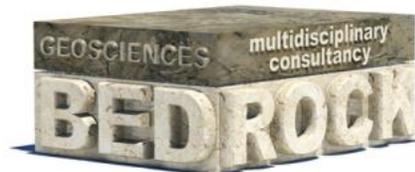


# The use of natural, industrial and archaeological analogues in support of deep borehole seal design

Russell Alexander<sup>1</sup>, Nick Jefferies<sup>2</sup> & Simon Norris<sup>3</sup>

1. Bedrock Geosciences, Auenstein, Switzerland (russell@bedrock-geosciences.com)
2. Amec Foster Wheeler, Didcot, UK (Nick.Jefferies@amecfw.com)
3. Radioactive Waste Management Ltd, Didcot, UK (Simon.NORRIS@nda.gov.uk)



# Background

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- In the UK, RWM is undertaking RD&D into borehole (BH) sealing because they expect that the Initial Site Evaluation will require a clear description, supported by reference to RD&D and technology demonstrations, of how site investigation boreholes might be sealed
- Here, we'll present results from one of the tasks undertaken as part of a three year project to develop **generic** approaches to seal deep repository site investigation BHs

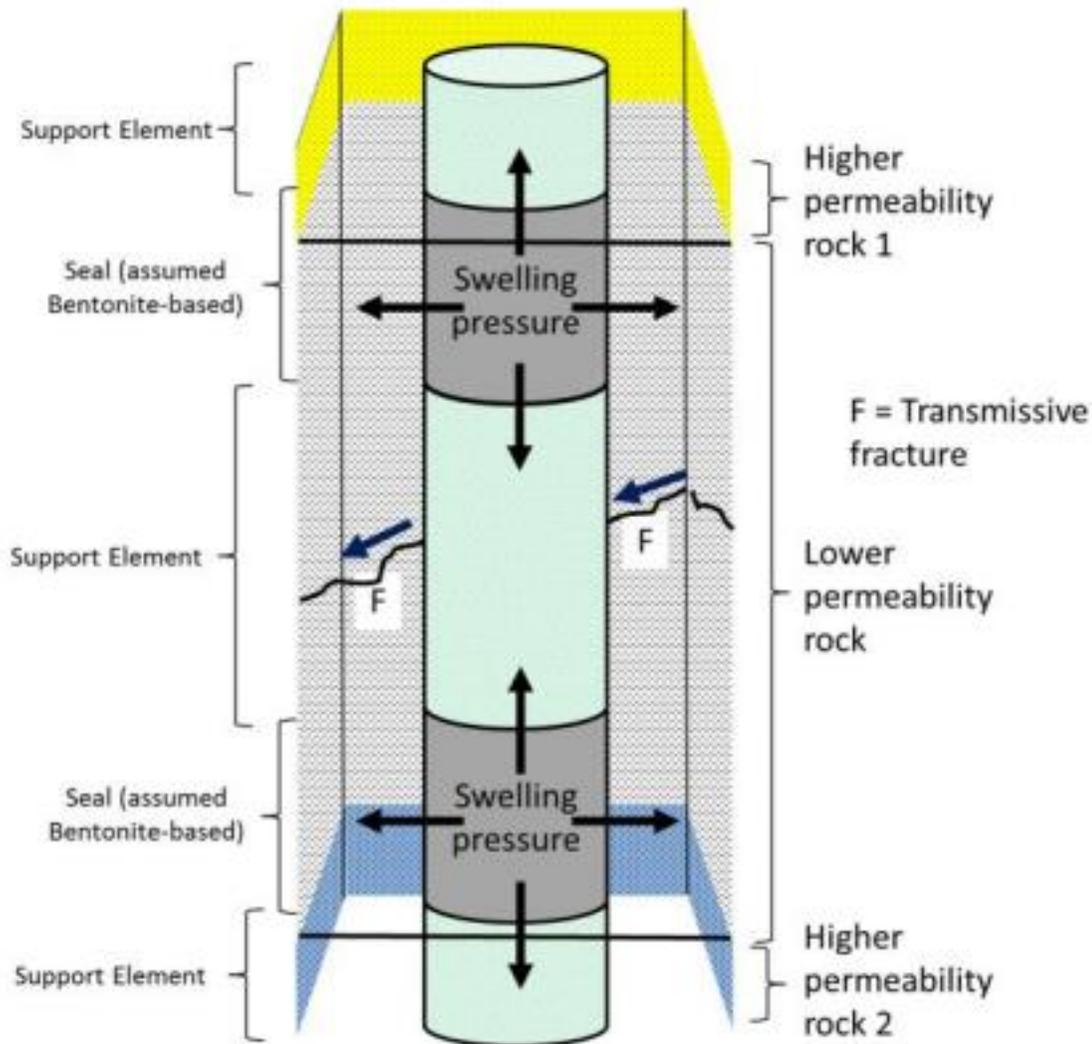
# Background

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**The key issues for generic research into BH sealing were identified to be:**

- to determine the functional requirements of BH seals**
- to determine the standard of sealing that is likely to be required from the BH seal recognising that this will be uncertain at the generic stage**
- to determine the most appropriate seal concept and emplacement method for each generic geological environment, and to evaluate the advantages, disadvantages and risks associated with the different sealing concepts**
- to identify how the quality of the emplaced seals and support elements can be assured**
- to identify appropriate materials for post-closure support elements, taking into account mechanical stability and chemical interactions with adjacent seals.**

