Natural Uranium Fluxes and their Use in Repository Safety Assessment

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Abstract. Studies of uranium series geochemistry have made relatively little impact on performance assessment where simplistic advection-dispersion models are still being used to derive dose and risk estimates. Supplementing the models with a scientifically sound representation of radionuclide migration would greatly enhance the rigour and transparency of the safety case. This paper outlines a methodology for employing natural geochemical fluxes as alternative indicators of repository safety by reference to case studies in Finland. The approach may be applied to any situation where a comparison between ambient exposure rates and increments received due to industrial activities is required.

Introduction

The regulations used to judge the acceptability of an underground repository for nuclear waste are invariably based on dose and/or risk (*e.g.* Vieno and Nordman 1999; Nirex 2000). The most commonly used measure is radiological risk, where conventionally a value of less than 10^{-6} per annum is deemed acceptable. In Finland, where the spent fuel disposal programme is advanced, an effective annual dose of 0.1mSv to the most exposed individual is taken as the limit. Similar constraints are now being employed when devising remediation targets for radioactively-contaminated land or former uranium mining areas. The choice of these measures, though routine with operational nuclear plant, causes many problems in an environmental context, since:

- There is great controversy surrounding the health effects of prolonged exposure to low doses of ionising radiation.
- Local factors are not easily taken into account, for example natural background varies substantially within and between countries.
- Assumptions need to be made concerning site evolution and future human behaviour that are almost impossible to justify.
- Neither dose nor risk is a concept easily understood by non-technical audiences.
- There are a number of shortcomings with the models employed.

For these reasons, the IAEA, among others, has initiated research into alternative indicators of safety, which may be used initially as an adjunct to dose and risk (Miller 2000). The current paper describes part of this work, focussing on the application of natural uranium series concentrations and fluxes in Finland.

Natural safety indicators

Analogues and performance assessment

Similarities between natural uranium occurrences and repositories for spent nuclear fuel have been recognised for many years, accounting for the large number of 'natural analogue' investigations undertaken world-wide (*e.g.* Chapman et al. 1991; Read et al. 1993; Duerden et al. 1994; Blomqvist et al. 2000). Surprisingly, and despite the resources expended by waste disposal agencies in carrying out the work, these studies have made relatively little impact on performance assessment (PA) procedures to date. This may be due to a number of reasons, including *inter alia*:

- The data derived are often qualitative and do not conform to the input requirements of existing models.
- The geochemical concepts are complex and have not always been well articulated by specialist investigators.
- Inertia on the part of workers involved in safety assessment calculations who are reluctant to abandon 'tried and trusted' methods, even where shortcomings are evident.

In addition, many of the analogue investigations, particularly the earlier studies, were self-contained and were not carried out with performance assessment in mind. Nevertheless, and whatever the cause, the direct consequence of this lack of interaction is that overly simplistic, physical dispersion models are still being used to derive desired dose and risk estimates for repositories many thousands of years after closure. Problems with such an approach are well documented. They may fail to satisfy either scientific reviewers, owing to omission of geochemical reactions, or the general public, due to the lack of transparency in the final results.

The main concern with performance assessment is that the modelling tools take no account of interactions between geochemical and hydrological processes. Further, they fundamentally misrepresent the known geochemical behaviour of the uranium series elements (Read and Hooker 1992; Burns and Finch 1999; Hellmuth 2001). The calculations are biased towards dispersion and dilution of activity in the far field where, normally, dose rates are estimated for a drinking well scenario (*e.g.* Vieno and Nordman 1999). Consequently, if there were to be an accu-

mulation of activity, as is often the case, sinking a well into groundwater above the repository would give rise to higher doses than those calculated.

A number of biosphere receptors (*e.g.* peat) also have the ability to concentrate radioactive species (Armands 1967; Halbach et al. 1980; Read et al. 1993; Porcelli et al. 1997) and, thus, neither the geosphere nor the biosphere can be regarded purely as a dilution medium. Indeed, Hellmuth (2001) states that "(the assumption of dispersion and dilution)… of ions released from the waste, transported through the geosphere by groundwater and finally reaching the biosphere is not supported by any observation from natural anomalies."

