BIOPROTA: international collaboration in biosphere research for radioactive waste disposal

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ABSTRACT

Consideration of the biosphere is important in the post-closure safety assessment of a geological disposal facility (GDF) as the biosphere acts as the receptor for any contaminants that may be released from the geosphere. Considerable uncertainty exists in the characteristics of the biosphere at times in the far future when any contaminant releases from a GDF would reach the accessible environment. These uncertainties include human behaviour, affecting environmental change as well as exposure modes.

A number of critical scientific issues have been identified through the practical application of the International Atomic Energy Agency reference biosphere approach within both site generic and site specific repository assessment projects. These issues are being addressed through an international collaboration programme, BIOPROTA. The purpose of this paper is to describe the BIOPROTA programme, its objectives and typical working method. The approach is illustrated with examples from the recent work programme including model intercomparison studies for the radionuclides ³⁶Cl and ⁷⁹Se.

Keywords: biosphere research, geosphere-biosphere interface, dose assessment models, waste disposal.

Introduction

POST-CLOSURE safety assessment of geological disposal facilities (GDF) for radioactive waste is commonly divided into consideration of the effectiveness of near-field engineered barriers at containing radionuclides, the role of the geosphere in limiting the further migration of any releases from the near field, and the assessment of the radiological impact of any eventual release to the biosphere. Special difficulties arise in the latter aspect because the rates of environmental change in the biosphere are faster, and justification of assumptions for human

* E-mail: ks@eden-ne.co.uk DOI: 10.1180/minmag.2012.076.8.36 behaviour, affecting environmental change as well as exposure modes, is problematic.

Several projects have been undertaken internationally to improve the long term assessment basis. Among these, the International Atomic Energy Agency (IAEA) BIOMASS Theme 1 (International Atomic Energy Agency, 2003) provided a clear basis for identifying, justifying and describing biosphere systems. In addition, the development of conceptual and mathematical models has been set out and a protocol for the application of data to these models developed. However, practical application of this methodology within site-generic and site-specific assessments has identified a number of critical issues, largely associated with processes for the key long-lived radionuclides of relevance to deep GDFs. The international collaboration programme, BIOPROTA (www.bioprota.org), which was established in 2002, seeks to address these key uncertainties.

The purpose of this paper is to describe the BIOPROTA programme, its objectives and typical working method. The approach is illustrated with examples from the recent work programme including model intercomparison studies for the radionuclides ³⁶Cl and ⁷⁹Se.

The BIOPROTA collaborative forum

The BIOPROTA programme is an international collaborative research forum aimed at national authorities and agencies responsible for achieving safe and acceptable radioactive waste disposal. In 2012 there are 20 funding organizations in 14 countries from East Asia, Europe and North America, with additional participation from associated technical support organizations and research institutions.

The principal objective of the forum is to provide underpinning scientific support to, and improve confidence in, long-term safety assessments. This is achieved through the application of good science to address important uncertainties in order to avoid unnecessary conservatism through a graded and iterative approach to assessments. This includes the sharing of knowledge and experience of organizations with a direct interest in the safe disposal of radioactive waste, but also includes relevant experience and information arising from research and assessments in other fields, such as management of NORM (naturally occurring radioactive material) and legacy contaminated sites. The forum also works to develop links with other relevant international organizations and working groups such as the IAEA (International Atomic Energy Agency), ICRP (International Commission on Radiological Protection) and IUR (International Union of Radioecology).

The BIOPROTA programme aims to identify common issues relating to biosphere assessments to which suitable approaches can be developed to the mutual benefit of all involved organizations through the efficient use of skills and resources. Issues may include the need for greater understanding of the processes governing behaviour of radionuclides in the environment, lack of radioecological data to represent such processes and/or uncertainty around the conceptualization of key processes. Such issues may be addressed through exchange of information within focussed work-

shops and/or the establishment of multilateral research programmes to develop appropriate assumptions and formulations upon which features, events and processes (FEPs) within the biosphere system may be represented for important radionuclides and to evaluate models in their ability to represent FEPs. The BIOPROTA programme approaches to addressing issues can be broadly categorized in three forms: (1) development of multi-organization collaborative projects to compare different model representations of biosphere processes for key radionuclides (model intercomparison); (2) organization of international workshops on key uncertainty areas; and (3) reporting to ensure knowledge dissemination. Benefits from the application of these approaches are discussed in this paper, with examples provided from the most recent work programmes.

