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(D3.1: Interpretation of matrix diffusion from real system analysis and experiments on different scales and D3.2: Natural chemical homologue behavior)

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Objective.

The objective of this deliverable is to supply all relevant background sources of the analytical and field pore water data, together with interpretations, from the Swedish site characterisation programme with a focus on matrix diffusion.

Background.

Two areas in Sweden have been studied to identify a suitable site for deep geological disposal of nuclear spent fuel waste. Both sites, Forsmark and Laxemar, are located close to the Baltic Sea coast and the hydrogeochemistry is documented in Laaksoharju et al. (2008, 2009). Several groundwater types which are now present in the bedrock can be associated with past climatic events in the late Pleistocene, including interglaciations, glaciations, deglaciations, and associated shore level displacements in connection with marine/non-marine transgressions and regressions. Among these, the last glaciation and post glacial period is the most important for the groundwater development in the Fennoscandian Shield. Generally, to varying degrees, both sites have been subjected to the same palaeoclimatic conditions following the last glaciation, i.e. since the last deglaciation (18,000-8000 BC). The climatic changes and resulting different groundwater types introduced into the bedrock since the last deglaciation stage to present conditions are well documented. In chronological order the most important are dilute glacial meltwaters, brackish waters (Littorina/Baltic Sea) and recent fresh waters. These successive water intrusions have interacted with the resident porewaters which can be considered an archive of the past hydrogeological (and therefore hydrogeochemical) history at the Forsmark and Laxemar sites.

The quantitative interpretation of the porewater-fracture groundwater interaction as a function of time is complex and depends on many factors such as the transport properties in the rock matrix, the distance to the nearest water conducting fracture, and the time period of fracture groundwater circulation with constant chemical and isotopic conditions, etc. (Waber et al., 2008, 2009; Waber and Smellie, 2009, and publications therein). Most demanding in such an interpretation is the case if a transient state (i.e. a difference in the chemical and isotopic composition between porewater and fracture groundwater) is established because of unknown conditions at the start of the interaction (initial conditions). In the situation of a steady state, on the other hand, at least a minimum and maximum time of interaction can be deduced more easily. In both cases, however, changes in the boundary conditions (i.e. the fracture groundwater composition) may become masked and superimposed in the course of the interaction. Furthermore, changes in the boundary conditions might not be equally present for all components. For example, the Cl^- concentration in fracture groundwater might grossly change with time while the water stable isotope composition remains similar as, for example, in the case of Littorina and Baltic Sea water, or the Cl^- concentration might remain similar whereas the water isotope composition changes dramatically as, for example, in the case of present day meteoric and past glacial meltwater infiltration.

The calculation of the concentration of chloride (or any other chemically conservative element) in the porewater (from out-diffusion concentrations) is inversely proportional to water content in the rock sample in question. The uncertainty of the indirectly derived porewater concentrations thus strongly depends on the accuracy of the water content determination and the degree to which the measured values represent *in-situ* conditions. The reliability of this method is addressed in Waber and Smellie (2008) and Waber et al. (2008, 2009, 2011).

The Forsmark Site.

The Forsmark site hydrogeologically can be subdivided into the ‘footwall’ (apart from the upper 150 m the site is basically of low transmissivity and a low frequency of single discrete water conducting fractures) and the ‘hanging wall’ (highly transmissive dynamic flow system to about 500 m and a high frequency of water conducting fractures including large subvertical deformation zones). These two hydraulic regimes reflect a different palaeohydrogeological evolution.

Generally at Forsmark, depending on the distance to the nearest water conducting fracture and the depth of the rock sample, the porewater preserves signatures of exchange with fracture groundwaters during Holocene, Pleistocene and pre-Pleistocene times (Waber et al., 2008). Furthermore, solute transport in the intact rock matrix appears to be dominated by diffusion, and matrix diffusion was identified to occur at least over several decametres into the rock matrix. Experimentally derived average pore diffusion coefficients for Cl^- are: metagranite to granodiorite = $1.2x10^{-10}$ m^2/s ± $0.40x10^{-10}$ m^2/s (n = 21), granodiorite to tonalite = $8.1x10^{-11}$ m^2/s (n = 1), aplitic granite = $9.4x10^{-11}$ m^2/s ± $4.2x10^{-12}$ m^2/s and fine-grained granite = $1.1x10^{-10}$ m^2/s ± $0.38x10^{-10}$ m^2/s (n = 3) at a temperature of 25 °C.

The footwall.