The complexity of the natural environment necessitates approximation but, in the past, the simplification has been taken to extremes. All geochemical processes affecting radionuclide release are reduced to two 'equivalent' parameters (limiting solubility and Kd) and, therefore, the mineralogical and geochemical controls are obscured. Of the various reasons advanced for the simplification process, or 'disaggregation' as it is often called, few stand up to scrutiny. The need for statistical sampling of parameters need not apply as deterministic treatment may well be adequate and limited computational power is far less of a constraint than hitherto.

The issue then is one of making natural system studies more relevant in an assessment framework while, at the same time, devising more justifiable radionuclide migration models that can be verified against observations from natural systems. The two aims are not incompatible.

The advantage of safety indicators

The purpose of PA is to help convince observers that disposal of radioactive waste in an underground repository is 'safe'. Therefore, the process of quantifying safety cannot be divorced from the need to communicate the results effectively. With operational plant, radiological dose can be calculated readily but, in a long-term performance assessment, the estimates are heavily reliant on assumptions made of barrier evolution, climate change and future human behaviour.

The situation is slowly changing and several countries have now embraced the concept of using natural geochemical concentrations or fluxes directly as indicators of repository safety (Miller 2000; Hellmuth 2001 Traber 2002). Such an approach offers the facility to compare the results of safety calculations directly with natural sources of exposure to ionising radiation; a concept more accessible to non-technical audiences than probabilistic dose or risk estimates.

This confers a number of advantages:

- abundant data are already available, in many cases for systems very similar to those envisaged for disposal.
- the values are measurable directly and readily understood; they can be compared directly to drinking water or other safety standards.

- reasoning by analogy is a powerful argument in convincing sceptical observers.
- the processes leading to contaminant migration and retention from an ore body should be similar to those that would operate in a repository, at least for simple waste matrices such as spent fuel.
- estimation of dose requires prior calculation of spatial concentrations as a function of time and, therefore, no additional modelling capability is required.

Comparison of model output with natural concentration ranges is an obvious means of assessing parametric uncertainty. It provides a check on the realistic bounds of the simulations and this is where emphasis has been placed to date. However, conceptual uncertainty is at least as important and is perhaps, the main area where indicators offer potential for enhancing performance assessment exercises. No amount of statistical sampling will compensate if the parameters being sampled do not reflect the processes actually occurring. For this reason, both parametric and conceptual uncertainties are addressed in the succeeding sections.

Methodology

For natural indicators to form part of the basis for a safety case there needs to be consensus on:

- The aspects of a natural system that may be compared to a waste disposal situation
- The means by which parameter values can be calculated.
- Methods used to compare them to repository-derived equivalents.
- The constraints on the approach.

Each of the above is addressed briefly to examine how such a consensus could be reached.

Parameter selection

Any one of three measures, either individually or in combination, may be used to place a repository-derived impact in the context of natural systems. These are respectively, concentration, flux and accumulated mass.

Concentration, expressed in mass terms or as activity, is the simplest measure to convey. It can be demonstrated, for example, that the uranium concentration in natural groundwaters falls within a given range and, even above ore deposits, does not exceed a certain threshold value owing to mineralisation reactions in the

subsurface. Current PA modelling tools do not include these reactions and so such a demonstration is not possible.

The number of factors affecting absolute radioelement concentrations in groundwaters is very large. However, source mineralogy, aqueous speciation and the solubility of derived phases constitute the major controls. Where two or more components can be shown to derive from the same host, relative measures of concentration provide a better guide to mobilisation and redistribution patterns. This is evident from the enormous body of research dealing with fractionation along the homologous rare earth (REE) series. The approach can also be applied to uranium and thorium, where conventionally 'immobile' Th is taken to be invariant in mass balance calculations (*e.g.* Braun et al. 1993). Thorium satisfies most of the requirements of an immobile element, notably very low aqueous solubility and occurrence in phases resistant to weathering. However, caution is needed when interpreting elemental as opposed to isotopic patterns since the mineralogical associations of ^{232}Th and ^{238}U do not necessarily coincide.