Addressing key biosphere modelling issues: intercomparison exercises

Key to dose assessments for many radionuclides is the appropriate representation of how radionuclides initially enter the human food chain (i.e. the behaviour in soils and uptake into plants). There are uncertainties associated with many of the long-lived radionuclides of interest to GDF assessments, which commonly result in the application of a cautious approach. When taken together for a set of parameters, a large number of cautious assumptions may lead to assessment results being unnecessarily conservative, giving rise to undue concerns about safety and inappropriate allocation of resources. Furthermore, by not making strenuous efforts to understand the science behind radionuclide behaviour, processes specifically relevant to radionuclides with unique characteristics may be omitted which may, under some circumstances, lead to dose underestimation. There is therefore a requirement to develop an adequate understanding of the system and what this means for radionuclide behaviour in order to generate broad confidence in assessment conclusions.

Soil–plant behaviour has been the focus of four collaborative model-intercomparison projects within BIOPROTA. These relate to the behaviour of carbon-14 (¹⁴C), chlorine-36 (³⁶Cl), selenium-79 (⁷⁹Se) and uranium-238 (²³⁸U) series radio-nuclides in soils and uptake into plants. The ⁷⁹Se and ³⁶Cl intercomparison projects are the focus herein. Information on the ¹⁴C intercomparison is

provided in a parallel paper (Smith et al., 2012) and in the project report (Limer et al., 2011). Results from the ²³⁸U series radionuclides project are reported in Limer et al. (2011). The broad approach taken to address assessment issues in each of these projects has been: (1) organization of focussed workshops to discuss the underlying science; (2) carrying out a FEP analysis, focussing on the soil-plant system, and, from this, developing a general conceptual model based on interactions between identified FEPs; (3) audit of available models against the FEP list; and (4) application of each model to a defined scenario with output being analysed as a means of identifying the key processes (and their mathematical representation) and/or parameters governing differences in results. Depending upon the results of the intercomparison exercise, further projects may be initiated to address remaining uncertainties. Some of the benefits associated with such an approach are demonstrated in relation to recent projects for which examples are provided.

Improving confidence in model approaches: the ³⁶Cl project

One of the objectives of undertaking model intercomparison exercises is to test the hypothesis that more complex (i.e. process-oriented) models give better results than conventional (radionuclide generic) models and this hypothesis was recently tested in a project focussed on the modelling of ³⁶Cl in soils and uptake into plants (Limer *et al.*, 2008). This isotope is of particular interest in respect of GDF's due to its long half-life (301,000 years), high mobility under many environmental conditions and, as a plant macronutrient, the possibility that it may be taken up in significant quantities resulting in a high accumulation potential in the food-chain.

Internationally there are a range of models available to represent the uptake of ³⁶Cl from soils to plants and these differ in their complexity, the processes incorporated and the mathematical representation of these processes. Ten different models were applied to a defined scenario within the BIOPROTA ³⁶Cl project. The range of processes considered in each model is listed in Table 1.

The model intercomparison indicated that, irrespective of the level of complexity of model, the processes included, or differences in data assumptions, the results were typically

Model			Conventional			Smarific artivity	Compartmentel	nento]	More	valumoo
MORT	EPRI	BIOMASS	1ASS JGC	Aquabios	Aquabios SAMM-TR	AquaCl36	SAMM-IR	EdF	IRSN	RSN NDA RWMD
Process										
Rainfall				×	×	×	×	×	×	×
Evaporation								×	×	×
Transpiration								×	×	×
Evapotranspiration				×	×		×	×	×	
Sorption to soil	×	×	×	×	×		×	×		×
Infiltration	×	×	×	×	×		×	×	×	×
Erosion		×	×							
Fertiliser						×	×	×		
Cropping	×	×	×	×					×	×
Irrigation	×	×	×	×	×	×	×	×	×	×

TABLE 1. Processes included in ³⁶Cl models.

within an order of magnitude The results do not therefore support the hypothesis that more complex (i.e. process-oriented) models perform better than conventional models in representing ³⁶Cl behaviour in soils and uptake into plants, to the extent necessary for post-closure safety assessment. Nonetheless, the results served to highlight the importance of clarity in both models and experiments with respect to which compartments of a system are assumed to be in equilibrium. In particular, where empirical data are used as input to a model, the representativeness of those measurements to the temporal and spatial assumptions within the model must be understood and correspond. This is particularly important for radionuclides such as ³⁶Cl for which specific activity or isotope ratio uptake models may be employed.

