SAANIO & RIEKKOLA OY
CONSULTING ENGINEERS

Bentonite studies: Current status - what remains to be done?

Heini Laine* & Russell Alexander

*heini.laine@sroy.fi

Recent reviews in the field of bentonite analogues

• **Posiva 2013.** Safety case for the disposal of spent nuclear fuel at Olkiluoto - Complementary Considerations 2012. Eurajoki, Finland: Posiva Oy. POSIVA 2012-11. ISBN 978-951-652-192-6.	POSIVA
• **M.Sidborn, N.Marsic, J.Crawford, S.Joyce, L.Hartley,A.Idiart, L.M. de Vries, F.Maia, J.Molinero, U.Svensson, P.Vidstrand & W.R.Alexander 2013.** Potential alkaline conditions for deposition holes of a repository in Forsmark as a consequence of OPC grouting. SKB R-12-17, SKB, Stockholm, Sweden.	SKB
• **W.R.Alexander 2013.** An assessment of the long-term durability of proposed shaft seal materials under highly saline groundwater conditions using natural analogues. Bedrock Geosciences Technical Report BG13-04 for NWMO, Toronto, Canada.	NWMO
• **Wilson, J., Savage, D., Bond, A., Watson, S., Pusch, R. & Bennett, D. 2011.** Bentonite - A Review of key properties, processes and issues for consideration in the UK context. Quintessa report. QRS-1378ZG-1.1	NDA
• **Laine, H. & Karttunen, P. 2010.** Long-term stability of bentonite – a literature review. Eurajoki, Finland: Posiva Oy. Working Report 2010-53.	POSIVA

Aims of this talk

- Identify the gaps in the current literature based on the recent reviews (previous slide)
- Elaborate thinking also to other concepts than addressed above
- Present potential novel study topics

FEPs discussed in this presentation:

Elevated temperatures

(Alkalinity)

Dilute groundwater

Mechanical stability

Highly saline groundwater

Microbial activity

Freezing/thawing

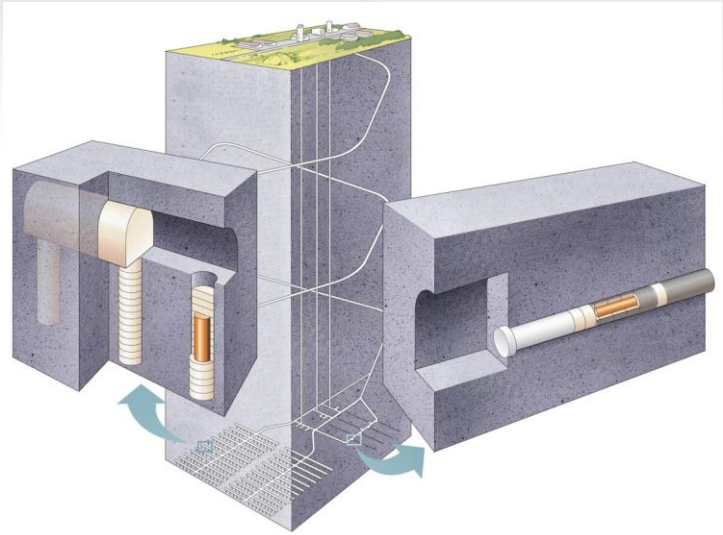
Cement –bentonite interaction is not discussed here (discussed in a separate presentation)

Posiva 2013 review on KBS-3 specific analogues (CCR)

- Main outcomes:
  - There is a vast data set available to be used to enhance confidence in process understanding and the over all safety case
  - Site specific conditions set greater demands in the natural analogues and their direct relevance
  - There are still topics that are not covered in the full potential by means of NA

Some examples on the future possibilities

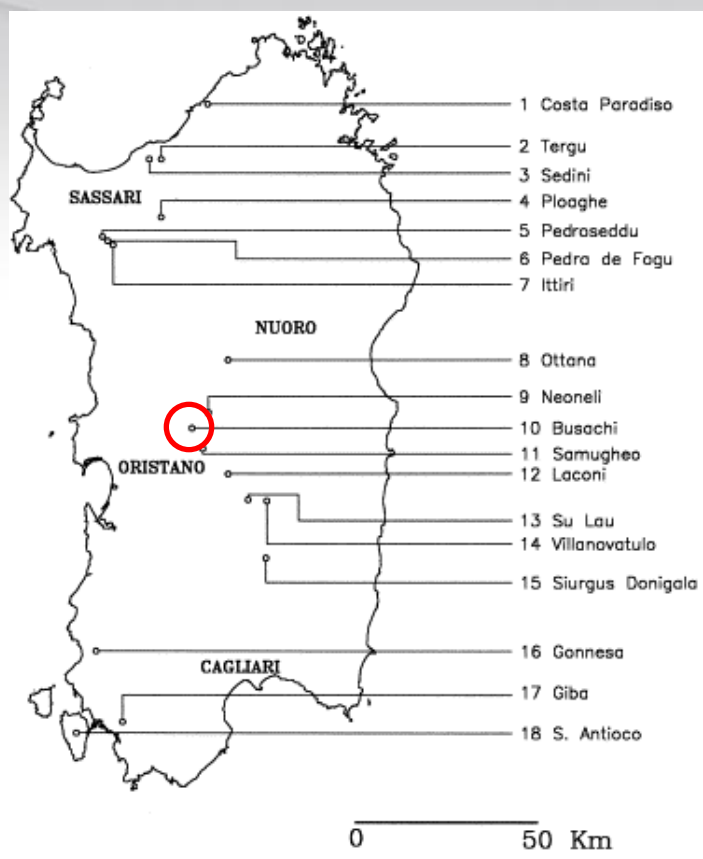
- Thoughts raised after Posiva 2012 safety case
- Other considerations for other repository systems



