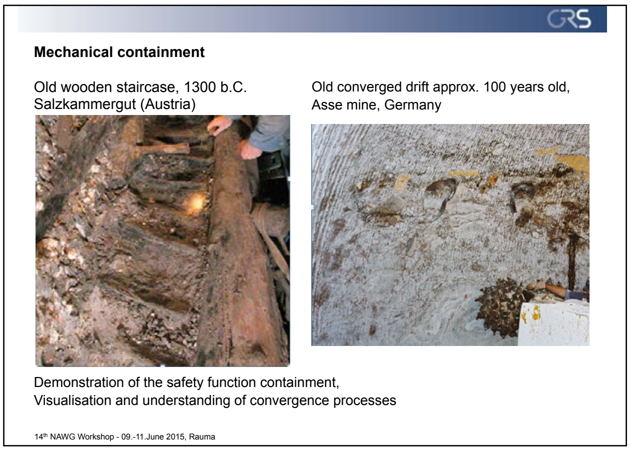
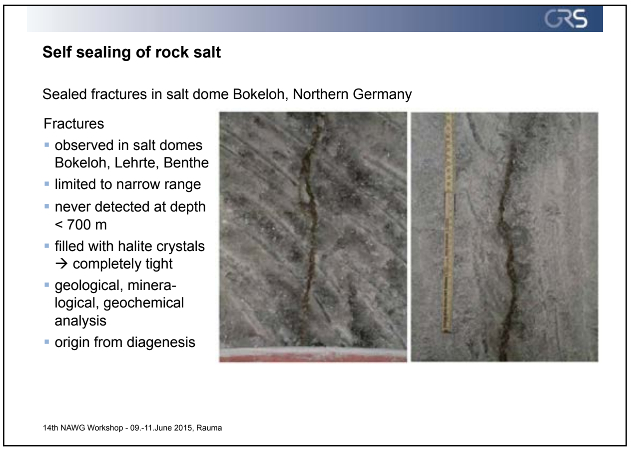
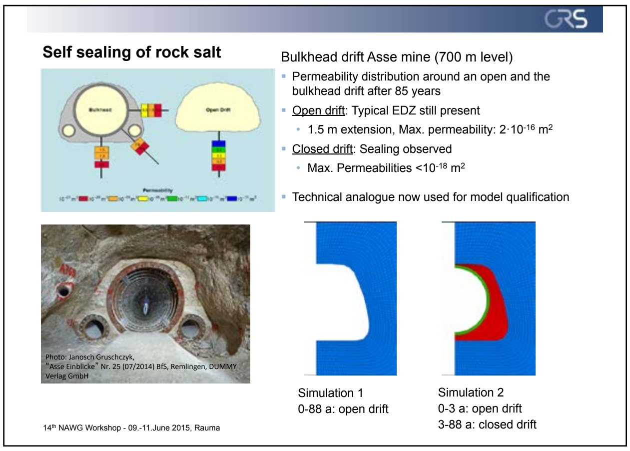
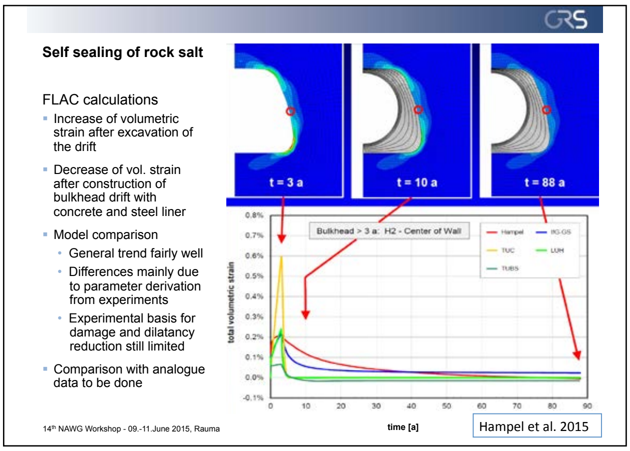
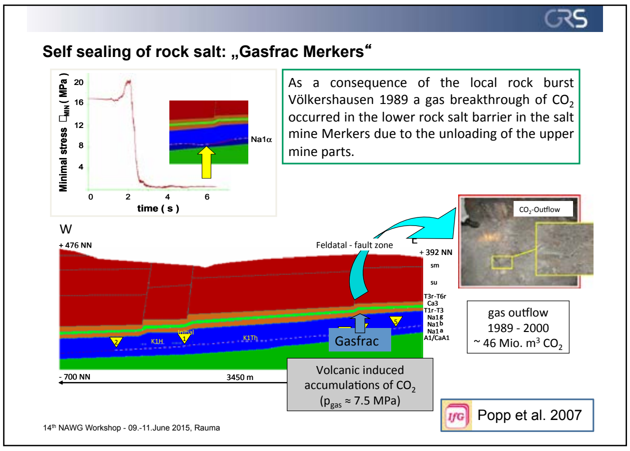
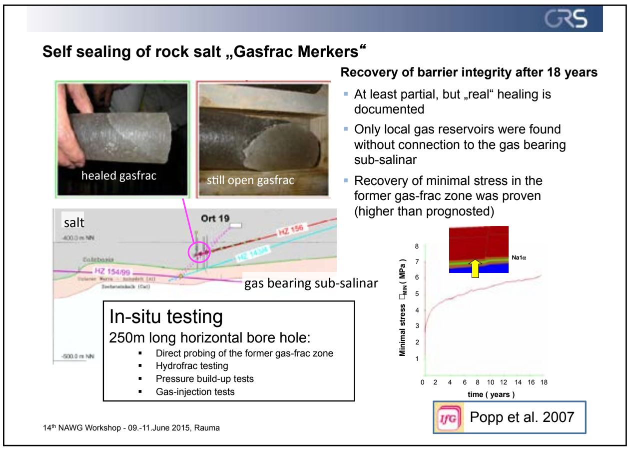


