



The Hydrochemical and Mineralogical Impact of Glaciation on Deep Groundwater Systems

Antoni E Milodowski, Martin, R Gillespie, Richard Metcalfe,
Mark B Crawford

British Geological Survey, Keyworth, Notts, NG12 5GG, U.K.

A thorough knowledge of the palaeohydrogeological conditions is required at potential sites for the disposal of radioactive wastes. This knowledge is an input for understanding the fundamental controls on groundwater flow and chemistry, and constructing models for the future hydrogeological evolution of a site. Most of the sites presently being evaluated for their suitability to host radioactive waste repositories have been affected by Pleistocene glaciations. There is a need to understand the impact of these glaciations on deep groundwater systems. A systematic, integrated investigation of rock mineralogy (especially fracture mineralogy) and groundwater chemistry can shed light on these impacts.

At Sellafield in northwest England, Nirex is investigating the suitability of Ordovician volcanic basement rocks, belonging to the Borrowdale Volcanic Group (BVG), as potential hosts for a repository of solid, low and intermediate-level radioactive waste (Nirex, 1994). The potential repository volume (PRV) is located at the margin of the East Irish Sea Basin (EISB), c. 3 km to the east of the present-day coast. The PRV lies within the upper BVG, at a depth of c. 650 m to c. 900 m below sea level (Ordnance Datum, bOD). The EISB is underlain by Carboniferous and older rocks, and infilled mainly with Permo-Triassic sediments, including halite-bearing, Triassic evaporites. The Lake District Massif lies to the east and consists of Lower Palaeozoic basement rocks, including meta-sedimentary rocks, granites and the BVG. In the neighbourhood of the PRV, Carboniferous to Triassic rocks overlay the BVG. During the Pleistocene, the area was glaciated on several occasions, resulting in variations to recharge conditions, and fluctuations in sea levels of several tens of metres. An important aim of a detailed site characterisation programme has been to establish the impact of the glaciations on the deep groundwater system. A major approach has been to undertake extensive, integrated investigations of rock mineralogy (including studies of fractures and matrix) and groundwater chemistry. These studies have used core and water samples from deep (to c. 2 km) boreholes, at 22 localities across the Sellafield area.

These investigations have revealed two main types of water: a. deep, Na-Cl dominated saline waters and brines, occurring in the cover and basement near the coast, but entirely in the basement further east; and b. shallow, fresh Ca-HCO₃ and Na-Ca-HCO₃ dominated waters, occurring mostly in the sedimentary cover. The salinities of the Na-Cl waters range from up to c. 30000 mg l⁻¹ Total Dissolved Solids (TDS) in the PRV, to c. 180000 mg l⁻¹ near the coast, 3 km to the west. The saline transition zone (STZ) between the fresh and saline waters varies in elevation across the area. The STZ falls from c. 400 m bOD near the coast to c. 650 m bOD 1.5 km inland, before rising to c. 300 m bOD in the vicinity of the PRV, and c. 150 m bOD 3 km to the east of the PRV.

The nature of the chemical variations in Na, Br and Cl indicates that the Na-Cl waters gained most of their salinity by halite dissolution. However, there are subtle variations in the chemistry of Na-Cl waters. In these, mass ratios of Br/Cl increase from west to east, from about 0.001 near the coast, to about 0.002, c. 3 km inland. Mass ratios of Na/Ca decrease over the same section, from about 30 to about 10. The isotopic signatures of saline waters in the east ($\delta^{18}\text{O}_{\text{SMOW}} = -7$ to -8‰ ; $\delta^2\text{H}_{\text{SMOW}} = -42$ to -46‰) are lower than in fresh waters ($\delta^{18}\text{O}_{\text{SMOW}} = -6$ to -7‰ ; $\delta^2\text{H}_{\text{SMOW}} = -35$ to -40‰), and in western brines ($\delta^{18}\text{O}_{\text{SMOW}} = -5$ to -6‰ ; $\delta^2\text{H}_{\text{SMOW}} = -32$ to -44‰). Preliminary $^{36}\text{Cl}/\text{Cl}$ ratios for Na-Cl groundwaters range from 2×10^{-15} (in the western cover), to 27×10^{-15} (in the PRV), consistent with ^{36}Cl being equilibrated

with the neutron flux in the Permo-Triassic sandstones and BVG respectively.

An explanation for these results is that basinal brines with an evaporite-derived salinity migrated downwards into the basement. The ^{36}Cl variations are consistent with these waters residing in the basement in the east for a substantial period of time (possibly > 1.5 Ma). During this interval, water/rock interactions in the basement are interpreted to have caused the observed variations in parameters such as Br/Cl and Na/Ca ratios. Subsequently, during the Pleistocene, cool, meteoric water recharged from the east, diluted the brines, and depleted them in heavy isotopes relative to the western brines. Most recently, meteoric waters with heavier isotopic compositions than the Pleistocene waters, have recharged the shallower rocks, interacted with carbonate minerals and produced bicarbonate-rich, shallow waters.

This scenario raises a number of questions: when did the cool recharge waters enter the deep system relative to the Pleistocene glacial episodes? where did the Pleistocene waters enter the deep groundwater system? what were the maximum depths to which these waters penetrated? and what was the effect of these waters on the geometry and flow paths of the deep Na-Cl dominated groundwaters? Detailed, coupled hydrogeochemical-mineralogical investigations have the potential to answer these questions.

Our mineralogical investigations have shown that geologically late Fe- and Mn-oxyhydroxide dominated fracture mineralisation extends to different depths in different boreholes across the Sellafield area. The base of this mineralisation corresponds approximately to the present depth of the STZ, between the bicarbonate and Na-Cl waters. Similarly, a geologically late generation of calcite in flowing features has been shown to vary in morphology, from 'nailhead' through 'equant', to 'scalenohedral' crystal forms across the modern STZ. An explanation for these correlations between mineralogy and water chemistry is that the distribution of oxide-dominated mineralisation reflects the maximum depth of penetration of oxidising recharge water, while the morphological variations in the calcite are controlled by the chemistry (possibly Mg, SO_4 , Sr or TDS concentrations) of the groundwaters. Geochemical modelling confirms that precipitation of calcite will occur due to mixing between deep, calcite-saturated saline waters and shallower, fresh HCO_3 waters. Hematite will precipitate with decreasing temperature accompanying this mixing. By dating the calcite (using U-series isotopes and ^{14}C) and oxide mineralisation (using K-Ar) it should be possible to constrain the stability of the STZ and the controls upon it exerted by glaciation. This work has demonstrated a potentially powerful mineralogical tool for placing limits on the impact of glaciations on deep groundwater systems.

References

Nirex 1994. The geology and hydrogeology of Sellafield, Volume 3: The hydrogeology. Nirex Report 524.