Themal alteration

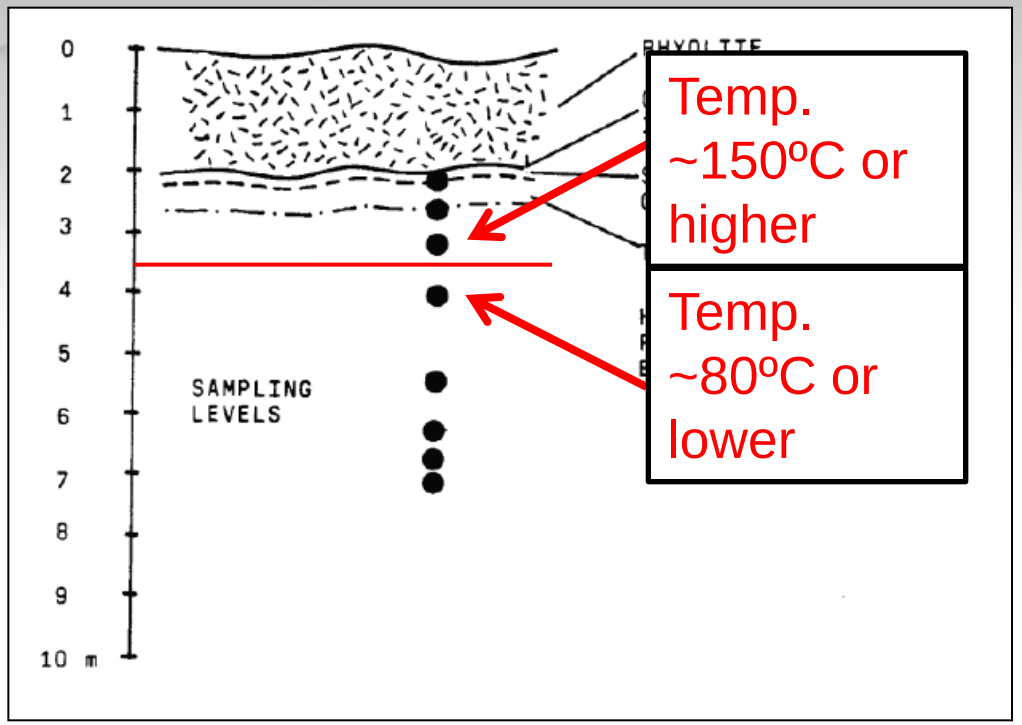
Buffer – thermal alteration/cementation

- Wide range of studies available (see Posiva 2013)
- Environments studied have little relevance to repository conditions
- Possibility:
  - Return to more relevant sites (e.g. Busachi) (Pellegrini 1999 and Woods et al. 2000) and apply a range of more relevant analytical techniques to better understand the site history and temperature profiles.



Busachi

- Original study: Pusch & Karnland 1988 evidence that significant heat-induced dissolution of smectite occurred at 150 to 200 ºC and precipitation of siliceous material occurred during cooling
- Open questions:
  - the flux of K through the site? Has this been a limiting factor?
- re-sampling at the site and standard clay analyses? More precice definition of temperatures?



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

Mechanical deformation

Buffer – canister sinking

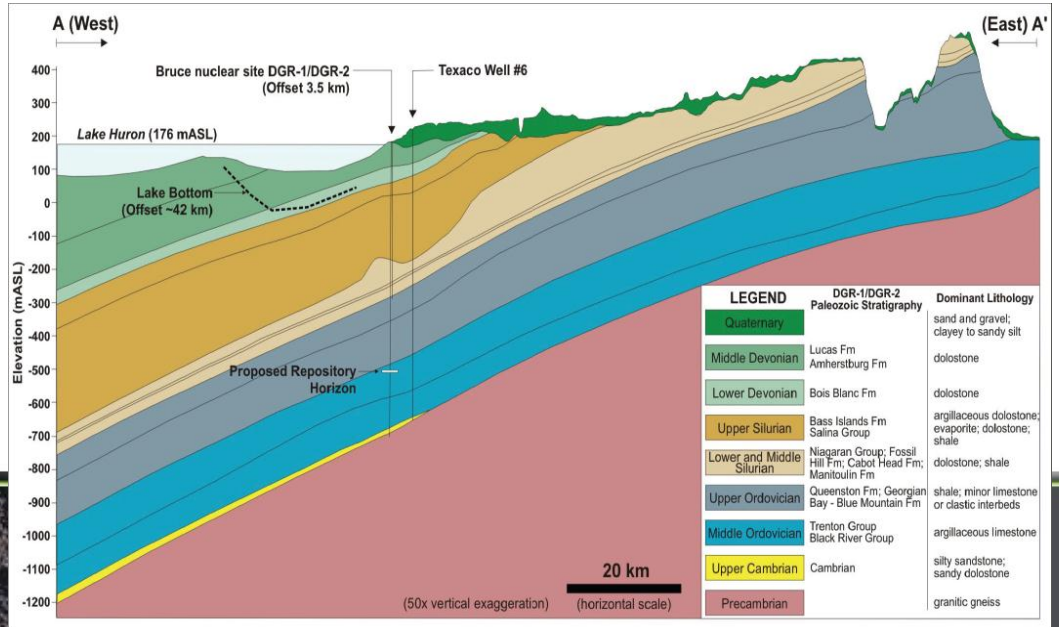
- No relevant studies available at the moment
- The approach of Keto (1999) could be repeated plus the process should be included into current URL experiments (e.g. FEBEX)