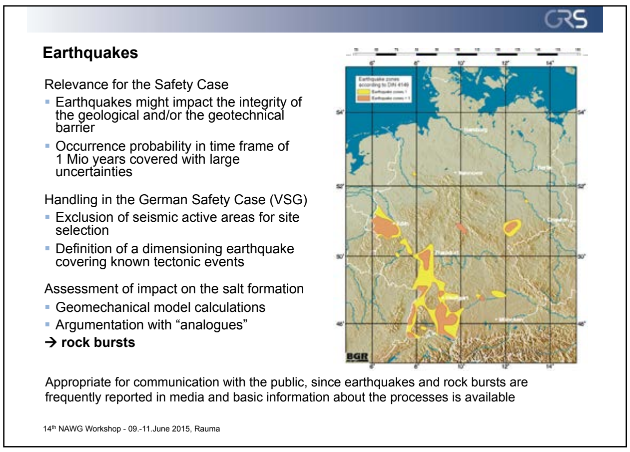


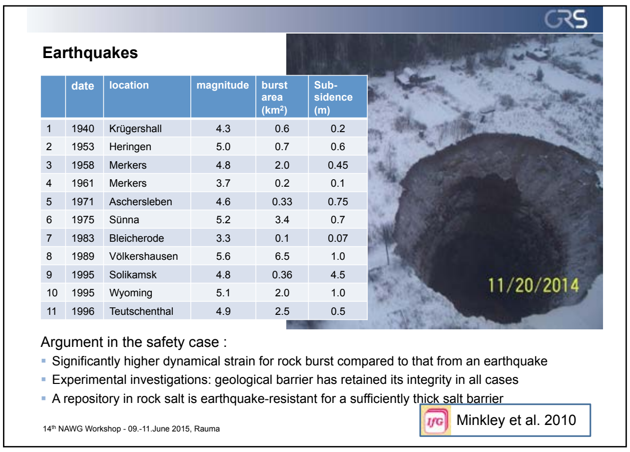




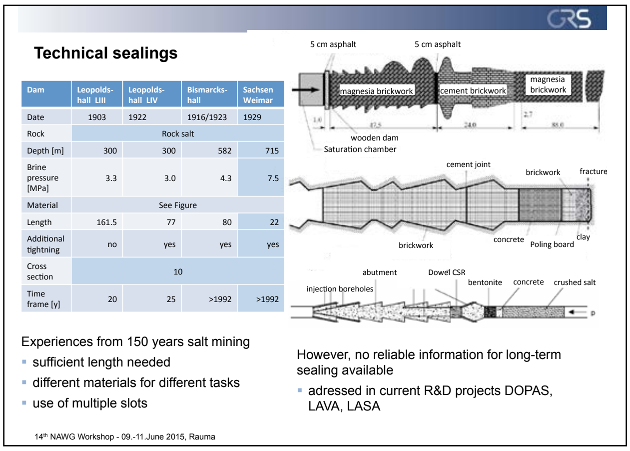




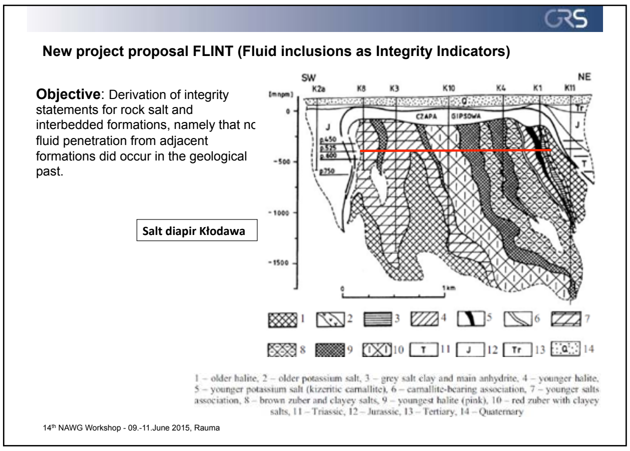


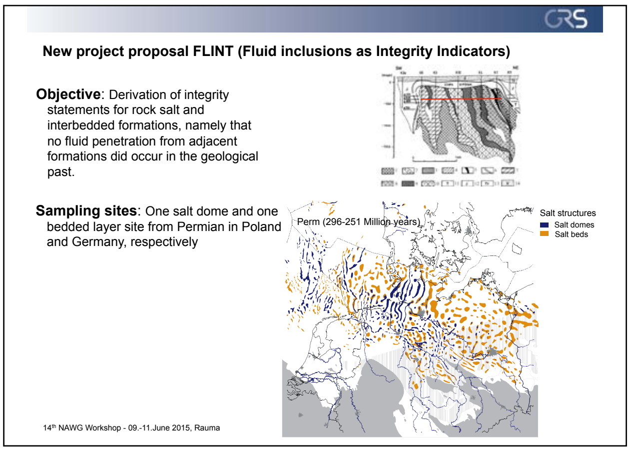


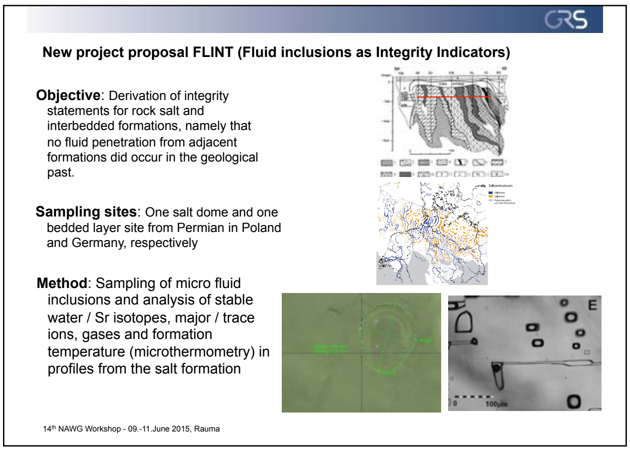
	date	location	magnitude	burst area (km²)	Sub-sidence (m)
1	1940	Krügershall	4.3	0.6	0.2
2	1953	Heringen	5.0	0.7	0.6
3	1958	Merkers	4.8	2.0	0.45
4	1961	Merkers	3.7	0.2	0.1
5	1971	Aschersleben	4.6	0.33	0.75
6	1975	Sünna	5.2	3.4	0.7
7	1983	Bleicherode	3.3	0.1	0.07