Addressing conceptual uncertainties in model approaches: the ⁷⁹Se project

Projects are often initiated with workshops to discuss the underlying science, with the result that new processes of potential relevance for key radionuclides may be identified. For example, during a workshop on the behaviour of ⁷⁹Se in soils and uptake into plants, volatilization of selenium from soils and plants was discussed. Such a process has in some cases been ignored in ⁷⁹Se assessment models to date, yet incorporation has the potential to significantly reduce the radiological significance of this radionuclide: for instance up to 6% loss per annum may occur (Smith et al., 2009). Although identified as potentially important, not all organizations will necessarily adopt the process within models. However, by identifying and recording processes of relevance for key radionuclides, assessors have the means by which inclusion or exclusion of a process may be justified. For example, exclusion of ⁷⁹Se volatilization might be justified in some cases as a means by which a requirement for a degree of conservatism can be incorporated within assessment models.

The 79 Se isotope is a redox-sensitive radionuclide with a complex environmental behaviour that is affected to a large degree by soil hydrological conditions. It may be present in soils in a number of different speciation states including selenate (+6), selenite (+4), elemental Se (0) and selenide (-2); organic forms may also be present. The different forms have the potential to coexist at any given time depending upon a

range of soil parameters, but particularly soil pH and redox conditions. The redox state affects not only mobility in soils, but also influences the ability of plants to take up 79 Se and the resultant distribution in plant tissues, which in turn governs entry into the food chain. In general terms, soil water content increases with soil depth, which in turn usually leads to more reducing conditions at depth, although the rooting zone of soil is not usually saturated, however, most soils experience dramatic changes in redox due to saturation at some point with saturation commonly cycling on a regular basis. In reducing conditions, ⁷⁹Se sorption is increased resulting in a reduction in bioavailability, but a higher potential for accumulation. The potential for volatilization from soils to occur is increased under more reducing conditions; however, highly saturated soils have a lower capacity for gas migration (Limer and Thorne, 2010; Smith et al., 2011).

The conventional approach to modelling ⁷⁹Se behaviour in soils and its uptake into plants requires the inherent assumption that the system is in equilibrium; an equilibrium exchange coefficient (K_D) is applied to represent the exchange between soil solids and soil solution and a concentration ratio applied to calculate plant concentrations in relation to the concentration in soil solution. Both parameters relate to total ⁷⁹Se concentrations. As K_D is determined under equilibrium conditions, soil redox induced changes in the form of ⁷⁹Se are not specifically considered; hence, the K_D approach may not be appropriate for a radionuclide with such complex environmental behaviour.

The range of values for $K_{\rm D}$ and concentration ratio is large, typically several orders of magnitude (International Atomic Energy Agency, 2009). One possible explanation for the range in values observed is variation in soil redox conditions at the time of analysis, which will greatly affect soil binding: variability in parameter values may therefore be an artefact of differences in experimental conditions. This leads to uncertainty in the selection of appropriate parameter values within assessment models.

As a result of discussions from the ⁷⁹Se project workshop and subsequent phase 1 report (Smith *et al.*, 2009), plus a parallel review of selenium behaviour relevant to dose assessment models (Limer and Thorne, 2010), three new models have been developed to represent soil behaviour and uptake into plants. The basis of the model varies in each instance.

Thorne (2010) presents a one-dimensional model, developed on behalf of Ciemat that is based on variation in soil hydrological conditions as an analogue for redox potential. The soil is represented by 10 separate layers for which separate $K_{\rm D}$ values are computed in relation to variations in soil layer hydrological conditions. Root uptake of ⁷⁹Se also varies with soil hydrological conditions and is limited to the unsaturated zone. Transfer to plants is represented by a soil-plant concentration ratio, but takes into account root distribution and total ⁷⁹Se concentration within soil layers. Losses from cropping (with potential return of organic matter to the upper soil layer) and volatilization (whereby rates are governed by soil hydrological conditions) are incorporated.

The ANDRA SAMM (SCM-ANDRA Multicompartment Model) approach (Miquel, 2008; Miquel and Basso, 2010) has some similar concepts to those described for the Ciemat model, but rather than representing the dynamic hydrology of multiple soil layers, SAMM integrates advection and diffusion processes between soil layers; the number of soil layers being user-defined. Diffusion of ⁷⁹Se in soils is controlled by the local concentration gradients defined on the basis of K_D in each soil layer. Transfer to plants is again represented by a soil-plant transfer factor. However, transfer is limited to the upper five soil compartments with root distribution reducing as a function of depth from 50% in the top soil layer to 1% in soil layer 5. Volatilization is excluded from the model.

The third model, developed by IRSN (Coppin et al., 2011) has recently been developed and is currently being validated. The model can represent different soil layers, the depths of which are user-defined. The selenium soil/ solution distribution within a soil layer is defined by using an equilibrium/kinetic model (E-K), which considers that the sorbed selenium is divided in two fractions: one labile fraction for which selenium is in equilibrium with the solution and another fixed (recalcitrant) fraction for which the sorption and desorption are kinetically limited. The fixed component is representative of selenium bound to organic matter for which only a slow release to the labile pool occurs as a result of mineralization processes. Account can be taken of the presence of stable selenium alongside the ⁷⁹Se in determination of the dynamics.

