Loch Lomond (Scotland)

Description: A well known natural analogue study of diffusive transport within clay sediments was that performed at Loch Lomond in the southern Highlands of Scotland (MacKenzie et al., 1983; 1984; 1990; Hooker et al., 1985; Falck and Hooker, 1990). Although the Loch Lomond study (which ended about 1990) involved the examination of only a few sediment cores from the loch, much valuable, quantitative information was obtained. At present Loch Lomond is landlocked and therefore freshwater, but about 7000 years ago the Flandrian marine transgression from the Firth of Clyde resulted in an incursion of seawater into the loch. This event is clearly recorded in the Loch Lomond sediment deposits which are about 6 m deep and contain a layer of marine detritus, about 1 m thick, overlain and underlain by freshwater sediments (Figure 1).

All the sediments are clay-rich, containing up to 80% clay in some horizons. A distinctive porewater chemistry characterises the marine sediment band, with higher measured concentrations of chloride, bromide and iodide. Migration of these halogens into the porewaters of the freshwater sediments above and below the marine layer has left a long record of diffusive transport.

A number of depth-concentration profiles were constructed for various elements from the analysis of the Loch Lomond sediment core samples, and these were used to quantify the diffusion processes. The sedimentation of the marine band was found to have occurred between 5400 and 6900 years ago, based on data from radiocarbon analysis and palaeomagnetic and palynological studies. Fixation of iodine, bromine, uranium and ^{226}Ra was clearly identifiable within the marine layer, a feature that correlated with the presence of organic carbon.

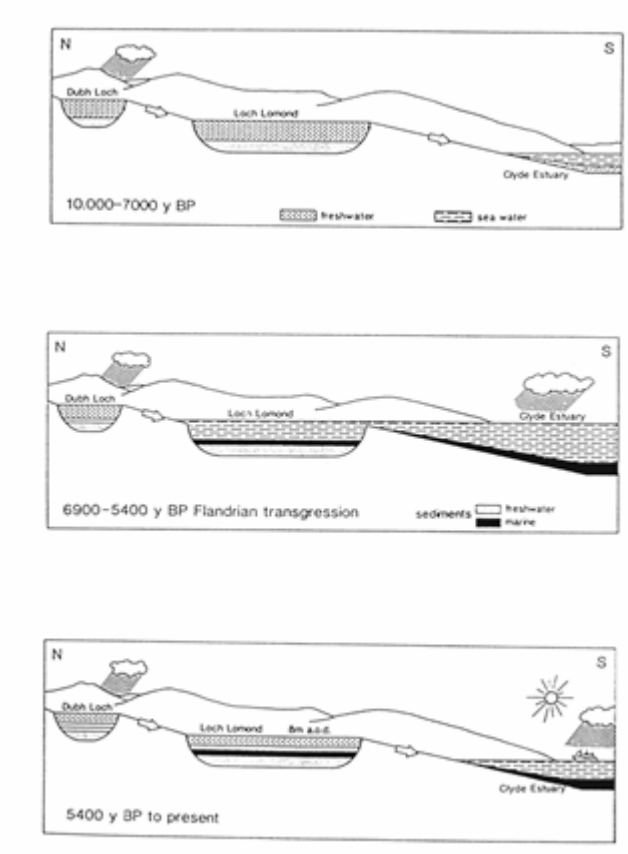

Figure 1 Schematic representation of the formation of the marine band within Loch Lomond during the Flandrian marine transgression 6900 to 5400 years ago (from McKinley, 1989).

Another core was taken from the Dubh Loch which lies just to the east of Loch Lomond, but at a higher elevation, and which did not experience the marine transgression. Analysis of the Dubh Loch core has shown that the effects observed in the Loch Lomond cores were indeed due to the marine transgression and not to a possible change in composition of eroded material supplied from the surrounding catchment.