Chemical alteration

- Saline environments
- Bentonite – copper and bentonite – steel interaction
- (cement – bentonite interaction, discussed in another presentation)

Buffer – chemical alteration in saline environment

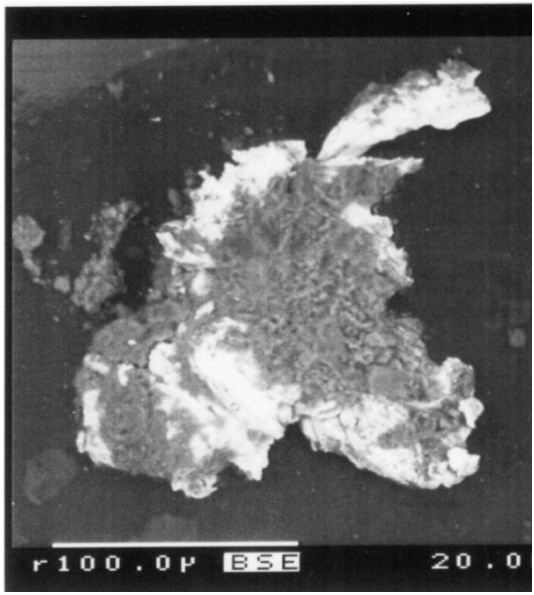
- Various studies available, but mainly marine salinities (cf. Bruce with Cl 370 gL^{-1})
- Relevance to KBS-3: no full characterisation of ‘reacted’ samples have been published
- Find an appropriate site and carry out a more rigorous analysis of all relevant parameters
- Of relevance also in evaporites