8	1989	Völkershausen	5.6	6.5	1.0
9	1995	Solikamsk	4.8	0.36	4.5
10	1995	Wyoming	5.1	2.0	1.0
11	1996	Teutschenthal	4.9	2.5	0.5

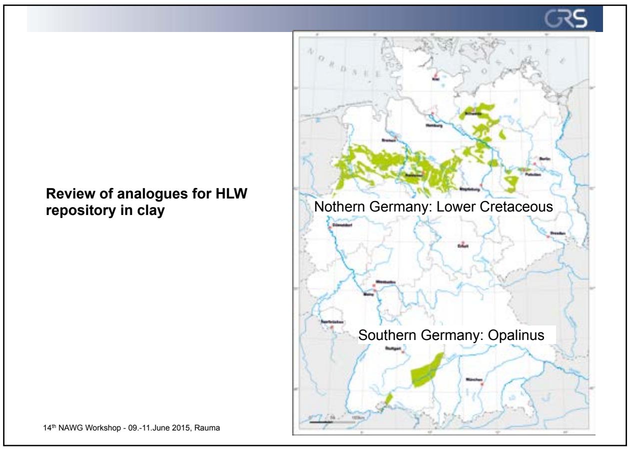


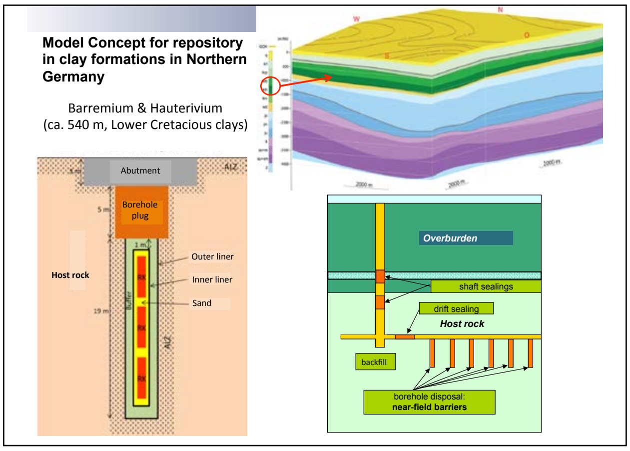
Dam	Leopolds-hall LIII	Leopolds-hall LIV	Bismarcks-hall	Sachsen Weimar
Date	1903	1922	1916/1923	1929
Rock	Rock salt			
Depth [m]	300	300	582	715
Brine pressure [MPa]	3.3	3.0	4.3	7.5
Material	See Figure			
Length	161.5	77	80	22
Additional tightning	no	yes	yes	yes
Cross section	10			
Time frame [y]	20	25	>1992	>1992