The sediment porewaters are reducing in nature with low Eh values. The porewater concentrations of chlorine, bromine and iodine decrease with distance from the marine sediments, as shown in Figure 2 for the case of bromine. Since the last marine sediments were laid down 5400 years ago, bromine has slowly diffused upwards and downwards into the enclosing freshwater sediments (Hooker et al., 1985). Modelling the bromine concentration profile in the porewater according to simple diffusion theory with reversible sorption and no advective flow or transport, produced an effective diffusion coefficient of 3×10^{-11} m^2s^{-1} (Hooker et al., 1985). This value was an order of magnitude lower than estimates based on laboratory sorption experiments, suggesting some mechanism of retardation of bromine was occurring in-situ. Subsequent and more detailed modelling assumed that halide interactions were occurring with organic matter in the sediments and that a slow rate of fixation (requiring several decades) was probably retarding bromine and iodine in the organic matter of the upper freshwater sediments. These interactions complicate calculation of the diffusion-dominated migration rates (Falck and Hooker, 1990).

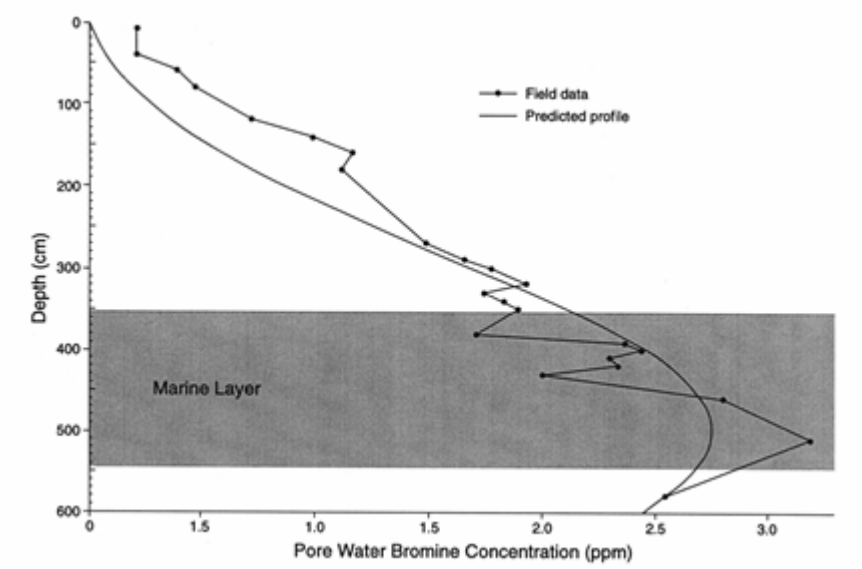

Figure 2 Measured and modelled (assuming an initial bromine porewater concentration of 10 ppm) bromine profiles through the Loch Lomond sediment core (from Hooker et al., 1985).

The studies of the Loch Lomond sediment cores have demonstrated that the sediments have remained an effective sink for bromine and iodine for thousands of years with only slow releases into the porewaters and slow diffusion towards the surface layers and the loch waters above.

Relevance: The findings from this study are most useful when applied to argillaceous repository environments, and would include the diffusive transport of halides within a bentonite backfill. This well-understood evolution of a clay-rich sediment system provides good illustrative analogue evidence for demonstrating the slow diffusive movement of radionuclides such as ^{36}Cl and ^{129}I through saturated low permeability clay barriers.

Position(s) in the matrix tables: The Loch Lomond study would occupy the Bentonite Clay/Diffusion box of the near-field matrix table. It also occupies the Diffusion/Plastic clays box under Nuclide migration in rock at low temperatures (<100 ^oC) in the Geosphere matrix table.

Limitations: The near-surface saturated loch sediments are not the same as bentonite material in a repository backfill environment nor are they the same as a host clay rock. The diffusion and inferred organic matter-halide interaction processes have been occurring at low temperatures (5-10 ^oC). Deeper formations would have higher temperatures (due to the geothermal gradient) which would increase any diffusion coefficients in the clays and speed up any water-rock interactions.