# Background



**Schematic illustration of a section of BH showing an arrangement of seals and support elements.**

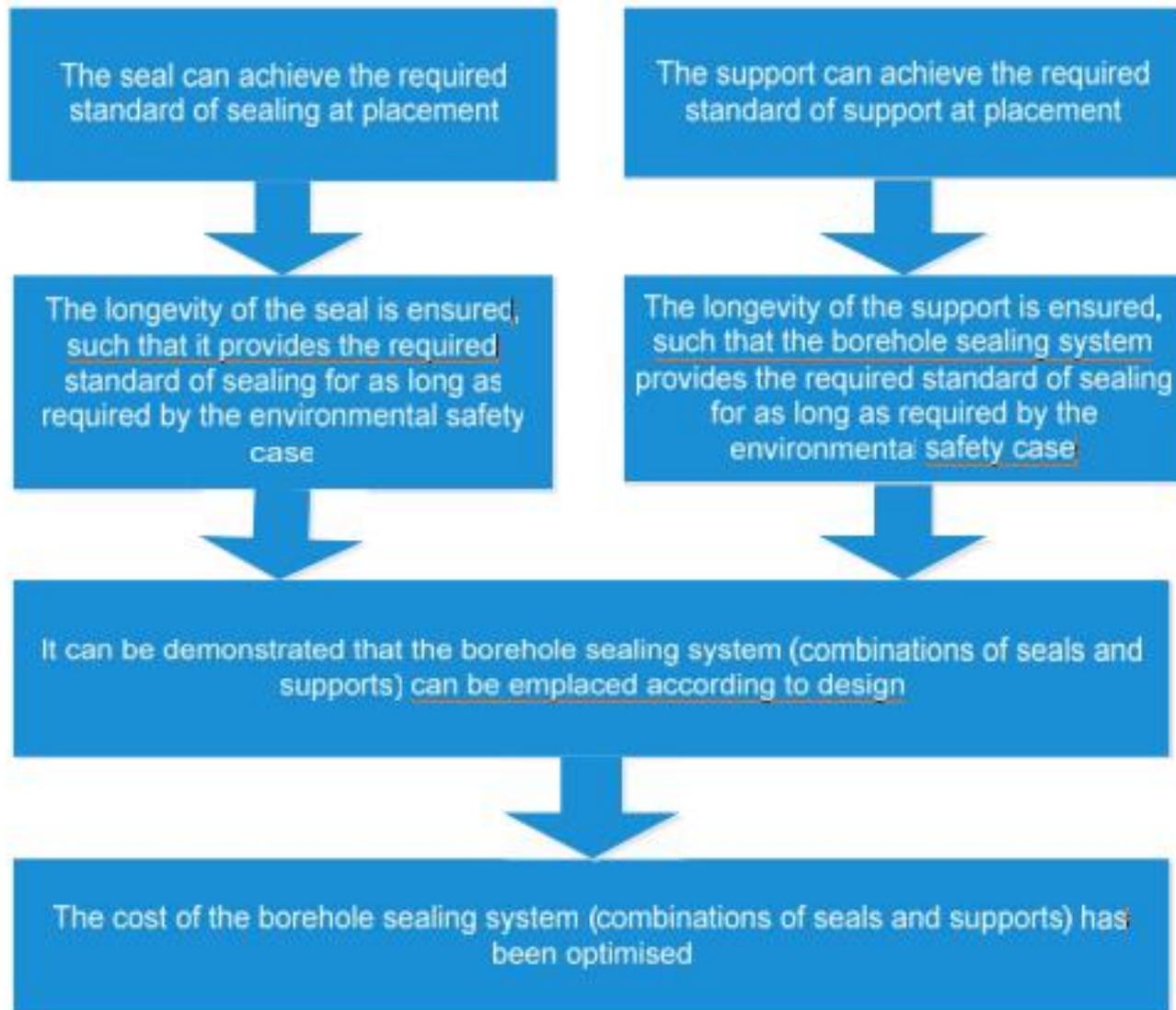
**Seals are located within the lower permeability rock. A support element is placed across a high transmissivity feature within the low permeability rock and in the adjacent higher permeability rocks**

# Background

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- The BH seals should provide resistance against groundwater flow and gas migration, thus limiting the potential for radionuclide transport in a range of geological settings of relevance for a future UK repository
- The use of information provided from natural, industrial and archaeological analogues will support generic designs for BH seals (and seal supports)
- There is **no research reported in the open literature** relating directly to NA of BH seals, but general information on the behaviour and properties of relevant materials does exist

# Background



# Background

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- This information has been assessed with the aim of extracting data of relevance to the project
- The main areas of interest concern:
  - materials **longevity** (for the BH seals and supports)
  - **interaction** between proposed materials
  - mechanical **stability** of BH seals and supports
- The materials of focus are **bentonite** (seals), **OPC** (Ordinary Portland Cement), **low alkali and salt cements** (supports), **copper** (bridge plugs), **steel** (BH casing) and a range of **lithologies** (BH walls)

# Organisation of presentation

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- This presentation is organised in three main sections:
  - Sealing materials, with focus on those materials (e.g. bentonite) and forms (e.g. pellets) **identified elsewhere in the project**
  - Supporting materials, which will be used to fill sections of BH between seals
  - Summary and conclusions
- **Note that reactions could be more or less significant than in the repository EBS due to differences in the relative volumes of materials present**

## Seals: general comment on bentonite

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**“Industrial bentonite, such as the ‘bentonite’ that will be used in the EBS of a radioactive waste GDF, is derived from natural bentonite rock but is usually processed to improve the smectite content (i.e. beneficiated) of the material. It can also be chemically treated to enhance its cation exchange, swelling or other physico-chemical properties (e.g. conversion or “activation” of natural calcium montmorillonite to sodium montmorillonite by treatment with sodium carbonate). **As such, the physical and chemical properties of industrial bentonite may differ significantly from that of the natural rock and this limitation must be borne in mind....”****