Nuclide flux (the rate of mass transfer per unit area normal to the direction of mass flow) can be measured directly in some situations, for example at springs or in spas but, more commonly, has to be inferred from other evidence. In the case of surface processes, including rock/soil weathering, surface flows, estuarine mixing and lake or sea deposition, there is an abundance of data from geochemical surveys and investigations in model catchments (*e.g.* Porcelli et al. 2001; Tarvainen et al. 2002.

Investigations in areas of high natural radioactivity demonstrate clearly that few trace elements are dispersed following alteration of primary ores. These include uranium (*e.g.* Miller et al. 1994 and references therein; Blomqvist et al. 2000); thorium (*e.g.* Braun et al. 1993; Read et al. 2002) and the rare earths (*e.g.* Cuney and Mathieu 2000). The secondary deposits so formed may reflect very significant mass accumulation and, in some cases, constitute more important sources to groundwater than the primary deposit (*e.g.* Blomqvist et al. 2000). Again this feature is not accounted for in the existing PA methodologies.

The uranium-thorium series nuclides with their wide range of daughter half-lives offer an additional advantage in that they provide the means of assigning time frames to contaminant migration events on a scale commensurate with the needs of a safety assessment. One example of the approach is the Broubster study in northern Scotland where source rock U depletion was matched with accumulation in surface peat deposits. The peat was dated radiometrically and this information, used in conjunction with mass balance calculations, allowed accumulation and historical flux to be inferred (Read et al. 1993). In a similar study on the Finnish Peräjävuoma peat, Porcelli et al. (1997) obtained enrichment factors relative to local waters of 10^5 and $6x10^5$ for U and Th, respectively.

Comparison of parameter values

In a recent investigation, Miller (2000) calculated elemental fluxes for four diverse geochemical environments in the UK; the Carnmenellis Granite, the Chalk aquifer

of the London Basin, the thermal springs at Bath and that part of the Oxford Clay formation surrounding the upper reaches of the Thames. He then compared gross-averaged concentration and flux data for uranium and other elements with the results of earlier PA calculations (Nirex 2000). He also alluded to the large total mass and activity contained in these systems when viewed against the inventory of a spent fuel repository in Scandinavia.

This type of exercise, though useful for illustrating the potential of natural safety indicators, needs considerable refinement before application to a repository safety case. In particular, it has to be ensured that:

a) There is commonality of scale between the natural sources cited and a repository. For example, the volume of the Carnmenellis Granite is estimated to be 800km^3. With large areas, the number of potential discharge points is increased and the effects of intrusion into the body correspondingly reduced. As a consequence the impact on individual receptors will be markedly different.
b) The mineralogy is comparable to the waste form being considered and would generate a similar source term (*e.g.* uraninite and spent fuel, monazite and synthetic phosphates).
c) The models used in the performance calculations accurately represent the processes occurring at the locations considered.

The most complete and relevant data set from which to develop an improved methodology applicable to a safety case for spent fuel disposal in Finland is Palmottu (Blomqvist et al. 2000). This site forms the focus of the case study described in the next section.

Scenario development

A crucial element of PA modelling is the definition of future states. At its simplest level, the process merely involves specifying the states *a priori* (*e.g.* Vieno and Nordman 1999). The other extreme is represented by the so-called 'dynamic simulation' approach in which an attempt is made to model system evolution explicitly (Thompson and Sagar 1993). Most commonly, however, both waste disposers and those regulating the industry favour the scenario development approach (*e.g.* Bonano et al. 1988; Eng et al. 1994).

Each scenario (*i.e.* a postulated sequence of future events) is derived by defining the separate components of the system of interest and their respective interactions. Various methods have been devised for representing the interactions and for formalising the judgement process. The weighting assigned to interactions is then used to indicate the level of treatment needed in the performance assessment calculations.

Over the past few years, considerable attention has been placed on 'FEP' (Features, Events, Processes) lists for scenario development, notably in Sweden (*e.g.* Skagius et al. 1995). Recognition that *processes* (chemical, physical, biological) act on *features* of the system (barriers, rock mass, groundwater) to produce

events is the first stage in constructing defensible scenarios that may be justified by recourse to nature.