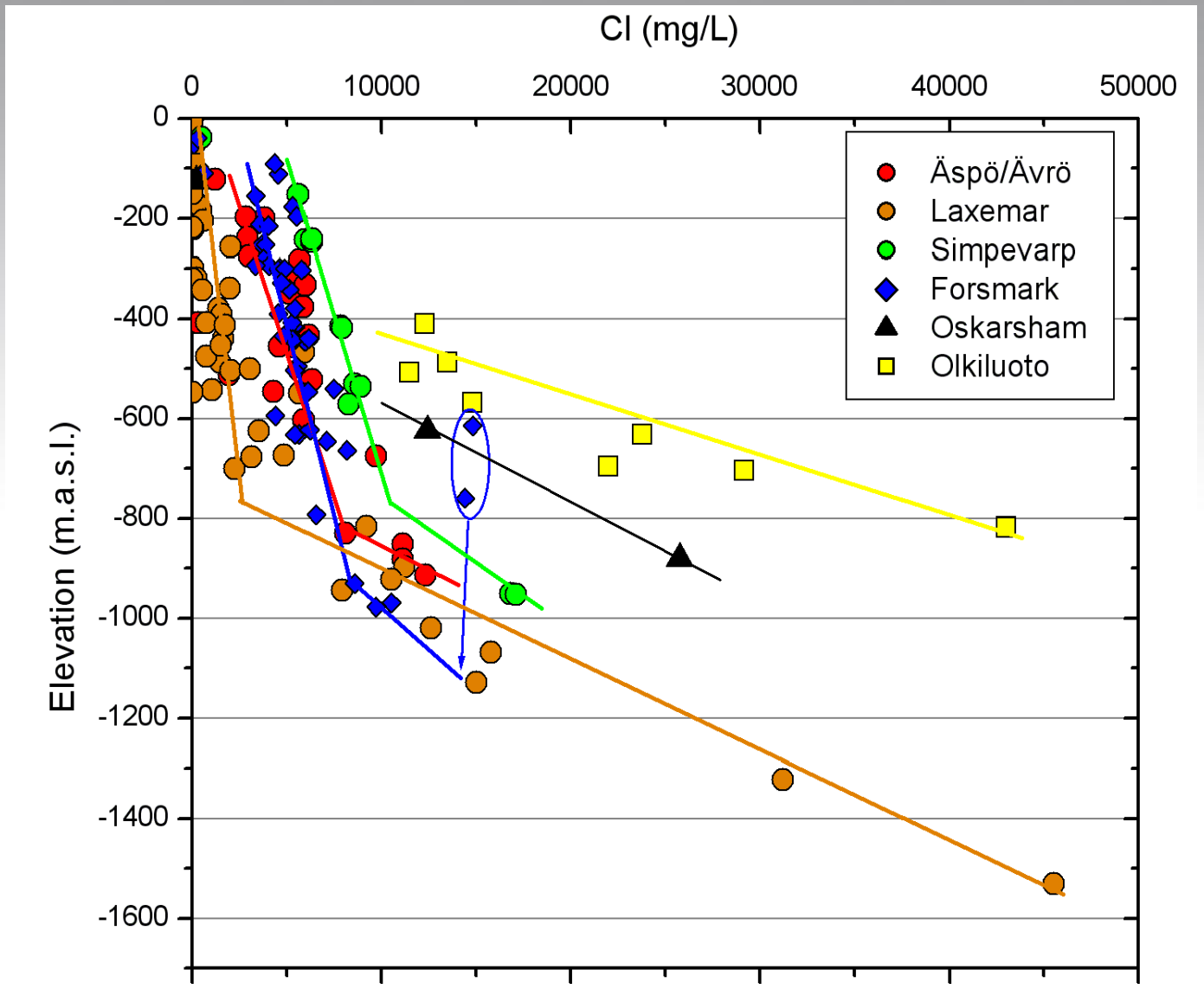


- Canister interaction – copper Kronan, qualitative data available
  - Data could be more rigorously integrated with existing/new laboratory data
- Canister interaction – steel
  - Existing natural analogue are not relevant enough for KBS-3 or other designs, although general observations from natural systems and the NF-PRO URL experiment suggest uptake of iron could reduce swelling pressures. These data need to be integrated with a more appropriate natural analogue study

Bentonite in saline environments – novel studies?

- Would provide direct site specific analogue
  - Montmorillonite occurs at all depths in Finland (Olkiluoto)
  - Also present at Sweden (Forsmark & Laxemar) sites.
  - Possibility: to study the occurrences of this mineral from the perspective of the controls that have preserved it
    - Mineralogical paragenesis?
    - How occurrences are affected by gw circulation? Or are they protected?
  - Not discussed in detail from this perspective previously.

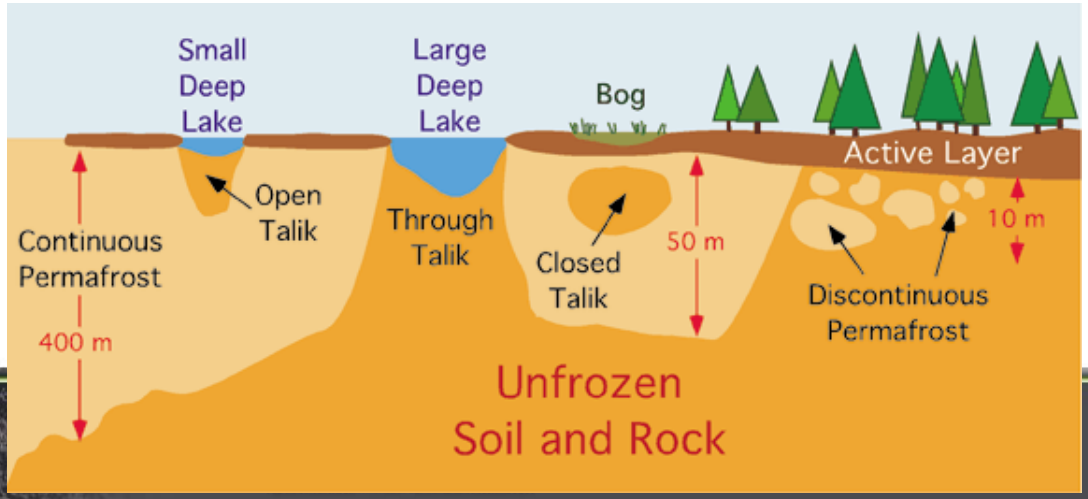



Cl (mg/L)
0
10000
20000
30000
40000
50000
0
-200
-400
-600
-800
-1000
-1200
-1400
-1600
Elevation (m.a.s.l.)
Äspö/Ävrö
Laxemar
Simpevarp
Forsmark
Oskarsham
Olkiluoto

- Groundwater chlorinity reaches around 45 gL^{-1} at depth
- At Larsmark and Forsmark sites, smectites are reported in closed and open fractures throughout the depth profile
- At Forsmark, dating of the fracture systems suggests that the smectites have been present since at least the Palaeozoic (i.e. 540 to 250 Ma BP) with the deep brines in contact with the smectites having been stable for at least several million years, if not much longer
- Similar understanding prevails also for Olkiluoto
- Unfortunately, no swelling pressure data are available for the material although sampling archived cores would be possible