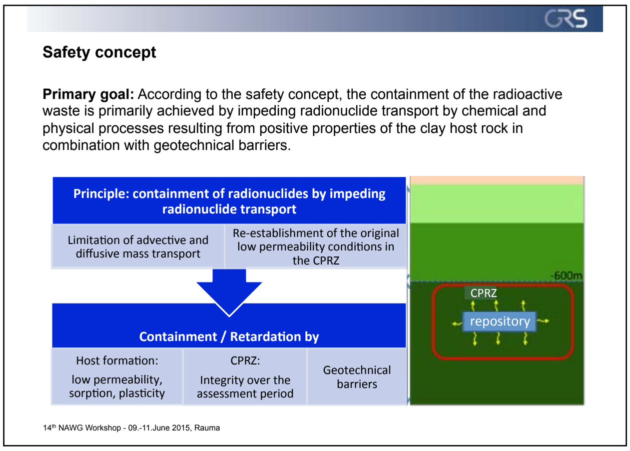


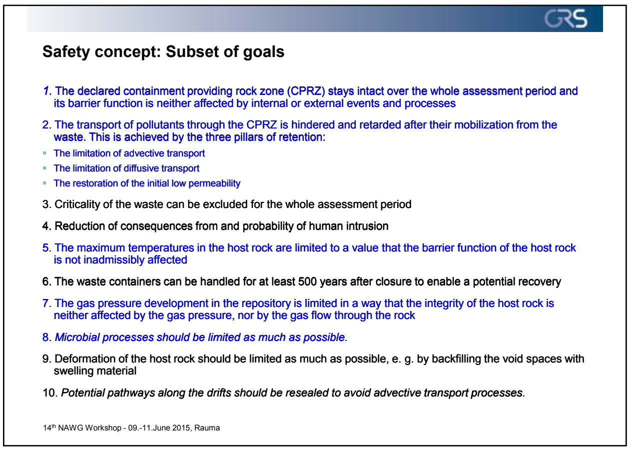












2. Goal: The transport of pollutants through the CPRZ is hindered and retarded after their mobilization from the waste

- **Limitation of advective transport**
- **Limitation of diffusive transport**
  - small diffusion coefficients
  - high sorption capacity
  - low solubility limits in the clay backfill and clay rock
- **Restoration of the initial low permeability of CPRZ**
  - self-healing of excavation disturbed parts of the host rock
  - in combination with the geotechnical barriers and backfill

Potential analogues

- **Transport of substances** in the pore water naturally occurred through geological past
  - Relevant processes can be deduced from tracer profiles in the rock pore water
- **Buffer capacity of the host rock** especially against pore water with cement phases
  - Clay formations in contact with cementitious pore water from natural geological environments
- **Resealing of fractures in the host rock**
  - natural analogues, processes that affected the host rock or comparable rock types
  - technical analogues like tunnel construction

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7. goal: Gas pressure in the repository is limited that the integrity of the host rock is neither affected by gas pressure nor by gas flow through the rock

Potential analogues: High gas pressures or gas flow might result in the fracturing of the host rock which potentially is a damage of the CPRZ integrity. Clay formations which are under a pressure load from gas can be found in natural and technical environments. Examples are natural gas resources in or covered by clay formations as well as technical gas storage in underground reservoirs. Both cases can be potentially suitable to serve as analogue for gas pressure effects to and gas transport processes in clay formations.

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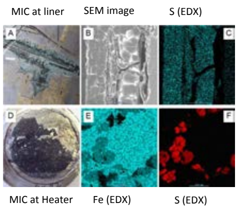
Goal: Microbial processes should be limited as much as possible.

Potential analogues: The microbial biota in a deep nuclear waste repository is expected to be represented through indigenous microbes that were inactive (or had very low metabolic activity) prior to construction and that become more active due to the emplacement of nutrients and electron donors. Additional microbe populations can be brought into the repository together with engineered materials.

- Anthropogenic analogues to the repository construction, e. g. from mining or tunnel construction or in-situ experiments
- Re-investigation of existing archaeological and natural analogues with respect to indications of microbial processes



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Complementary considerations for the safety case

Heini Reijonen, Nuria Marcos
Saanio & Riekkola Oy

and

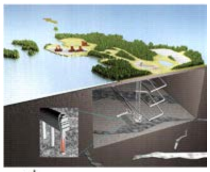
Marja Vuorio,
Posiva Oy

NAWG 2015 9.-11.6. Eurajoki, Finland

POSIVA

Contents

- Complementary considerations (CC) - background
- CC in TURVA-2012 safety case
- Future outlook for TURVA-2020 safety case



KBS-3V repository at Olkiluoto

POSIVA 2014-09-12 2

Complementary considerations - background

- Part of the safety case
- Regulatory requirements (STUK YVL D.5)
  - 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 with **natural analogues** or observations of the **geological history of the disposal site**.

POSIVA 2014-09-12 3

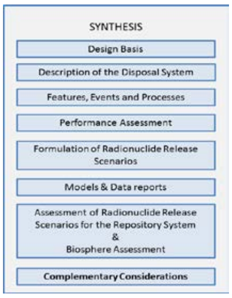
Complementary considerations - background

- "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."
- Complementary considerations have been described (NEA 2004, 2009) as evaluations, evidence and qualitative supporting arguments that lie outside the scope of the other reports of the quantitative safety assessment.