At the very least, observations on natural systems provide a check on the realism of scenarios derived by elicitation or other methods. At best, they constitute the basis of a viable alternative to current approaches.

Palmottu case study

Site characteristics and model conceptualisation

The Palmottu U-Th mineralisation in SW Finland (Fig. 1) was discovered by airborne radiometric surveys in the late 1970's and was a target of intensive uranium exploration from 1979 to 1984. The ore body consists of narrow pegmatite veins (2-10m) cutting granite and mica gneiss. The mineralisation extends to depths of at least 400m and total resources are estimated at around 1,000 tonnes (Kaija et al. 2002). The deposit proved to be too small for mining purposes, but provided an excellent opportunity for studying radionuclide transport along well-identified groundwater pathways in the fractured crystalline rock of the Fennoscandian Shield. Analogue studies have been in progress at Palmottu since 1987 (Blomqvist et al. 2000), aimed at characterising the geology, hydrology and hydrochemical setting of the uranium mineralisation.

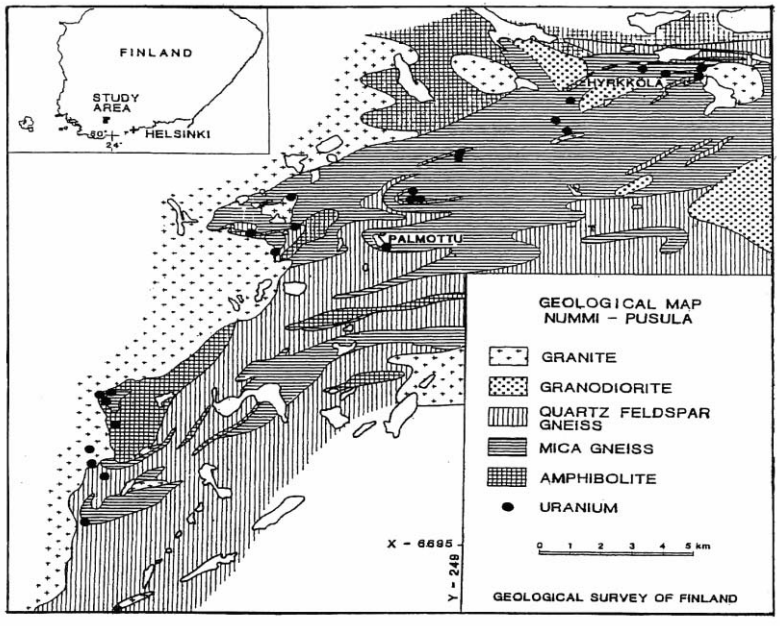


Fig. 1. Geology of the Palmottu study site

The present groundwater flow system dates back to the last deglaciation and the subsequent emergence of the landmass some 10,000 years ago. The uranium deposits extend from near the bedrock surface through a redox transition zone into a strongly reducing groundwater environment. This has allowed the full cycle of uranium redox chemistry under natural conditions to be studied.

Glacial tills or glaciofluvial formations largely cover the bedrock. In local depressions, post-glacial clays were deposited during different stages of Baltic Sea ingress and retreat. Soon after deglaciation, peat started to form in these depressions and has now reached a thickness of 3-4m. Near the exposed U mineralisation it contains several hundreds of ppm of uranium in ash.

In many respects, the geological history of the Palmottu deposit resembles the postulated evolution of the repository proposed for spent U fuel at Olkiluoto (Crawford and Wilmot 1998). These include the inventory, host rock and geographical setting. The deposit is limited in size, contains uranium mainly in the form of UO_2 and has been subjected to a series of perturbations since its emplacement, including an extensive drilling campaign as also envisaged at Olkiluoto. The site experienced continental ice margin conditions and a unique feature of the Palmottu exercise is that the influence of palaeoclimatic changes on groundwater geochemistry formed a central part of the investigation throughout. However, the modelling approaches used at the analogue site (Blomqvist et al. 2000) are very different to those employed in the most recent Finnish safety case, TILA-99 (Vieno and Nordman 1999). Here, a number of discrete canister failure scenarios (small/large hole, canister disappearance after 10,000 years) were combined with hydrodynamic dispersion in the far field. Thus, rather than consider the repository as a geochemically anomalous 'anthropogenic ore body', the safety case assumes that the system becomes homogenised and indistinguishable from its surroundings. The effects of this fundamental difference in model conceptualisation are now explored.