**Alexander & Milodowski (2014)**

# Seals: HDBC (high density bentonite block concept)

- **When emplaced, the seal material comprises of well-fitting blocks of highly compacted bentonite**
  - *(e.g. pre-dried to a water content of about 6% and then compacted to a dry density of  $1,900 \text{ kg m}^{-3}$ )*
- **Like to understand how bentonites with similar densities behave in the BH. Issues are likely to be around**
  - homogenisation
  - erosion
  - cation exchange with groundwater



Jackson et al. (2014)

# Seals: HDBC (high density bentonite block concept)

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- Physico-chemical data on natural bentonites from the vast majority of existing NA studies **have little relevance to the HDBC**

**If we were to go further:**

- **New samples could be collected (using triple barrel drills) and which have come from bentonite mines (e.g. Tsukinuno in Japan) or elsewhere (e.g. Kato Moni in Cyprus) where the bentonite is under a natural confining pressure**
- **These samples could then be assessed to establish bentonite densities, degrees of saturation etc for comparison with the properties required of a bentonite seal**

# Seals: bentonite pellets

- Of interest would be whether the boundaries between pellets act as interfaces or whether the expected swelling behaviour of the bentonite will eradicate any such boundaries, so producing homogenous seals over long time periods.
- **Reworked** lapilli-derived bentonites (such as are expected in the Pinatubo lahar fields in the Philippines) would appear to offer a realistic analogy of bentonite pellets
- Modern Pinatubo is around 35 ka old, but the original somma volcano is around 1 Ma old, offering the possibility to examine reworked lapilli-derived bentonites of relevant ages

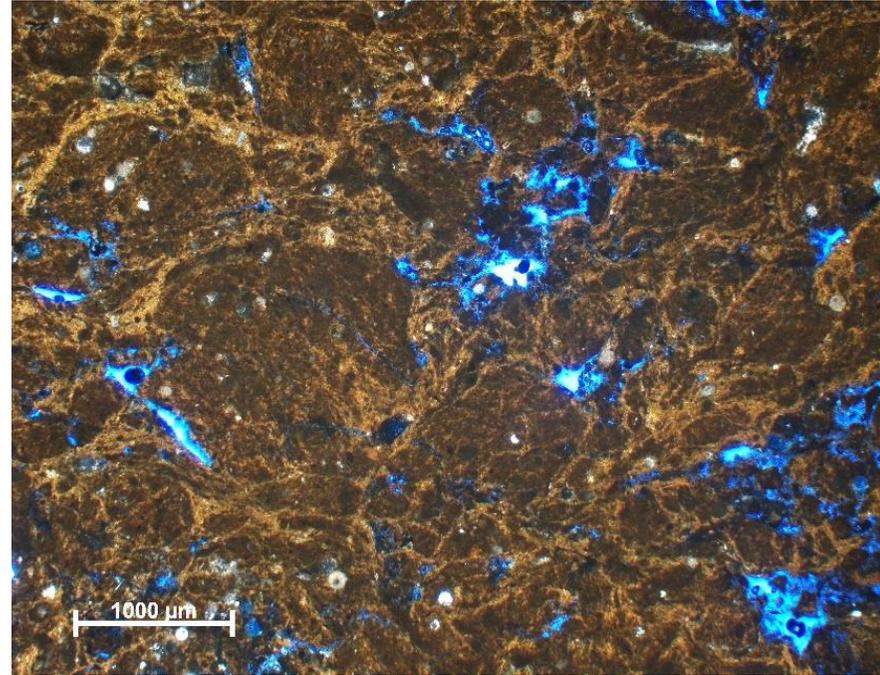


Courtesy C. Arcilla, University of the Philippines

# Seals: bentonite pellets

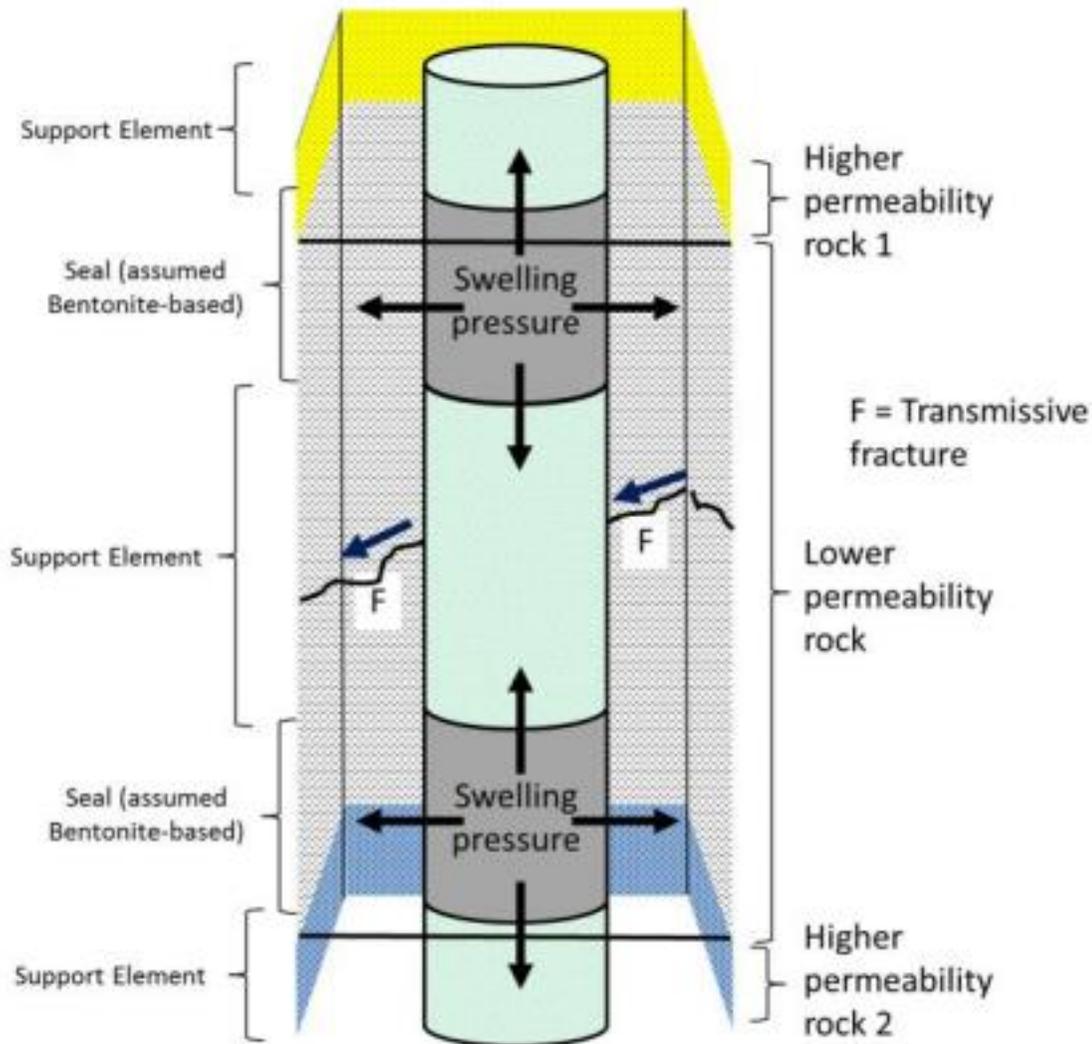
➤ The LPB (Long-term Performance of Bentonite) study at Kato Moni (Cyprus) concluded that both natural and industrial bentonites are intrinsically susceptible to shear under certain loads, implying the bentonite will continue to creep to fill voids or to homogenise density variations over long time periods – although the final density may be rather low