POSIVA 2014-09-12 4

Complementary Considerations in TURVA-2012 safety case

- Report supporting especially
  - FEP, '
  - Performance Assessment, and,
  - Formulation of Scenarios reports.



Neall, F., Alexander, R., Laine, H., Marcos, N., Hjerpe, T., Smith, G. & Vuorio, M. 2013. Safety case for the disposal of spent nuclear fuel at Olkiluoto -Complementary Considerations 2012 (POSIVA 2012-11)

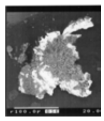
POSIVA 2014-09-12 5

NA related contents of CC report

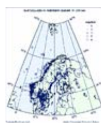
- Evidence for the long-term stability of the host rock, e.g.:
  - Understanding of the climate driven processes
  - Understanding of the site stability: e.g. historical earthquake record, low erosion rate
- Evidence for the suitability of the repository design and materials
  - Natural analogues for **long-term durability** of spent fuel, copper, iron, bentonite and cementitious materials,
  - Specifically reviewed against Olkiluoto site and design
- Evidence for limited rates of radionuclide migration in the repository system
  - e.g. NAs for diffusion-dominated transport in bentonite buffer
- Analogues for potential future conditions in the surface environment
  - Analogues used to understand biosphere conditions at the site during the dose assessment time frame (10 000a)
- The evolution of the repository system beyond one million years
  - NA used for the very long-term geological history of the site and its surroundings

Few examples:

Native copper and smectite in Finnish bedrock fracture at Hyrkkölä, Marcos (2002):



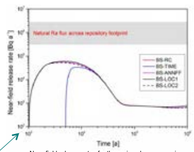
Earthquakes recorded in Northern Europe from 1375 to 1964 (University Of Helsinki):



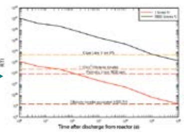
POSIVA 19/11/2015 6

In addition to NAs, CC includes also discussion on

- Geological disposal as waste management option
- Complementary Indicators (such as fluxes and concentrations of naturally-occurring radionuclides), introduced in CC → and used in the safety assessment (AoS).
  - In the period beyond a few thousand years *complementary safety indicators* using fluxes and concentrations of naturally-occurring radionuclides have been used (according to IAEA, 2003).
  - alternative indicators, such as "crossover times" have been used in illustrating safety (according to IAEA, 2003)



Near-field release rates for the various base scenario calculation cases, compared with range of natural Ra-226 flux through the repository area around repository depth



Radiotoxicity index of 1 tonne and 9000 tonnes of Finnish spent nuclear fuel

POSIVA 19/11/2015 7

Feed back from STUK on CC

- Overall CC has been found to be comprehensive
- A better integration of discussions in CC to safety functions and performance targets has been suggested.

POSIVA 2014-09-12 8

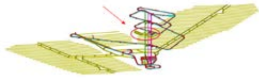
Future developments for the next TURVA-2020 safety case

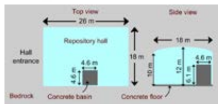
- Updates needed

POSIVA 2014-09-12 9

Updates related to design

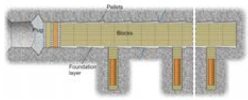
- Low and intermediate waste repository will be included in the safety case
  - Final materials and design
  - Location at ~-180 m depth





(Nummi et al. 2012)

- Deposition tunnel backfill design is to be updated before the operation licence application (around 2020)
  - No drastic changes expected i.e. swelling clay material



(Aution et al. 2013)

POSIVA 2014-09-12 10

New information on NAs

- CC report listed **NA studies overview on relevant to Posiva's safety case** (Appendix C in CC)
  → These have been brought forward already by publishing paper (supported by Posiva) on the **CC methodology and topics for further research** in Swiss Journal of Geosciences (see Reijonen et al. 2015)
- **New studies** also started and **new information** from e.g. Greenland Analogue Project (GAP), long-term cement studies (LCS) and Cyprus NA project (CNAP)
  - the final results will be available for TURVA-2020.
- Posiva is currently participating in several **NA projects**:
  - Finalising the GAP project
  - **Saimaa project** for searching information on stagnant ice sheet conditions
  - Olkiluoto **fracture montmorillonite** is being investigated in more detail
  - Olkiluoto "Merireikä" (sea borehole) project to investigate paleo gw chemistry

NA / site data

POSIVA 19/11/2015 11

Refined scope

- **Increased comprehensiveness and in-depth discussion**
- **Better integration to safety functions and performance targets**
  - **Will be obtained also by better cross referencing between safety case reports**
- **Considering also for the next CC report:**
  - **Comparison** with activity releases and concentrations in the repository system
    - Activity concentrations
    - Activity fluxes in groundwater
    - C-14 uptake by biomass
  - Comparison of radiation dose from different sources (Chapter 7 of POSIVA 2012-10)
    - Radon in indoor air
    - Natural radioactivity in the body
    - External radiation from the ground
    - Cosmic radiation
    - Medical X-ray exams
    - Medical radioisotope exams
    - Nuclear weapons tests and Chernobyl fallout

