The Preservation of Climate-Induced Chemical Signatures in Groundwater. A Feasibility Study in the Context of the Sellafield Site, UK.

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UK Nirex Ltd have identified Sellafield, West Cumbria as a potential site for the deep disposal of solid low and intermediate level radioactive waste for which a post-closure safety case is now being developed. If, following further geological site investigation and related research, Nirex decide that they have sufficient evidence to support such a case, it is anticipated that they will apply formally to Her Majesty's Inspectorate of Pollution (HMIP) and the other Authorising Departments, for an authorisation to dispose of radioactive wastes. This paper focuses on the geochemical aspects of multidisciplinary feasibility study aimed at extending HMIP's ability to assess models of past groundwater evolution and to provide a scientific basis for the evaluation of future climate changes on post-closure performance of a potential repository at Sellafield. An overview of the approach, which incorporated modelling of climate change, groundwater flow and groundwater chemistry is given in an accompanying paper (Boulton *et al.*, this issue).

Geochemical signatures provide information extending back over tens of thousands or, more rarely, millions of years and represent key data against which regional long-term groundwater flow models can be tested. Importantly, scope exists for alternative conceptualisations of a system to be formulated whereby the implications of different chemical signatures are compared. Considerable uncertainty remains, however, in the definition of initial recharge signatures during past climates and also regarding the preservation of these signatures in specific groundwater regimes. Both these aspects have been examined in the context of the Sellafield Region.

The use of ^{2}H and ^{18}O signatures in constraining groundwater ages relies upon the fact that the isotopic signal of recharge water is dependent on local climatic influences such as temperature, latitude and altitude. A methodology has been developed for deriving the isotopic signature of recharge waters through time, incorporating advection of precipitation through an ice sheet. Taking reasonable parameters for groundwater flow rates and dispersivities the preservation of a Sellafield-specific ^{18}O recharge signal has been examined for specific rock types. The results demonstrate that signal preservation of a non-reactive recharge signal may be poor in the Ordovician Borrowdale Volcanic Group (BVG) (Figure 1). Results for the overlying St Bees Sandstone suggest good signal preservation, although the strongly depleted $\delta^{18}O$ (‰ SMOW) values predicted have not been recorded in Sellafield groundwaters. This suggests that alternative conceptual models for glacial recharge will require consideration. Moreover, it demonstrates the feasibility of using palaeohydrogeological techniques to constrain models of long-term groundwater flow.

Noble gas (Ne, Ar, Kr, Xe) measurements in groundwater have been successfully used in number of palaeohydrogeology studies. The principal of the noble gas thermometer (NGT) is that the solubility of noble gases in water is temperature dependent, thus, the temperature at which water equilibrated with the atmosphere, ie at recharge, can be calculated to approximately ±2°C. Noble gas solubilities, however, decrease exponentially with increasing salinity, although few groundwater studies have yet evaluated this effect. At the Gorleben site in Germany anomalously low NGTs were explained by exsolution and noble gas concentration in association with an aquitard Suckow & Sonntag (1993). In the context of Sellafield, comparison of groundwater salinities with NGT determinations suggests a controlling relationship may be operative (Figure 2). In a regionally dipping aquifer, noble gases exsolved due to salinity may be unconfined and thus escape, leading to anomalously high NGT determinations in groundwaters. Preliminary calculations suggest that at salinities of 1 mol dm^{-3} NaCl, salinity-corrected NGTs are in the order of 8 °C to 9 °C lower than uncorrected values such that uncorrected warm recharge temperatures may in fact reflect cool recharge. Thus, depending on the openness of the groundwater system to gas migration, saline exsolution may potentially give rise to either anomalously low or high NGTs. These effects should be considered in palaeohydrogeological studies in saline groundwaters.

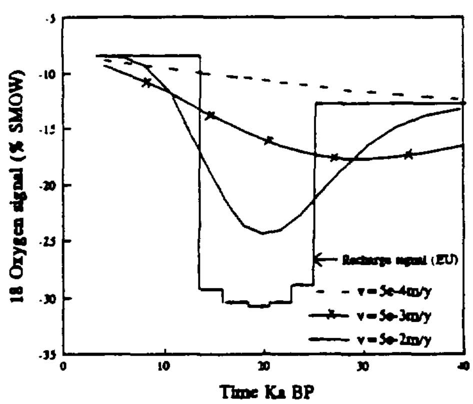

Figure 1
Comparison of $\delta^{18}O$ recharge signals to model groundwater signals in the BVG for varied porewater velocities and a dispersion length of 30m.

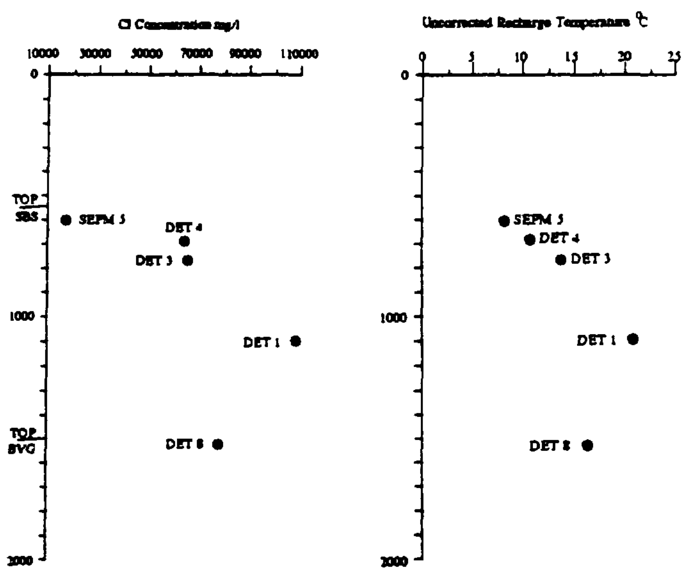

Figure 2
Comparison of salinity and uncorrected NGT profiles in Sellafield borehole 3. Adapted from Nirex Report 524. A salinity control on NGTs is suggested by the similarity of the profiles.

Boulton, G S, (1996). Modelling the signal of long-term environmental change in the geochemistry of groundwater - an approach to testing models of long-term rock mass properties (this issue).

Suckow, A and Sonntag, C, 1993. The influence of salt on the noble gas thermometer. In Proc. Meeting on Isotopes of Noble Gases as Tracers in Environmental Studies IAEA Vienna, pp, 307-318.