➤ However, in the micro-pellet bentonite, anastomosing shear plane networks are produced and this appears to prevent shear displacement to some degree **and this may need to be considered in the final design of a bentonite seal -**



Alexander et al. (2017)

# Seals



**Schematic illustration of a section of borehole showing an arrangement of seals and support elements.**

**Seals are located within the lower permeability rock. A support element is placed across a high transmissivity feature within the low permeability rock and in the adjacent higher permeability rocks**

# Seals: potential bentonite reaction

## Bentonite-OPC

- Harpur Hill and Herbert's Quarry industrial analogue sites (former cement works) appeared promising, but ultimately neither included sites where OPC fluids reacted with clays



# Seals: potential bentonite reaction

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## Bentonite-OPC

- Despite extensive work conducted in Jordan, there are **no unambiguous NA data on OPC-bentonite reactions**
- Techer et al. (2006) state that the (Daba) clays have reacted with alkaline solutions, dissolving smectites and leaving zeolites as reaction products
- But Pitty & Alexander (2011) noted that “Zeolites have been found.....in the matrix and the weight of evidence suggests that these phases have been produced by interaction with alkaline fluids. However, it is not yet possible to rule out if the matrix zeolites are a primary diagenetic feature of the sediments without further isotopic analysis.”
- **But BH geometry works for us, could have sacrificial loss. So no further assessment required**

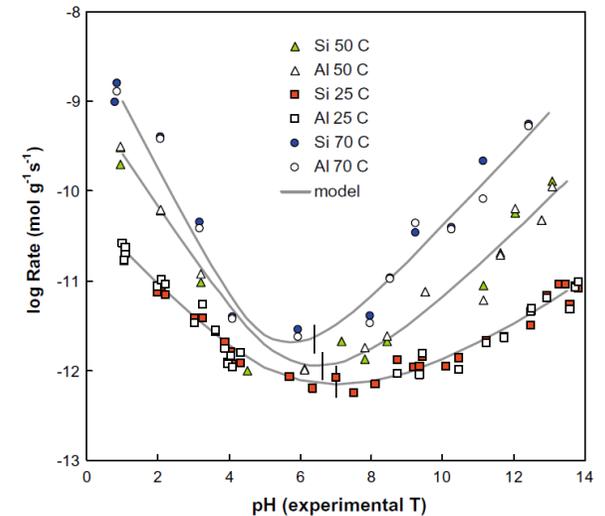
# Seals: potential bentonite reaction

## Bentonite-low alkali cement

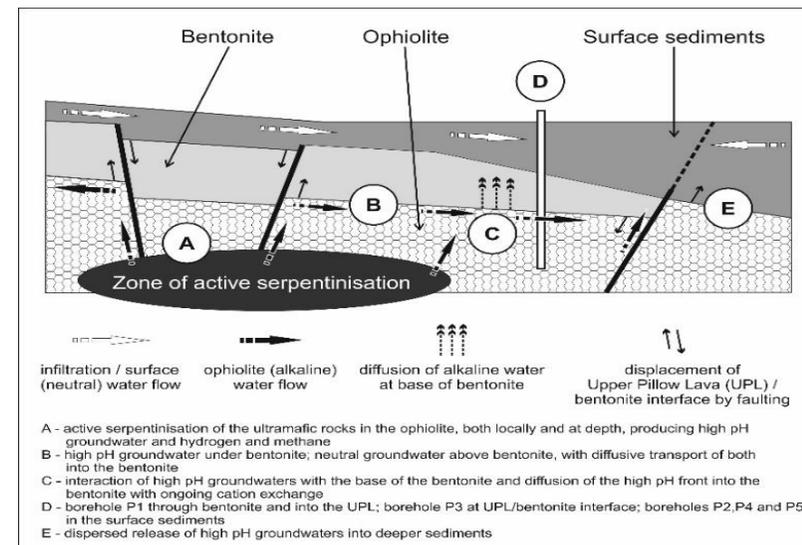
➤ Work in Cyprus and the Philippines indicates that bentonite reaction would appear to be minimal and that the bentonite retains its swelling capacity and acts as a seal, effectively trapping the alkaline groundwaters below it