POSIVA 19/11/2015 12

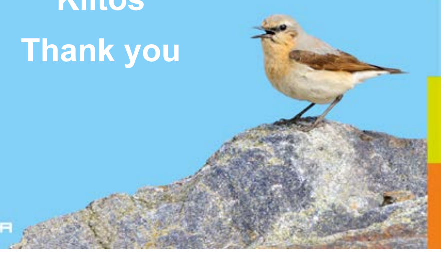
Conclusions

- The new CC will account for the requirements and additional feedback from STUK
- Updates will include
  - Design updates
  - New information from recent research and from the open literature
  - Some new topics
- Better integration with the other reports of the safety case
  - Better connection between the CC and safety functions and performance targets
- In-depth discussion → support for the overall conceptual understanding of the safety case

POSIVA 19/11/2015 13

Kiitos

Thank you



POSIVA

23. Norris, Milodowski, Alexander, West & McEvay: Using natural analogues to build stakeholder confidence in geological disposal "A catalogue of natural analogues for radioactive waste management".

Radioactive Waste Management

Using natural analogues to build stakeholder confidence in geological disposal – "A catalogue of natural analogues for radioactive waste management"

Simon Norris, Tony Milodowski, Russell Alexander, Julie West & Fiona McEvoy

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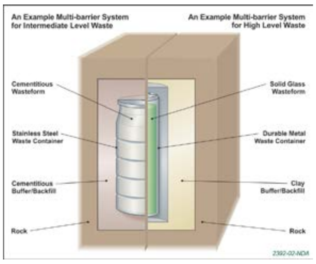
UK Geological Disposal Facility

- Site yet to be identified
- Work currently generic
- Multi-barrier system

Possible host rocks:

- Higher strength rocks
- Lower strength rocks
- Evaporites



Radioactive Waste Management 3

RWM's "Geological Disposal Environmental Safety Case Strategy" - 1

RWM report NDA/RWM/090 2012

"Analogues can be helpful in demonstrating understanding of aspects of GDF performance and provide evidence that certain materials can survive for long periods.

However, they do not provide conclusive proof that these materials will survive for the required periods in the environments of a particular GDF, as the conditions under which the analogue material has survived may not match those expected to occur or evolve in a GDF.

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RWM's "Geological Disposal Environmental Safety Case Strategy" - 2

RWM report NDA/RWM/090 2012

Therefore, analogues will be used with caution, and can only ever provide supporting arguments in an Environmental Safety Case.

Nevertheless, appropriate analogues can be helpful in providing a long-term practical demonstration to support the theoretical and mathematical safety arguments."

Radioactive Waste Management 5

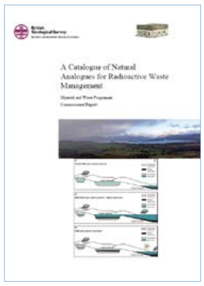
Catalogue of natural analogues for radioactive waste disposal

Aim

- •To provide a reference source of safety-relevant examples from natural systems that could be used to support a safety case
- •Set of "flyers" summarising key points

Scope

- •Target audience: Aimed at regulators, policy makers, managers, decision makers and others involved in the waste management process who are not necessarily specialists in natural systems
- •Suitable for the general scientific audience
- •All aspects of disposal relevant to UK situation
- •HLW, ILW, range of repository rocks and concepts
- •"Classical" analogue projects
- •Natural system data



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Objectives and scope of the natural analogues catalogue

To produce examples of data from natural systems (including archaeological analogues) that could be used to support a safety case for a GDF

For each analogue example, the following information is provided:

- A description of the specific aspects of components of the GDF that the natural or archaeological analogue system represents or is applicable to
- A summary description of the geoscientific characteristics of the natural analogue
- Key safety-relevant observations
- A summary of limitations of the use of the analogue
- Images of systems that can be used in RWM publications, subject to appropriate acknowledgements and copyright criteria
- A list of references where further information can be obtained

Radioactive Waste Management 7

Catalogue organisation

The examples grouped into 4 main sections, which are then subdivided according to their main safety barrier functions:

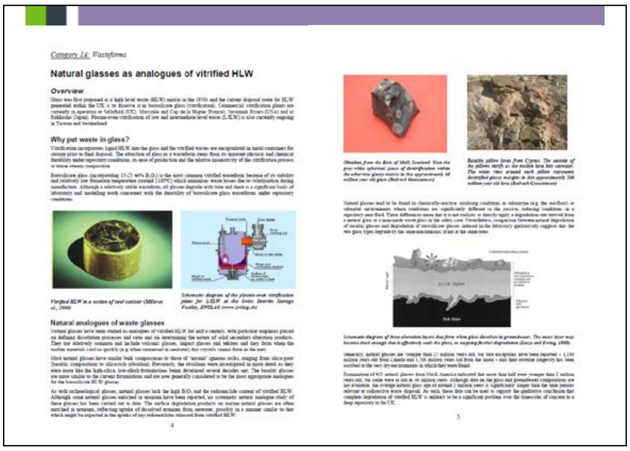
1. The Engineered Barrier System
   - Wasteforms e.g. natural glasses as analogues of vitrified HLW
   - Container performance e.g. Roman Legionary nails from Inchtuthil
   - Near-field barrier materials e.g. Cements from Hadrian's Wall, England
2. The Natural Barrier System
   - Long-term performance of clay host rocks e.g. The Dunarobba Forest, Todi, Italy
   - Long-term performance of fractured crystalline host rocks e.g. Tono, Japan (rock stability)
   - Long-term performance of salt host rocks e.g. natural brine inclusions in salt diapirs, Germany
   - Long-term isolation concepts e.g. Matrix Diffusion (e.g. Kamaishi, Opalinus Clay)
3. Radionuclide migration in natural systems
   - Retardation in natural systems e.g. Loch Lomond (iodine and chloride migration)
   - Colloid migration in natural systems e.g. Poços de Caldas, Morro do Ferro, Brazil
4. Natural analogues and the Safety Case
   - Generic safety case
   - Database and model testing e.g. thermodynamic databases examples from Oman and Jordan
   - Whole system performance e.g. Cigar Lake, Canada – a natural analogue for an entire repository?