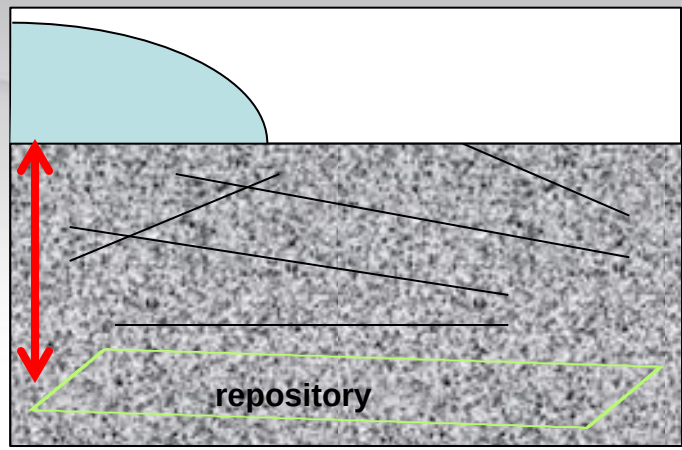
Assessing the impact of future glaciations and periglacial processes

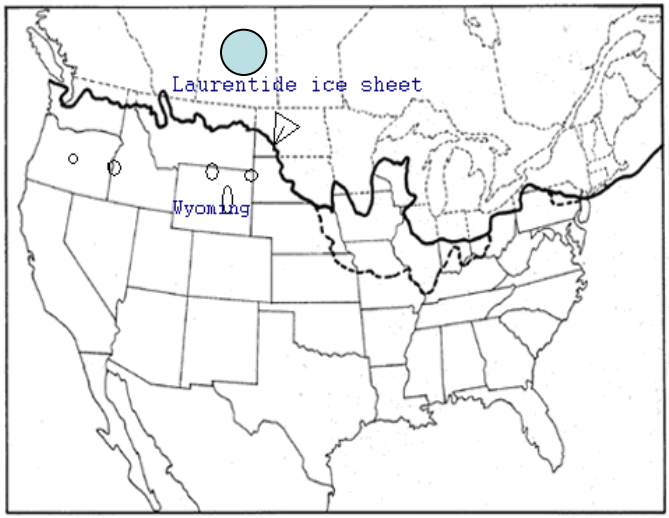
- Chemical erosion
- Freezing and thawing



Assessing the impact of future glaciations and periglacial processes

- Chemical erosion due to dilute conditions combined with flow
  - Unlikely at repository level due to thick bedrock buffer
  - However, only one NA study available
  - Possibility: to study bentonites quantitavely under exposure to meteroric waters and constraints to erosion rate and relevant conditions
  - E.g. in Canada within the boundaries of LGM there are several bentonite occurrences, erosional history of these deposits?



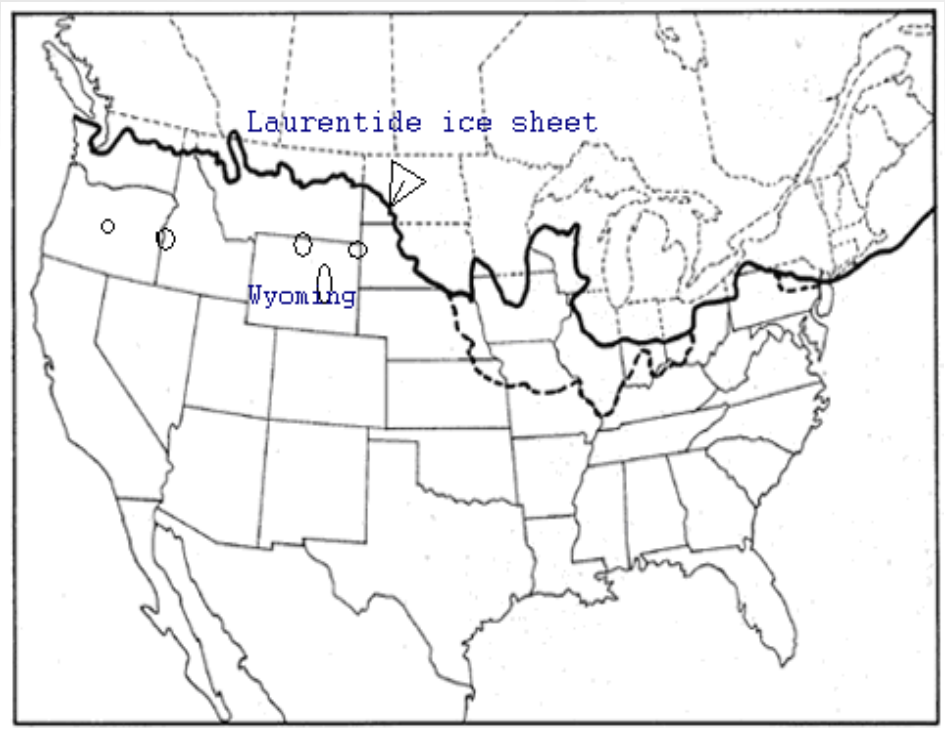


- Puura & Kirsimäe (2010) have considered erosion of bentonites in very general level in SKB TR-10-24.
- They conclude: Geological literature data on historical bentonites do not consider colloid formation in low ionic strength water as relevant mechanism for smectite mobilization. However there are no studied cases where this could be a relevant mechanism.
- They are focussing only on K-bentonites and try to look for an analogue there and possibilities to study erosion profiles.