➤ In Cyprus, the base of the bentonite shows a very small degree of reaction, with the smectites changing to palygorskite, Philippines to sepiolite

Milodowski et al. (2016)



Rozalen et al. (2009)



# Seals: potential bentonite reaction

## Bentonite-low alkali cement

- Palygorskite formation - is consistent with other natural systems and lab data (e.g. Moyce et al., 2014)
- Finally, while the Cyprus study notes that alkaline reaction has changed the nature of the exchangeable cations in the bentonite, the physical properties of the material remain largely unchanged
- Philippines – see Fujii et al. (this workshop)
- **No further work is recommended...**



# Seals: potential bentonite reaction

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## Bentonite-metal

- Interaction between bentonite seals and the metals in the BH casing and/or seal emplacement systems could cause a loss of bentonite isolation properties as, for example, metal ions from the canister exchange with cations in the clay
- For example, Wersin et al. (2008) noted that a simple mass balance estimate indicates that substantial amounts of montmorillonite (10–30%) would be transformed if all the iron from the super-container steel shell reacted with the bentonite buffer at Olkiluoto to produce a non-swelling Fe clay. **This would induce a decrease in swelling pressure from 7 MPa to 2 Mpa**
- But, at that time, these authors could find no iron/clay reaction analogues

# Seals: potential bentonite reaction

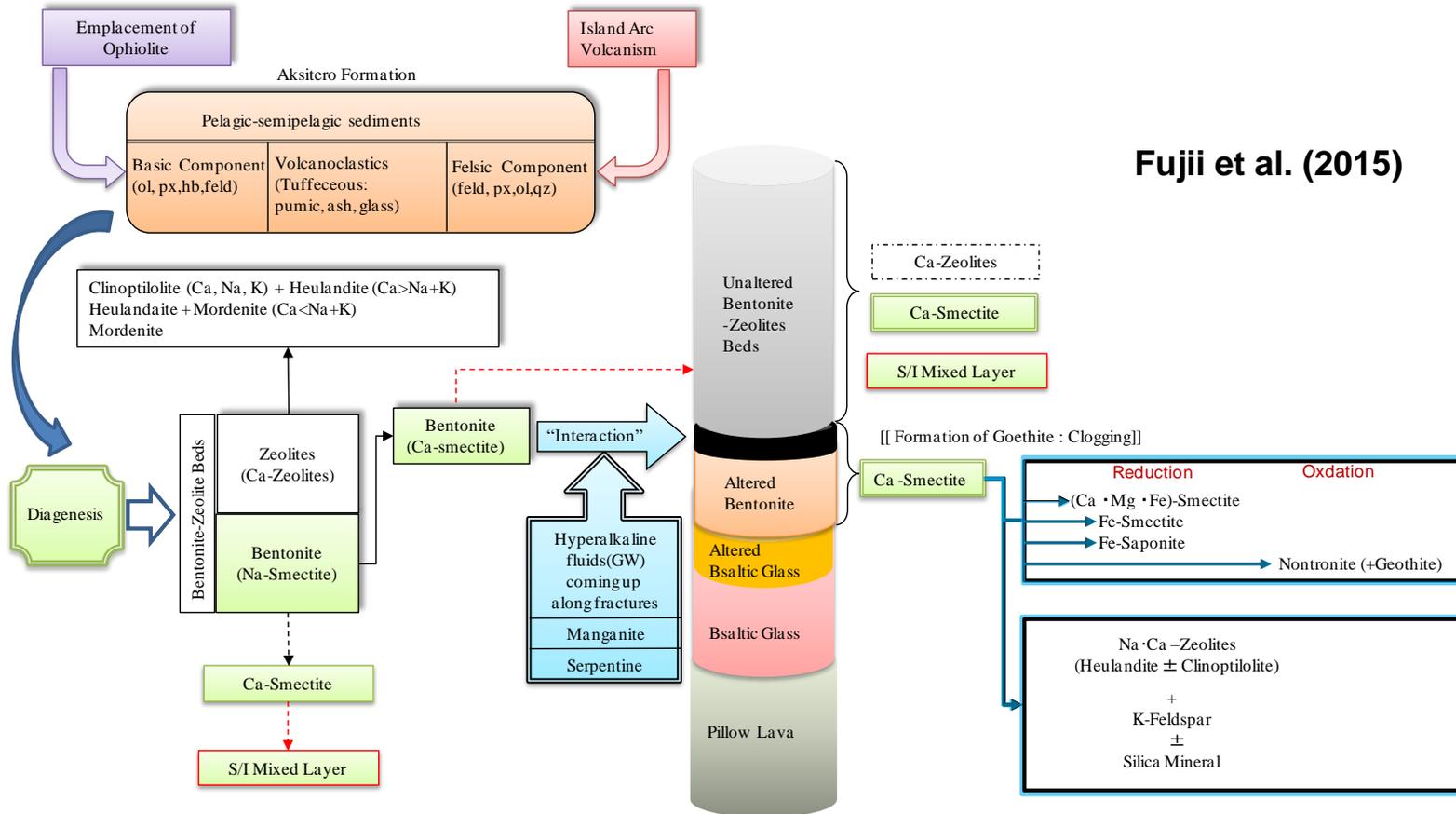
## Bentonite-metal

- Preliminary studies were carried out at the Cortijo del Aire bentonite quarry, but the smectite content (15 wt%) was deemed too low
- At the Kawasaki bentonite deposit in Japan, Fukushi et al. (2010) reported **hydrothermal reaction** producing Fe-rich smectites. But the secondary clay (and opal) were formed by dissolution and subsequent precipitation from the interaction of the original bentonite with the hydrothermal solution
- This is not what would be expected in a BH seal