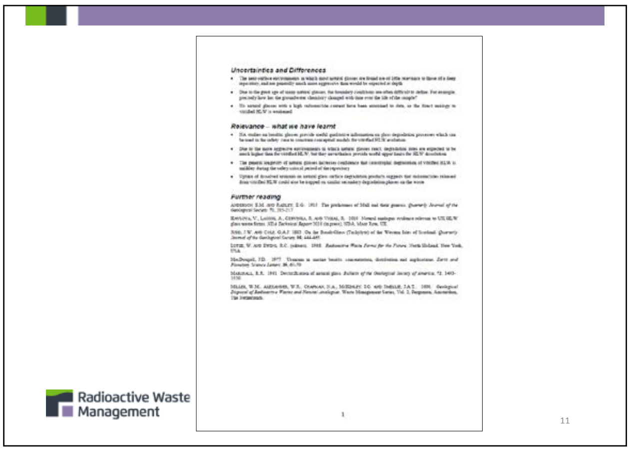
Radioactive Waste Management 8

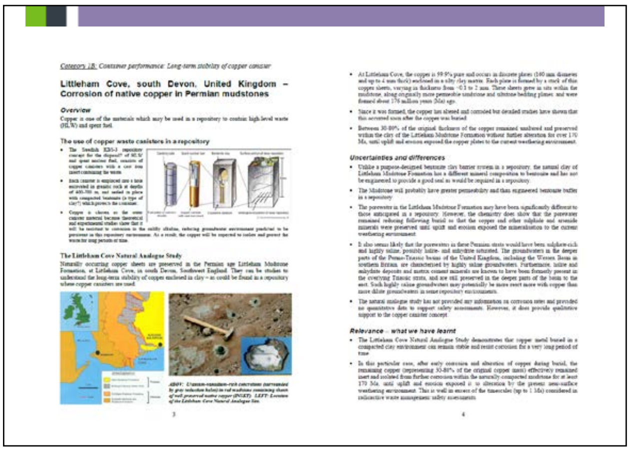
23. Norris, Milodowski, Alexander, West & McEvay: Using natural analogues to build stakeholder confidence in geological disposal "A catalogue of natural analogues for radioactive waste management".