→ Resulting in conclusion that relevance to the repository conditions would be rather poor, but some interesting data could be obtained…

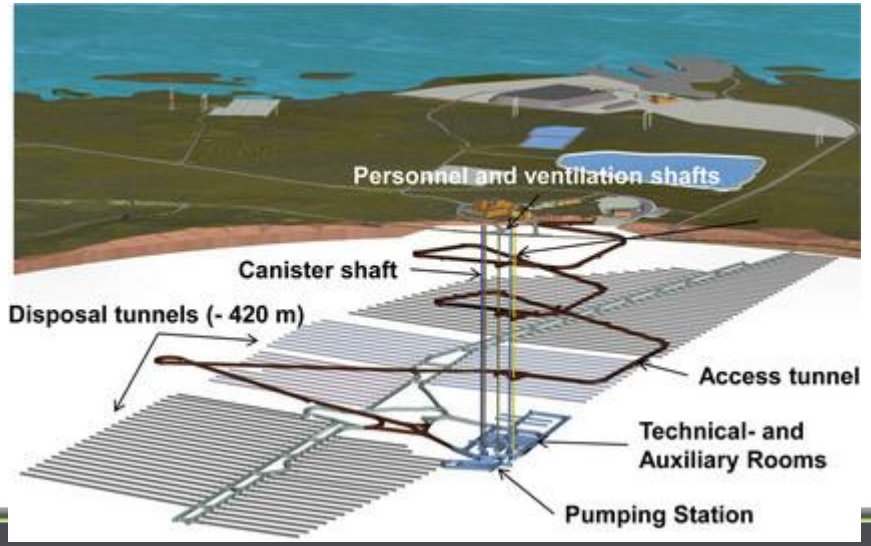
Freezing and thawing

- Permafrost is not expected to reach repository level
- Nevertheless, the process is considered in safety case
- No NA studies available
- Possibility: study those bentonites occurring within the glaciated/permafrost areas in more detail



- Related to both chemical erosion by dilute waters and freezing/thawing observations the knowledge on the erosional histories at the bentonite sites are poorly constrained → better constraints, better analogues

- Note: although these processes are not necessarily relevant at the repository depth the data is of interest regarding the closure components backfilling the access tunnels in repositories
- Also, important to be able to screen out some processes



Chemical stability of the bentonite in dilute conditions – novel studies?

- How do the smectite minerals occur in general in those environments where intended to be emplaced?
- E.g. in Finland → no bentonites → but smectites occur in various environments
- Montmorillonite/smectite occur as a fracture mineral at the selected site at Olkiluoto (reported in Posiva working reports)
- As discussed earlier from the saline point of view → same data set can be looked at from the dilute point of view

Benefit: Would provide direct site specific analogue

- Again, a possibility: to study the occurrences of this mineral from the perspective of the controls that have preserved it
  - Mineralogical paragenesis?
  - How occurences are affected by gw circulation? Or are they protected?
- Not discussed in detail from this perspective previously.

- Montmorillonite occurring in crystalline bedrock in general
  - There are many reported montmorillonite occurrences e.g. in Finland, but no detailed review has been done
  - Most data is reported as observations and are often not supported by quantitative analysis
  - In general montmorillonite is found in addition to **fracture filling mineral** as **alteration product in ancient weathering profiles** (Precambrian), **soils** and in **hydrothermally altered rocks**.

"The bedrock at Olkiluoto consists of high-grade metamorphic schists and gneisses and, therefore, many of the minerals were originally stabilised at high temperatures and pressures (e.g. cordierite, garnet and sillimanite). These high-grade metamorphic minerals are potentially more prone to rock-water processes than minerals more frequently found in crystalline granitic bedrock. This is indicated by the presence of smectites at Olkiluoto in the rock matrix and in the fracture zones that result from retrograde metamorphism rather than later rock-water interactions. ***The detailed properties of the smectites at the site (e.g. cation exchange capacities, chemical compositions, grain size distributions) are not well known****."*

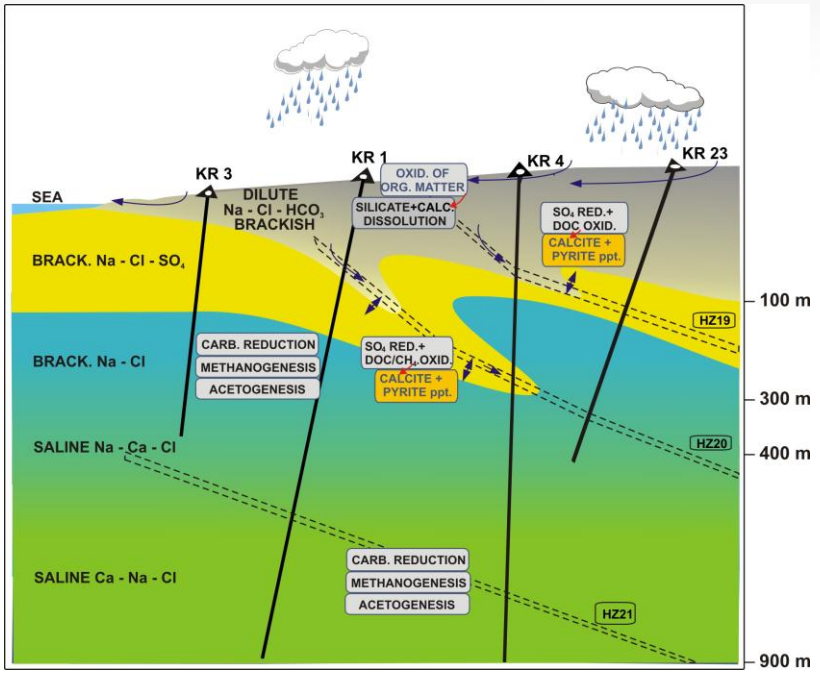
- Posiva FEP report (Posiva 2012-07)