Quantitative information: The quantitative information from the study includes an estimate of an effective diffusion coefficient of 3 x 10^{-11} m^2s^{-1} for bromide in saturated clay-rich sediments. Although this result is most useful when applied to the diffusive transport of halides within a bentonite backfill, the information would also be appropriate to modelling matrix diffusion of ^{36}Cl and ^{129}I in a fractured clay rock with flowing fractures in the far-field.

Uncertainties: There are uncertainties associated with the compositions of the interstitial porewaters 5400 years ago i.e. the source term concentrations of the dissolved halides. The exact characteristics of the implied retardation interactions between the sediment organic matter and the migrating bromide and iodide anions are not known. Chlorine migration was assumed to be conservative i.e. chloride anions diffused with no sediment interactions.

Time-scale: The time-scale of the Loch Lomond natural analogue is geological i.e. Quaternary (Holocene <10,000 years). In particular, the processes of sedimentation and diffusion have occurred over the last 7000 years.

PA/safety case applications: Previous uses of the analogue study in a PA or safety case may be in published PA reports by Nagra.

Communication applications: Nagra used the Loch Lomond study in the analogue video. It would be interesting to know how the video has been used in communication and the reactions of the video viewers.

Current use of the analogue study in communication and dialogue include:
At http://www.nuclearfaq.ca/cnf_sectionE.htm , a Canadian website set up to answer frequently asked questions in the nuclear and radioactive waste management field, the Loch Lomond study is considered an important analogue for the 'protective clay buffer' of a deep spent fuel repository i.e. with reference to the potential isolating capacity of the buffer. Reference to a clay buffer rather than backfill seems odd when the temperature of a buffer system would be so much greater than the ambient temperature of the loch sediments. This is a small, but good example of the inappropriate use of the Loch Lomond analogue information, since the technical i.e. temperature implications had not been fully appreciated before associating the loch sediment environment with that of a buffer.

References:

Falck WE and Hooker PJ (1990) Quantitative interpretation of Cl, Br and I porewater concentration profiles in lake sediments of Loch Lomond, Scotland. BGS Technical Report, WE/90/3.

Hooker PJ, MacKenzie AB, Scott RD, Ridgway I, McKinley IG and West JM (1985) A study of natural and long term (10^3 - 10^4 year) elemental migration in saturated clays and sediments, part III. BGS Technical Report, FLPU 85-9; also CEC Radioactive Waste Management Series, EUR 10788/2, CEC, Luxembourg.

MacKenzie AB, Scott RD, McKinley IG and West JM (1983) A study of long term (10^3 - 10^4 year) elemental migration in saturated clays and sediments. Institute of Geological Sciences Technical Report, FLPU 83-6.

MacKenzie AB, Scott RD, Ridgway IM, McKinley IG and West JM (1984) A study of long term (10^3 - 10^4 year) elemental migration in saturated clays and sediments, part II. BGS Technical Report, FLPU 84-11; also CEC Radioactive Waste Management Series EUR 10788/1, CEC, Luxembourg.

MacKenzie AB, Shimmield TM, Scott RD, Davidson CM and Hooker PJ (1990) Chloride bromide and iodide distributions in Loch Lomond sediment interstitial water. BGS Technical Report, WE/90/2.

McKinley IG (1989) Applying natural analogues in predictive performance assessment. Unpublished internal Nagra report, Nagra, Wettingen, Switzerland.

Added value comments: The analogue information on the retardation aspects e.g. the very slow releases of Br and I over time from the marine band, could be extended to the fixation of ^{226}Ra for the last 5,000 years. The retardation information could be used in an illustrative way to show how effective relatively undisturbed shallow clay-rich sediments can be as barriers to elemental diffusive transport and how deeper, thicker clays would afford an even greater degree of efficiency as barriers to the migration of radionuclides.

Potential follow-up work: Possible follow-up work would involve the collection of fresh samples and an analysis and elucidation of the probable retardation interactions that have occurred between the sediment organic matter and the bromine and iodine elements.

Keywords: clay, sediments, halides, diffusion, backfill

Reviewers and dates: Paul Hooker, Enviros Consulting (July, 2003)