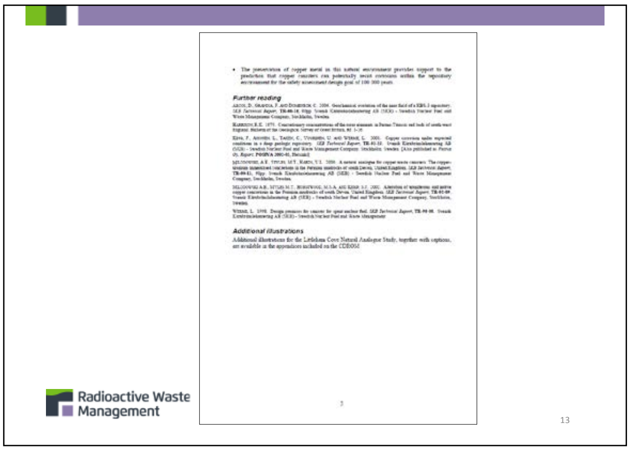
A few snapshots

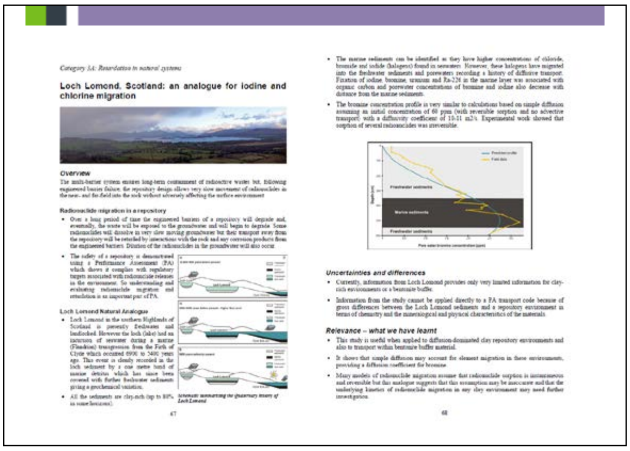
Radioactive Waste Management

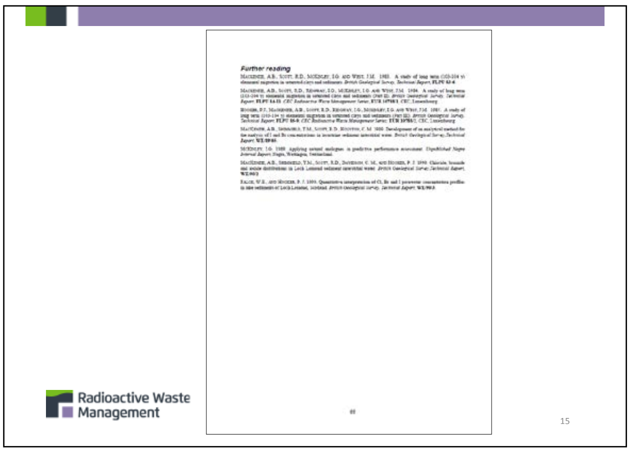












What the catalogue 'is', 'is not' and 'could be'

- It is a high-level summary 'short snapshots' of a range of natural analogues
  - Demonstrates a good understanding of natural processes
- Aimed at regulators, policy makers, managers, decision makers and others involved in the waste management process who are not necessarily specialists in natural systems
- Fills the gap between 'specialists in the field' and 'non-scientists'
- Respects the intelligence of the audience without overestimating knowledge – the layered approach provides a route to further information
- Presented using clear scientific language in an accessible way

Example……

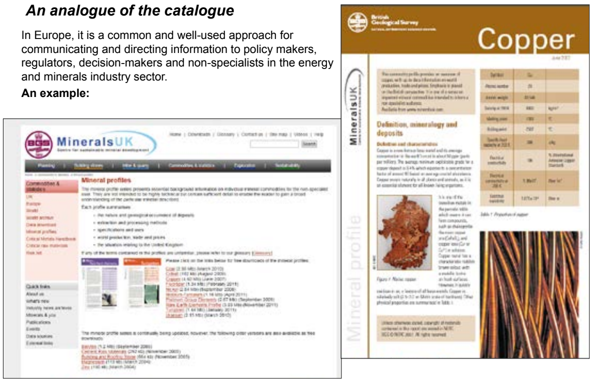
Radioactive Waste Management

23. Norris, Milodowski, Alexander, West & McEvay: Using natural analogues to build stakeholder confidence in geological disposal "A catalogue of natural analogues for radioactive waste management".

An analogue of the catalogue

In Europe, it is a common and well-used approach for communicating and directing information to policy makers, regulators, decision-makers and non-specialists in the energy and minerals industry sector.

An example:



What the catalogue 'is', 'is not' and 'could be'

- Is not a catalogue of **all** known natural analogues
- It is not intended to be highly technical but to contain sufficient detail to enable the reader to gain a broad understanding of the particular analogue described
- Does not intend to reach **all** audiences
  - It is one component of a multi-pronged approach
- Combined with other forms of communication, it could be a powerful resource
  - e.g. social media could signpost to the catalogue
- Combined with an interactive web-based interrogation tool of the different components of a GDF, it could be accessible to anyone across the web
- Considerable potential as an education tool aimed at A level students and or undergraduates

Radioactive Waste Management

Going forward….

1. Catalogue to be completed and published ("*A Catalogue of natural analogues for radioactive waste management*", BGS Commissioned Report CR/10/XXX, 2015)
2. RWM's gDSSC being updated, to be ready for publication late 2016 – will include use of analogues, building on 2010 gDSSC and related intervening studies
3. *Modelling Alkali-Rock Reaction: the Maqarin Natural Analogue*, C. Watson, D. Savage, J. Wilson and S. Norris. *In prep.*
4. *Minimal alteration of montmorillonite following long-term reaction in natural alkali solutions*: implications for geological disposal of radioactive waste, A. Milodowski, S. Norris and W. R. Alexander. *In prep.*

Radioactive Waste Management

19

For open discussion

The catalogue was collated from a generic perspective without a specific site, reflecting UK position

1. How could it be used more usefully?
   Could it be taken further?

2. Could it link to the CCR approach?
   Could CCR be used to support a UK Safety Case?

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20

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This book contains abstracts and extended abstracts submitted for presentation at the 14th Workshop of the Natural Analogue Working Group (NAWG), held in Rauma, Finland on 9–11 June 2015. In addition, the PowerPoint presentations are provided in the Appendix at the end of this book.

The workshop organisers would like to express their sincere thanks to Posiva Oy for providing the workshop facilities in Vuojoki and Olkiluoto and for organising an interesting visit to their low and intermediate level nuclear waste disposal facility. The editors wish to thank all authors for their contributions and to them and the other workshop participants for their active involvement in the discussions during the workshop. We also offer our thanks to the Geological Survey of Finland (GTK), which supported the organisers in numerous ways and provided the opportunity to publish this book in the GTK publication series.

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