With respect to the porewater, the following characteristics can be noted:

- In the porewaters occurring locally in the shallowest levels down to about 200 m depth, only a weak influence of Holocene time fracture groundwater is developed that can be associated to the exchange with brackish marine Littorina type fracture groundwater.
- At intermediate depths below 200 m, porewater is dilute to moderately brackish in composition with stable isotope signatures depleted in the heavy isotopes down to about 640 m depth. A transient state with respect to higher mineralised fracture groundwater is established. These porewaters have evolved from pre-Pleistocene meteoric to brackish fracture groundwater of warm climate origin and support the very long average residence time derived for some groundwaters from this area.
- At the greatest depths sampled (>700 m), the origin of the saline Ca-Na-Cl type porewater cannot be related to any fracture groundwater because of the absence of such data. Their origin is, however, even older than that of the brackish type porewaters at intermediate depths.
- In conclusion, the porewater composition of the footwall bedrock suggests an evolution with a well developed component of rock-water interaction in a weakly active hydraulic system (mainly limited to the shallow zone) at least during Holocene and Pleistocene times.

The hanging wall.

- Down to about 700 m, the porewater is generally dilute and has characteristic signatures of interaction with glacial and brackish marine type fracture groundwaters. Compositional changes are related to the fracture frequency (mainly deformation zones ZFMF1 and ZFMA2) and do not show a regular distribution with depth.
- The porewater composition indicates that the system became essentially saturated with dilute glacial type water after the last glaciation (Weichselian) and such cold climate glacial water was circulating for a considerable time period in the fractures down to more than 500 m depth. Since the last deglaciation, this cold climate porewater signature has become overprinted with a brackish marine Littorina type signature as indicated by Cl^-, Mg^{2+} and $\delta^{18}O$ in porewaters sampled closer to the conducting fracture.

- In the shallow zone of the hanging wall bedrock, the brackish marine Littorina and/or Baltic Sea signature is now becoming overprinted by the circulation of present day meteoric groundwaters.
- In conclusion, the porewater is dominated by rapid exchange with fracture groundwater within a few thousands of years in a hydraulically very active system.

The Laxemar site.

The regional groundwater flow in the Laxemar subarea, in contrast with Forsmark, is strongly driven by topography with a general gradient from the high elevated areas in the west to the Baltic Sea in the east. The flow pattern is largely governed by the mutual connections of the deformation zones which characterise the region. The topography also results in localised areas of recharge/discharge which represent groundwater circulation cells of varying depth and extent, and therefore of varying ages. The maximum effect of groundwater circulation is considered to be down to depths of around 1,000 m.

In common with Forsmark, depending on the distance to the nearest water conducting fracture and the depth of the rock sample, the porewater at the Laxemar site preserves signatures of exchange with fracture groundwaters during Holocene, Pleistocene and pre-Pleistocene times (Waber et al., 2009). However, the Littorina Sea signatures are much weaker because of greater topography and hydraulic gradients resulting in a diluted Littorina end-member water which was limited in the extent of infiltrating into the bedrock. Furthermore, last deglaciation meltwaters and present meteoric groundwaters extend to much greater depths than at Forsmark because the bedrock is more fractured and hydraulically transmissive.

Experimentally derived average pore diffusion coefficients for Cl^- are: Ävro granite = 5.8×10^{-11} m^2/s ± 2.7×10^{-11} m^2/s, quartz monzodiorite = 8.4×10^{-11} m^2/s ± 5.5×10^{-11} m^2/s, diorite = 3.8×10^{-11} m^2/s ± 4.1×10^{-12} m^2/s (n = 2) at a temperature of 25 °C.

With respect to the porewater, the following characteristics can be noted:

- Matrix diffusion was identified to occur at least over several decametres into the rock matrix.
- There is some evidence of warmer climate meteoric influences from temperature maximums during the Pleistocene prior to the last glaciation.
- Down to depths of about 400 m, the porewater is of a dilute $Na\text{-}HCO_3$ type and present-day meteoric influence dominates in porewaters.
- Cold climate influence from the last glaciation occurs in the still dilute $Na\text{-}HCO_3$ type porewaters between about 135-350 m depth and sometimes down to about 500 m depth.
- A distinct change in chemical and isotopic composition to a highly mineralised $Na\text{-}Ca\text{-}SO_4$ and $Ca\text{-}Na\text{-}SO_4$ type porewater is observed between about 430-550 m depth and sometimes to 600-750 m depth. One possibility is that these signatures may have evolved from fracture groundwaters influenced by permafrost-related freeze-out processes.
- Modelling of a porewater profile extending from a conducting fracture into the intact rock matrix (Waber et al., 2007, 2009) indicated that changes in fracture groundwater composition during Holocene time left their (superimposed) signatures a few metres into the rock matrix. Further into the rock matrix, older (i.e. prior to the last glaciation), warm climate signatures are still preserved, lending additional support to the hydrogeochemical conceptual model.

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