Each model is being applied to a defined scenario with the objective of evaluating the

performance of each, in relation to the conventional model approach, thus determining the degree of model complexity required to represent ⁷⁹Se behaviour. Results of the intercomparison will be made available in a future version of Smith *et al.* (2011).

Addressing broad conceptual knowledge gaps: focussed workshops

In some instances broad conceptual gaps may be identified and focussed workshops are organized with the aim of bringing together the scientific and assessment community to share knowledge and experience on specific assessment questions. Recently, workshops have been held to address gaps in knowledge on the behaviour of radium-226 (²²⁶Ra) in soils and uptake into plants, the behaviour of redox-sensitive radionuclides at the interface between the geosphere and the biosphere, and approaches for demonstrating compliance with environmental protection objectives for GDF non-human biota assessments.

It has become apparent in recent years that there are similar radionuclide migration and dose assessment issues for nuclear legacy sites and within the NORM (naturally occurring radioactive material) industry. As such, greater effort is being placed on fostering a knowledge-sharing network through workshop participation. The workshops on ²²⁶Ra and behaviour of redox sensitive radionuclides at the geosphere-biosphere interface (reported in Smith et al. (2010a) and Smith and Smith (2011), respectively) were particularly successful in drawing together experts from these different fields. Identified issues relating to ²²⁶Ra are being addressed, in part, through an ongoing project investigating disequilibrium in the ²³⁸U decay series.

In other instances it is evident that there are important gaps in developing assessment approaches. This is particularly the case for nonhuman biota (NHB) assessment approaches that, throughout Europe, are largely developing in relation to conventional releases to the environment (i.e. gaseous and liquid industrial effluents). Whereas results from recent NHB assessments (e.g. Smith *et al.*, 2010*b*; Torudd, 2010) indicate that potential doses from post closure releases are low (and below current guidance levels applied internationally), focussed workshops have provided the opportunity for the radioactive waste assessment community to discuss conceptual issues in the application of assessment methods to GDF scenarios. The workshops have also provided the opportunity to discuss mechanisms for compliance demonstration with active members of the International Commission for Radiological Protection (ICRP) Committee 5 that are responsible for the further development of the ICRP reference animals and plants assessment framework.

Reporting and dissemination

Reports are generated on all workshops organized through BIOPROTA and on specific projects. Each is subject to a review process whereby participants and sponsors are invited to make comment on draft material and, with the agreement of project sponsors, all reports are made publicly available on the forum website, www.bioprota.org.

In addition to annual workshop reports and the focussed workshop reports outlined above, the website contains reports detailing: (1) model review and comparison for the spray irrigation pathway (BIOPROTA, 2005a); (2) modelling the inhalation exposure pathway (BIOPROTA, 2005b); (3) model review and comparison for ¹⁴C (BIOPROTA, 2005c); (4) model intercomparison with focus on accumulation in soil (BIOPROTA, 2005d); (5) application of biotic analogue data (BIOPROTA, 2005e); (6) modelling processes in the geosphere-biosphere interface zone (BIOPROTA, 2005f); (7) guidance on site-specific biosphere characterization and experimental research and field research protocols (BIOPROTA, 2006); (8) investigation of ³⁶Cl behaviour in soils and uptake into crops (Limer et al., 2008) and dose assessment uncertainties and variability (Limer et al., 2009); (9) non-human biota dose assessment: sensitivity analysis and knowledge quality assessment (Smith et al., 2010b); and (10) modelling the abundance of 79 Se in soils and plants for safety assessments of the underground disposal of radioactive waste (Smith et al., 2009).

Summary

Focussed projects within the BIOPROTA forum have identified processes and parameters which have the greatest influence on, and relevance for, model representation of radionuclide behaviour in the environment. Identification of these processes and parameters enables site characterization studies to be targeted as a means of further improving confidence in safety assessments.

It has been the intention of the forum to work in a complementary way to other programmes, including the IAEA EMRAS and EMRAS II programmes. As such, representatives of relevant work programmes are invited to participate in BIOPROTA workshops to achieve knowledge sharing and ensure efforts are complementary. An additional benefit of the forum approach is that, by bringing together groups and knowledge bases from different geographical and cultural regions, alternative assumptions appropriate to reference futures may be identified. Maintenance of the forum website to allow wide dissemination of workshop and project reports ensures that shared knowledge is maintained.

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