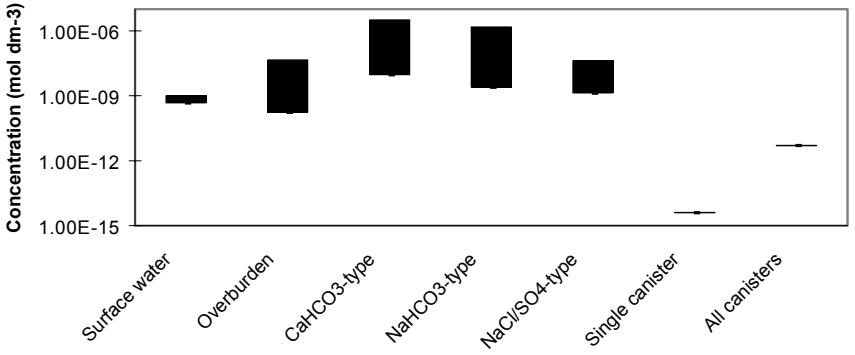
Concentration and flux estimates

The TILA-99 performance assessment was designed to be transparent and is readily amenable to comparison with alternative approaches. Dose calculations relate to a well for drinking water that is assumed to be located in the vicinity of the repository or in the groundwater discharge zone. This is the only exposure pathway considered. The model employed assumes that the annual releases from the repository into the biosphere are diluted in 100,000 m^3 of water and that an individual drinks 500 dm^3 of this water per year (Vieno and Nordman 1999).

The activity entering the biosphere essentially depends on the values chosen for solubility in the near field and equilibrium distribution coefficients (Kd) for barrier components and the host rock. In the case of uranium, solubility ranges from $3x10^{-7}$ to 10^{-4} mol dm^{-3} for reducing and oxidising conditions, respectively. Such

values do not seem unreasonable (*e.g.* Burns and Finch 1999) but when combined with sorption, diffusion and dispersion terms the net effect is marked.

No ^{238}U releases are predicted for the reference damaged canister scenarios. Even for cases where the canister is assumed to 'disappear', the maximum release rate from the geosphere is given as only 1.2 Bq y^{-1} (and then after 1 million years). This equates to a concentration of $4x10^{-15}$ mol dm^{-3}. In comparison, uranium concentrations measured at Palmottu range from around $4x10^{-9}$ mol dm^{-3} for the most reducing waters to $>10^{-6}$ mol dm^{-3} nearer the surface (Kaija et al. 2002). The safety case considered release from only one canister, however scaling up to match the total inventory at Palmottu (~1000 t), is insufficient to account for such a large discrepancy. If all the canisters were assumed to disappear there would still



be 10^3-10^6 times less uranium in the simulated well water than is actually observed in the geologically similar Palmottu system (Fig. 2). The difference is entirely due to assumptions made regarding the geochemical behaviour of uranium. This is not only a feature of TILA-99 (Vieno and Nordman 1999) but of all performance assessment calculations (*e.g.* SKB 1999; Nirex 2000). Indeed, the Finnish approach is rare in its clarity of procedure, allowing such checks for realism to be made.

Fig. 2. Uranium concentrations in Palmottu surface and groundwaters compared to PA predictions for a drinking water well.

It is important to note that these calculations do not suggest that the disposal concept is 'unsafe'; the highest concentration of U found in Palmottu groundwaters would produce a dose of less than 0.25μSv y^{-1} constrained by the solubility of secondary β-uranophane, which is abundant at shallow depths. The issue is one of increasing confidence in the predictions.