- Possibilities?
  - Review the existing literature
  - Based on this locate possible targets for more detailed natural analogue studies
  - Bring understanding of the montmorillonite occurrence on site to higher level
  - Regionally try to find site where montmorillonite would occur close to surface in larger quatities than what's found in fractures → allow also consideration of weathering/erosion…

- Dilute conditions could be studied also through meteoric water
- Controlled erosion experiments at bentonite sites where a stream flows through deposit (cf. Kamei study)?
- Is there something to be gained in addition to the experimental work in the lab? Care has to be taken also between studies made by using natural occurrences, actual bentonite products and on the other hand extremely purified montmorillonites.

To sum it up

- Fracture smectites should be studied considering the full range of groundwater compositios
- Surficial processes could be studied separately in case appropriate site is found



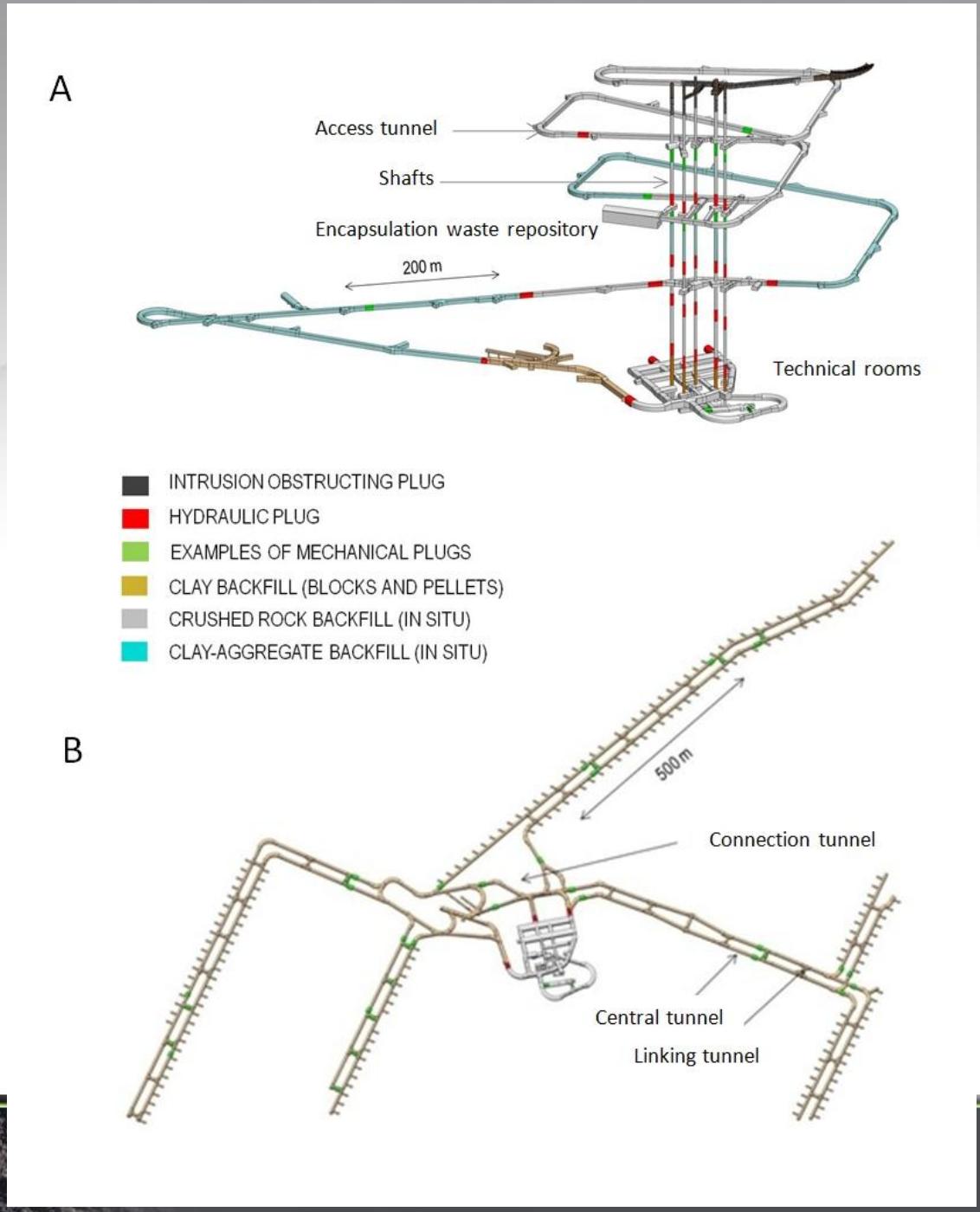
Microbial activity

- Discussion raised (again) at Montpellier 2012 Clay conference
- Basic problem seems to be how it can be assured that microbial activity does not affect the safety funtions of the clay host rock or clay based EBS components? See e.g. Meleshyn (2012)

- Low metabolic rates are suggested to be relevant during long periods of time.
- Metal-clay interactions, role there?
- New information on natural colloid populations are of interest if microbial processes are to be modelled.
- Some work has been done, but new studies are welcome

Bentonite – is that all there is?