Cortijo del Aire, Almería, Spain (left). The red staining of the bentonite is clear during sampling (right). From Marcos (2004)

# Seals: potential bentonite reaction



Further analysis of existing samples from Saile recommended

# Seals: potential bentonite reaction

## ➤ Bentonite-copper reaction

- Kronan cannon
- Littleham Cove native copper
- More focussed NA work recommended

## ➤ Bentonite-groundwater reaction

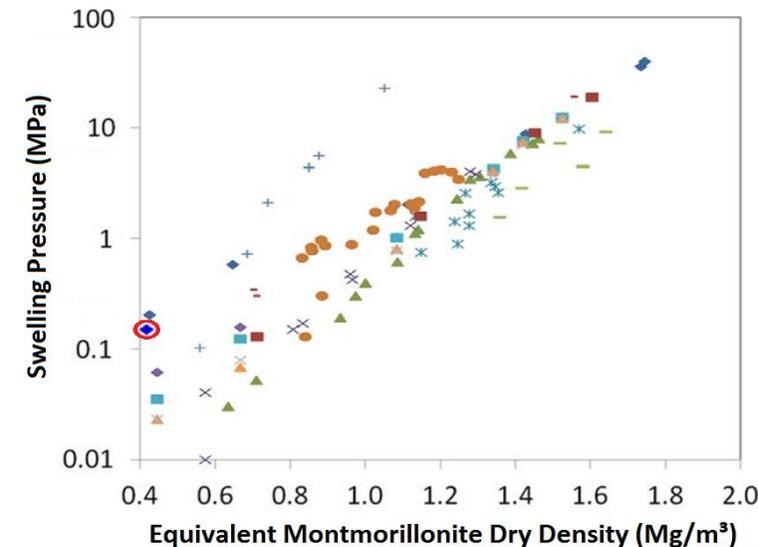
- Freshwater - Fennoscandian Shield, IBL
- Saline water - Fennoscandian Shield (self analogue)
- Brines - Bruce site (self analogue), Dead Sea

## ➤ Illitisation

- Large body of NA data available



Milodowski et al. (2000)



Kremer & Alexander (2015)

# Seals: potential bentonite reaction

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## Bentonite-non-clay

- **Quellon HD is a bentonite (~90%) - magnetite (~10%) mixture**
  - **smectite-rich (49 wt%) bentonite which includes 7 wt% magnetite reported in Morocco**
  - **minor magnetite in the Otoy bentonite of California**
- **Micro-silica – essentially pumpable sand/micro-silica mix**
  - **Non-glacial loess (e.g. Chinese desert sand loess)**
- **No data exist, but potential sites could be assessed**

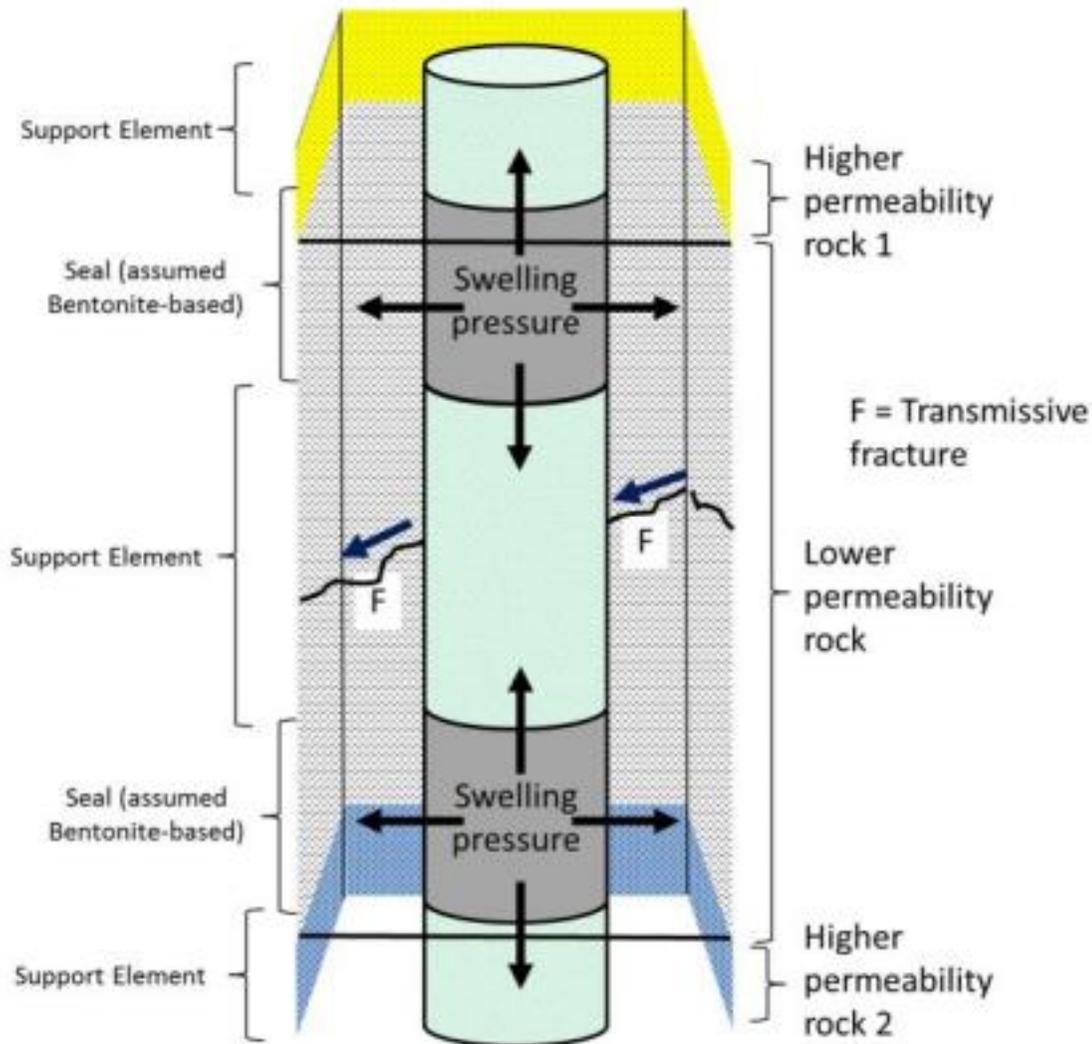
# Seals: alternative materials

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## ➤ Evaporites (bedded halites)

