Geochemistry and radioactive waste management: issues and conflicts

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Introduction The multi-barrier representation of a radioactive waste disposal system has wide currency in the international community and is conventionally considered in terms of: 1) the wasteform itself; 2) the engineered barrier; 3) protection afforded by the host geology; 4) the potential for dispersion of activity in the surface environment

Geochemical considerations are not confined to the geological barrier but influence all aspects of containment from performance of grouts and backfill to the bioavailability of contaminants in the soil zone. As with related scientific disciplines, a procedure is required which can account explicitly for uncertainties in the effect of geochemical processes on system performance. These may arise from, *inter alia*, an incomplete understanding of the disposal system, the inherent variability of natural systems and the unavoidable subjectivity involved in choosing models for long-term predictions. This paper examines the problem of dealing with such uncertainties and illustrates the disparate and often conflicting approaches to treating geochemical information in performance assessments and conventional scientific investigations, respectively.

Performance assessment: uncertainty, subjectivity and bias Performance assessment procedures are being employed by several countries to demonstrate that a radioactive waste disposal facility will not give rise to unacceptable risks over timescales of many thousands of years. The complexity of the natural environment obviously necessitates a simplified representation in probabilistic risk assessment (pra) models which contain many approximations, assumptions and caveats. Consequently, procedures have been developed for treating uncertainty and bias throughout the various stages of an assessment. In this context, the following definitions apply:-

uncertainty reflects our incomplete knowledge of parameter values, both spatially and temporally (parametric uncertainty) together with deficiencies in our conceptualisation of the system (model uncertainty).

bias results from prejudicial selection of models, parameters or data owing to incomplete definition of the disposal system and/or limitations in calculation methods.

Identifying sources of uncertainty can often be facilitated by considering four distinct stages within the assessment; conceptual model development, selection of model parameters, selection of key data and mathematical modelling. Expert elicitation is frequently used for the first two of these, whereas selection of data and modelling, itself, are amenable to structured sensitivity analysis. Parametric uncertainty is normally incorporated into a risk assessment by using statistically defined distributions of parameter values. Conceptual model uncertainty is addressed in most countries by considering a range of possible 'scenarios', as discussed elsewhere.

Quantification of bias is more difficult and, here, the choice of conceptual model(s) can exert an over-riding influence on all subsequent calculations. Alternative conceptualisations of a system may have been overlooked even though they may be consistent with available information from a site. Relatively few attempts have been made to address this issue in the field of geochemistry but those studies which have been undertaken follow two distinct routes: 1) a top-down approach in which the results of predictive modelling are assessed by retrospectively analysing each component of the modelling procedure; 2) modular construction ie a 'bottom-up' approach.

Both involve a certain degree of subjectivity. In the case of the former, one may derive a false sense of security from apparent conformity in predicted and measured results when the actual processes operating and associated boundary conditions are not sufficiently well understood. Similarly, for the modular approach an assurance is needed that all of the necessary components of the system are represented and that their interactions are explicit. Thus, while the advantages of expert systems, for example, are readily apparent they may be negated if deployed in an uncritical manner.

Subjectivity is accepted as an integral part of performance assessment but is rarely acknowledged in traditional geochemical investigations. More commonly an individual researcher advocates his interpretation of observed features, often to the exclusion of alternatives. Without doubt, consideration of uncertainty and bias issues are implicit in his reasoning though the justification for selecting one conceptual model over another is not documented.

When incorporating geochemical concepts into post-closure assessments of radioactive waste disposal, however, the issue is not, if, but, at what stage, subjective judgements should be employed.

Conceptualisation of geochemical systems The tenet of the geological sciences 'the present is the key to the past' has been adopted enthusiastically by the waste disposal community and extended to the maxim 'the past is the key to the future'. Inherent in this statement is an appeal to the geosciences to provide assurance regarding the future performance of a natural system. Such assurance is far in excess of the demands normally imposed on earth scientists and tends not to reflect the provisional nature of geological understanding. Clearly,there are practical limits to the confidence which can be gained from either 'natural analogues' or 'predictive modelling'. Thus, rather than strive to 'validate' waste disposal concepts using these methods; an impossible task, a more realistic aim would be to employ models in testing plausible hypotheses of system evolution in a structured manner. One might, for example, make alternative selections of the processes thought to dominate, examine a variety of boundary conditions and re-assess interactions between the hydrogeology and geochemistry of the region.

As an illustration of the former, consider a clay-rich aquitard separating a body of saline water from fresh. If one postulates osmosis as a dominant mechanism, concentration gradients will develop where the transport of charged species is impeded. Thus, trace elements may accumulate at the interface, or within the aquitard, where groundwater salinities are high. If, however, the semi-permeable behaviour of the aquitard is neglected, as is usual in radionuclide transport calculations, purely diffusive gradients for individual ions will promote dilution. Each of these two situations represents a discrete conceptual model which may be analysed for its impact on risk.

By far the most contentious assumption in geochemical modelling is the attainment of equilibrium. It is generally accepted that many rock–water systems are not at equilibrium. Equally, it is recognised that reaction rate data are severely limited. Given that justification for the equilibrium approach is appropriate only when dealing with closed, non-dynamic systems the problem appears intractable. This may indeed be so at the quantitative level but, nevertheless, the bias introduced by neglecting kinetics can be assessed. Consider two extremes: 1)-reaction kinetics are very slow relative to groundwater flow rates, thereby allowing migrating species to be treated independently; 2) -reaction kinetics are sufficiently rapid that local equilibrium is attained.

These two extreme models were investigated in a published study of neptunium transport through glauconite-rich sand. The experiment, employed a column pre-equilibrated with a synthetic groundwater solution under oxic conditions and a ^{237}Np source. Groundwater was pumped through the column and the outflow monitored for 187 days. The results showed a leachate concentration almost two orders of magnitude less than the inflow with most of the injected Np concentrated in the first few centimetres of the sectioned core.

The experiments were simulated using a conventional chemical transport model incorporating an electrostatic description of the adsorption process. The first set of simulations assumed rapid adsorption kinetics and local equilibrium, so that the partitioning of aqueous Np species ($NpO_2CO_3^-$, NpO_2^+, NpO_2OH) throughout the column responds immediately to the effects of adsorption. Predicted concentrations of sorbed Np agreed well with observations close to the source but decreased more rapidly than actually observed with no breakthrough.

The second set of simulations assumed very slow kinetics, such that each transported species behaves independently. Predicted concentrations of sorbed Np again fell rapidly but breakthrough similar to that measured could be explained by conservative transport of NpO_2OH accounting for about 3% of the source.

Thus, by ignoring the influence of reaction kinetics and assuming local equilibrium, no breakthrough was predicted whereas, in fact, a small but possibly significant concentration of Np did pass through the column. It is likely that the actual situation lay between these two extremes as rate constants for the interconversion of Np species are estimated to be of the order of a few hours.

Discussion This paper has briefly highlighted efforts made to distinguish between alternative models of geochemical behaviour. The column case examined the null hypothesis implicit in almost all geochemical modelling exercises, that the system is in thermodynamic equilibrium. Such approaches are still relatively rare. This serves to emphasise the need for formalised model and data elicitation prior to performing numerical calculations, followed by critical peer review subsequent to completion.

One method suggested for reducing uncertainty and bias at the conceptualisation stage might function by posing a series of logical questions producing, if appropriate, several plausible alternative models plus their associated limitations. The probability of each conceptual model being substantially correct could then be quantified in terms of the amount and quality of data available to support it.