- In addition to bentonite, the backfill materials planned to be used in the various types of repositories to seal and eventually close the repository cover bentonite/sand, bentonite/aggregate, other clays, till, etc.
- Variability of materials and conditions to be expected vary to those of the repository in the acces routes coming to the surface.
- NAs have not been utilised to their full potential in these cases.


A
Access tunnel
Shafts
Encapsulation waste repository
200 m
Technical rooms
INTRUSION OBSTRUCTING PLUG
HYDRAULIC PLUG
EXAMPLES OF MECHANICAL PLUGS
CLAY BACKFILL (BLOCKS AND PELLETS)
CRUSHED ROCK BACKFILL (IN SITU)
CLAY-AGGREGATE BACKFILL (IN SITU)
B
500 m
Connection tunnel
Central tunnel
Linking tunnel

Couple of examples

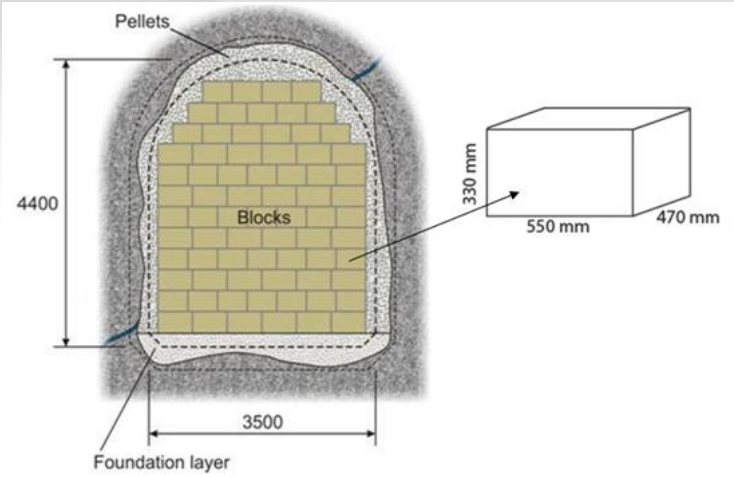
- Bentonite/sand mixtures in saline conditions
- How NAs can help in estimating the sulphide fluxes from the Clay backfill in KBS-3V concept?

1. Bentonite-sand mixtures in saline conditions

- Many natural bentionites are closer to bentonite-sand mixes than pure industrial bentonite
- Although not a focus of the original CNAP study, the data have been re-assessed from this viewpoint for a recent NWMO study
- This topic is discussed in another talk in more detail

2. Sulphide fluxes from backfill

- In the Performance Assessment within the Posiva Safety Case 2012, the maximum sulphide concentrations in the backfill have been assessed by thermodynamic calculations and by comparison of these results with **natural analogue** data in Wersin et al. (2013).



Wersin, P., Pitkänen, P., Snellman, M., Román-Ross, G., Smith, P., Filby, A., Kiczka, M. 2013. Sulphide fluxes in the spent nuclear fuel repository at Olkiluoto. Eurajoki, Finland: Posiva Oy. Working Report (to be published).

Reference backfill material (Friedland clay + bentonite pellet components), is compared to Opalinus Clay, Collovo-Oxfordian clay and Boom Clay.

- These rock types, which geochemical properties have been extensively investigated, display fairly similar mineralogy as the backfill material,
- notably all contain pyrite and siderite as accessory minerals.
- The combined set of information suggests that undisturbed pore waters of OPA, COx and Boom Clay display low levels of sulphide (below detection limit), presumably in equilibrium with pyrite, the main sulphur phase.

- Information gained in the clay formation studies on the sulphide concentrations and knowledge on the controls over higher consentrations is utilised in the performance assessment.
- See Posiva (2012) for more details (6.6.3 Production of sulphide and microbial activity in backfill).

Safety case for the disposal of spent nuclear fuel at Olkiluoto – Performance Assessment 2012. Eurajoki, Finland: Posiva Oy. POSIVA 2012-04. ISBN 978-951-652-185-8.

Selecting the data

- The use of NAs in safety cases development are likely to be
  - site specific
  - repository design-specific
- Emphasis is to be placed on the appropriate use of relevant NA data on bentonite longevity.
- “Complementary Considerations” or “Complementary Evaluations” type approach provides a natural place for such a case specific review
  - Also helps in pointing out research needs.
  - Can be done at any stage of the disposal project on a relevant level of detail.

Way forward

- What's missing?

- Review

Proceedings of NAWG WS 2013 planned (Laine & Alexander 2013)

- New studies?

Thank you!