- In the German national programme, sealing boreholes not considered explicitly, but the reference safety assumption is that long-term creep of the host rock will ensure that all underground openings will eventually close, thus restoring the original barriers
- While actual analogue data are not used *per se*, information from observations in the tunnels (e.g. Navarro, 2013) have been used to assess the creep models
- Wolf & Noseck (2015) also noted that, **due to the importance of crushed rock salt in the repository design, it was essential to find appropriate NA support for the laboratory data that has been the basis of the safety case.** But this is not referring to borehole seals, rather to shafts and drifts.

# Seal supports



**Schematic illustration of a section of borehole showing an arrangement of seals and support elements.**

**Seals are located within the lower permeability rock. A support element is placed across a high transmissivity feature within the low permeability rock and in the adjacent higher permeability rocks**

# Seal support materials

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- **Much of the information relevant to seal supports has already been covered, but additional points include:**
  - **mechanical stability**
  - **chemical interactions with adjacent clay seals**
  - **seal support/host rock interaction**
- **Cementitious materials**
  - **See talk on cements from this morning, many of the points are completely applicable here. For example, quantitative evidence for porosity sealing would be advantageous**
  - **Active tectonic regimes where seal disruption could be possible could be assessed with sites in Jordan**

# Cement seal support/host rock interaction

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- In general, this reaction will be independent of the site hydrochemistry, but clearly there is a greater chance of minimising reaction with the host rock when either **high bicarbonate or high sulphate groundwaters** are considered as the **carbonation** or production of **ettringite** could block the outer pores of the cement and lead to effective minimisation of the degree of leaching
- However, the overall evolution of any borehole support-host rock annulus will also be coupled with other site aspects, such as the regional and local hydrology, host rock mineralogy and site stability
- In addition, when looking borehole seal supports, there is a question of **mass balance** when considering the relatively small volumes of cement in comparison with the much larger volume of host rock
- **Generally, this would suggest localised rock reaction of a limited extent**
- **A more detailed examination of the natural cements in Jordan**

# Seal supports: alternative materials

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## ➤ Baryte

- Commonly used in the oil and gas industry
- Baryte-bentonite reaction could occur, sites to examine it identified in Greece and USA
- Baryte-cement reaction reported in the lab, caused cement destabilisation. Jordan data indicate possible stability
- No data exist, but potential sites could be assessed

# Conclusions

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- **The Borehole Sealing Project has produced several generic seal and seal support designs for RWM's generic host rocks**
- **The designs are reasonable and our modelling and short-term lab experiments indicate that they can be emplaced appropriately and perform as required. Phase 3 project will include large-scale laboratory experiments and, potentially, in situ borehole sealing test**
- **Nevertheless, there are certain open questions where information unique to natural analogues – e.g. material longevity, including the impact of long-term reaction with groundwaters and other materials (host rocks, borehole liners etc) - can build confidence in the long-term behaviour and performance of borehole seals**
- **BUT little or no NA studies focussed on borehole seals**
- **But new NA sites/sample re-analysis identified for most aspects of potential interest**

# Acknowledgements

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- **The work presented here was funded by RWM Ltd (the UK radioactive waste management agency) as part of the project ‘Sealing of deep site investigation boreholes: Phase 2’**
- **Project was undertaken by a team comprising of:**
  - **Amec Foster Wheeler (UK)**
  - **Bedrock Geosciences (CH)**
  - **Clay Technology (SE)**
  - **Galson Sciences (UK)**
  - **Nagra (CH)**
  - **Schlumberger (UK)**
  - **Quintessa (UK).**