The approach is easily extended to alternative exposure pathways or other inventory components. For example, the maximum ^{226}Ra flux to the biosphere is estimated at around 2MBq y^{-1} in the reference scenarios and in the (absolute) worst case scenario as 70MBq y^{-1}. Bearing in mind that the disposal concept involves emplacing a significant radioactive heat source at depth, it is not unreasonable to compare these values to those for natural thermal spas. It can be seen from Table 1

that the annual ^{226}Ra release, even for the worst case scenario and regarded as highly pessimistic by the authors (Vieno and Nordman 1999), is substantially less than that found, for example at Bath Spa (Kellaway, 1991). The latter is by no means exceptional in European terms (Carlé 1975; Traber 2002).

Table 1. Annual releaes of ^{226}Ra calculated from the recent PA exercise and Bath Spa.

	Reference	High flow	Worst case	Bath Spa
^{226}Ra (MBq/y)	6.7×10^{-6}	1.9	70	200

As a drinking water well was the only exposure route considered in the recent PA exercise, no quantitative data on surface accumulation of activity are available. Nevertheless, the authors comment on the long term evolution of the candidate sites and the mix of freshwater, forest, peatland etc. environments that could exist. Data for a number of surface media are available from Palmottu, including stream and lake water, overburden, sediments, till and peat (Kaija et al. 2002). Since the last glaciation, the lake sediments are estimated to have accumulated about 130kg uranium with an additional 100kg in two local peat bogs. The Palmottu peats are not particularly radiogenic and the values may represent a lower bound. More than 1t U was found in peat associated with the much smaller ore body at Broubster (Read et al. 1993) whereas several percent U has been found in other Scandinavian and Russian peat bogs.

Similar verification calculations to those described above may be carried out for concentrations, fluxes and accumulated masses of other naturally occurring elements. Nor are artificial isotopes necessarily precluded as discussed below.

The potential role of natural indicators

Several radiologically important elements do not occur at significant concentrations in nature. Miller (2000) highlights this as a severe limitation of natural safety indicators and on the use of analogues in general. Though true to an extent (chemical analogues exist in the majority of cases and artificial isotopes are now widespread in the environment) it applies equally to any alternative measure of performance currently available. Importantly, there are no *additional* problems when compared to the use of dose or risk constraints but there are important advantages, including the transparency of the method and the ease of communicating the results to regulators, scientific reviewers and the Public. At the very least, the use of natural safety indicators, by focussing on the behaviour of geochemical systems, provides a firm basis for extrapolation. Thus, though the use of natural safety indicators is not problem-free, most limitations are procedural and may be overcome by enhancements to the current methodology. For example, new research into the weathering of DU munitions is already furnishing useful data on the likely fate of trace impurities.

Now that arguments in favour of natural safety indicators have become more widely accepted, it is worthwhile assessing the extent to which they could replace

dose/risk estimates or whether, as suggested by Miller (2000), they should be viewed as complementary.

A common feature of all PA modelling carried out to date is the hierarchical sequencing of computer programs whereby detailed process-specific models are progressively simplified until one reaches a stylised representation of the entire 'disposal system'. Only at the last stage are parameters sampled and thus, the often-substantial uncertainties associated with the more fundamental aspects of modelling tend to be hidden. This provides much of the rationale for natural indicators, *i.e.* a more transparent safety case. Criticism of PA calculations is at least partly justified, therefore, as in actuality the models do not describe 'the system' but are merely simplistic calculations of groundwater-mediated radionuclide transport. Calibration against more detailed models (where carried out) rather than provide surety often tends to introduce additional errors.

It is possible that, in future, the additional layer of 'PA modelling' could be excluded from safety assessments in favour of an explicit treatment of geochemical transport. This would provide a means for direct comparison of output with natural fluxes and related indicators. If required, conversion of nuclide concentration profiles to dose or risk values could be performed easily by post-processing. In the short term, however, it is likely that both approaches will be applied in tandem.

One consequence of adopting natural indicators as performance measures is the greater visibility of the models used to simulate radionuclide migration. Chemical transport models have to be employed and shortcomings with respect to geochemical site data, thermodynamic/kinetic constants, hydrogeological characterisation etc. are more readily apparent. This is not necessarily a disadvantage if it stimulates the research needed to remedy the deficiencies.

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