# References and further reading

- Alexander, W.R. (2017). Sealing site investigation boreholes: Phase 2. The use of natural, industrial and archaeological analogues in support of the borehole sealing project, Amec Foster Wheeler report 202580/07 for RWM Ltd, Harwell, UK (*in prep*).
- Alexander, W.R. (2017). More realistic treatment of long-term cement degradation: support from ancient natural cements. Proc. NAWG-15 Workshop, Prague, 22-25 May, 2017. SURAO Technical Report 2017-XX, SURAO, Prague, Czech Republic (*in prep*).
- W.R.Alexander and A.E.Milodowski (eds) (2014). Cyprus Natural Analogue Project (CNAP) Phase IV Final Report. Posiva Working Report WR 2014-02, Posiva, Eurajoki, Finland.
- Alexander, W.R., Reijonen, H.M., MacKinnon, G., Milodowski, A.E., Pitty, A.F. & Siathas, A. (2017). Assessing the long-term behaviour of the industrial bentonites employed in a repository for radioactive wastes by studying natural bentonites in the field. Geosciences Special Issue on Geological Disposal of High Level Radioactive Waste - The Relationship between Engineered and Natural Barriers. Geosciences Special Issue on Geological Disposal of High Level Radioactive Waste - The Relationship between Engineered and Natural Barriers, eds. R.Lunn, S.Harley, S.Norris & J.Martinez-Frias. Geosciences 7, 5; doi:10.3390/geosciences7010005
- Alexander, W.R., Börgesson, L., Hedström, M., Jefferies, N. and Wilson, J. (2017). Sealing Site Investigation Boreholes: Phase 2. Aspects of the evolution and longevity of bentonite seals. Amec Foster Wheeler Report 202580/09 for RWM Ltd, Harwell, UK (*in prep*).
- Fujii, N., Yamakawa, M., et al. (2015). Alkaline alteration processes of bentonic sediment in the Philippines natural analogue and perspective on the survey of the active type. In Alexander, W.R., Ruskeeniemi, T. and Reijonen, H.M. (eds) (2015). Proceedings (abstract book) of the NAWG-14 Workshop, Rauma, Finland, 9-11 June, 2015. Geological Survey of Finland (GTK) Guide 61. GTK, Espoo, Finland. [http://tupa.gtk.fi/julkaisu/opas/op\\_061.pdf](http://tupa.gtk.fi/julkaisu/opas/op_061.pdf)
- Fukushi, K., Sugiura, T., Morishita, T., Takahashi, Y., Hasebe, N. and Ito, H. (2010). Iron–bentonite interactions in the Kawasaki bentonite deposit, Zao area, Japan. Appl. Geochem., 25, 1120-1132.
- Jackson, C.P., Jefferies, N.L. et al. (2014). Sealing deep site investigation boreholes. Phase I report. AMEC Report 201257/002 Issue B for RWM Ltd, Harwell, UK.
- Jefferies, N. et al., (2017). Sealing Site Investigation Boreholes: Phase 2. Task 12: Final Report, Amec Foster Wheeler report 202580/14 for RWM Ltd, Harwell, UK (*in prep*).
- Kremer, E.P. & Alexander, W.R. (2015). Long-term durability of shaft sealing materials under highly saline groundwater conditions. In Alexander, W.R., Ruskeeniemi, T. and Reijonen, H.M. (eds) (2015). Proceedings of the NAWG-14 Workshop, Rauma, Finland, 9-11 June, 2015. Geological Survey of Finland (GTK) Guide No. 61. GTK, Espoo, Finland.
- Marcos, N. (2004). Results of the Studies on Bentonite Samples from Serrata de Nijar, Almería, Spain. Posiva Working Report 2004-24. Posiva, Olkiluoto, Finland.

# References and further reading

- W.M.Miller, W.R.Alexander, N.A.Chapman, I.G.McKinley & J.A.T.Smellie (2000). Geological disposal of radioactive wastes and natural analogues. Waste management series, vol. 2, Pergamon, Amsterdam, The Netherlands.
- Milodowski, A.E., Styles, M.T. & Hards, V.L. (2000). A natural analogue for copper waste canisters: The copper-uranium mineralised concretions in the Permian mudrocks of south Devon, United Kingdom. SKB Technical Report, TR-00-11, SKB, Stockholm, Sweden.
- Milodowski, A.M., Norris, S. and Alexander, W.R. (2016). Minimal alteration of montmorillonite following long-term reaction in natural alkali solutions: implications for geological disposal of radioactive waste. *Appl. Geochem.* 66, 184-197.
- Moyce, E.B.A., Rochelle, C.A., Morris, K., Milodowski, A.E., Chen, X., Thornton, S., Small, J.S. & Shaw, S. (2014). Rock alteration in alkaline cement waters over 15 years and its relevance to the geological disposal of nuclear waste. *Appl. Geochem.* 50, 91-105.
- Moyce, E.B.A., Milodowski, A.E., Morris, K. & Shaw, S. (2015). Herbert's Quarry, South Wales – an analogue for host-rock alteration at a cementitious radioactive waste repository? *Min. Mag.* 79, 1407 – 1418.
- Navarro, M. (2013). Die vereinfachte Berechnung der Konvergenzrate salzgrusverfüllter Hohlräume im Steinsalz. GRS Report GRS-307, GRS, Braunschweig, Germany. (in German).
- Pitty, A.F. and Alexander, W.R. (eds), (2011). A natural analogue study of cement buffered, hyperalkaline groundwaters and their interaction with a repository host rock IV: an examination of the Khushaym Matruk (central Jordan) and Maqarin (northern Jordan) sites. Bedrock Geosciences Technical Report 11-02, NDA-RWMD, Harwell, UK.
- Reijonen, H.M., Kurosawa, S., Ito, M. & Alexander, W.R. (2017). International Bentonite Longevity (IBL) project: an introduction. Proc. NAWG-15 Workshop, Prague, 22-25 May, 2017. SURAO Technical Report 2017-XX, SURAO, Prague, Czech Republic (*in prep*).
- Reijonen, H.M. and Alexander, W.R. (2015). Bentonite analogue research related to geological disposal of radioactive waste – current status and future outlook. *Swiss Journal of Geosciences* 108, 101-110. DOI 10.1007/s00015-015-0185-0
- Rozalen, M., Huertas, F.J. & Brady, P.J. (2009). Experimental study of the effect of pH and temperature on the kinetics of montmorillonite dissolution. *Geochim. Cosmochim. Acta* 73, 3752-3766.
- Techer, I., Khoury, H.N., Salameh, E., Rassineux, F., Claude, C., Clauer, N., Pagel, M., Lancelot, J., Hamelin, B. & Jacquot, E. (2006). Propagation of high-alkaline fluids in an argillaceous formation: case study of the Kyushaym Matruk natural analogue (central Jordan). *Journal of Geochemical Exploration* 90, 53-67.
- Wersin, P., Birgersson, M., Olsson, S., Karnland, O. & Snellman, M. (2008). Impact of corrosion-derived iron on the bentonite buffer within the KBS-3H disposal concept: The Olkiluoto site as case study. SKB Technical Report 08-34, SKB, Stockholm, Sweden.
- Wolf, J. and Noseck, U. (2015). Natural Analogues for containment providing barriers in rock salt: results from the German research project ISIBEL. *Swiss Journal of Geosciences*, 108, 129-138.