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Chapter 5 Modelling radionuclide transport in the geological environment: a case study from the field of radioactive waste disposal.

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Contents

5.1 Introduction

This chapter provides a short introduction to modelling the distribution of radionuclides in the geological environment (or geosphere). The chapter begins with the basic principles of radionuclide geosphere transport modelling are examined, with the emphasis on identifying the processes and structures of relevance to contaminant transport. Approaches for including these relevant features in transport models used to assess the long-term performance of radioactive waste repositories are then discussed, followed by an explanation of how such models should be tested to ensure that all relevant mechanisms have been included and are represented in an appropriate manner.

There are several differences between repository performance assessments and the use of the models to assess groundwater resources or pure research on radionuclide chemistry in the geosphere and these will be examined and explained, mainly by use of examples from the deep geological repository programme in Switzerland[1] . The main difference is that, in a repository performance assessment (or PA), the radionuclide transport models are used as a tool to assess risk and safety and are applied in a relatively crude fashion. Obversely, building confidence in the results of the calculations employs series of tests which are much more thorough, better documented and requiring of much greater effort than anything generally considered in more traditional areas of radionuclide transport modelling. This dichotomy will be highlighted with appropriate examples from the literature and the subsequent effects on the progress of the modelling of radionuclide transport in the geological environment discussed.

5.1.1 Sources of radionuclides

A wide range of radionuclides can be found in all components of the geological environment – the number of specific radioisotopes identified being primarily limited by the sensitivity of the analytical techniques used. These radionuclides include:

- "Primordial" isotopes which are sufficiently long-lived to have persisted in measurable quantities since nucleosynthesis of the elements which make up the earth (>~5 x 10^9 years ago)
- Radioactive daughters (usually shorter lived) which are continually produced by the decay of primordial isotopes; of particular relevance are the "natural decay series" chains of daughters produced by the decay of ^{238}U, ^{235}U and ^{232}Th.
- Radionuclides produced in situ by nuclear reactions; of particular importance are neutrons from the background of spontaneous fission of some actinide elements which can produce radionuclides by (n, α) and (n, p) reactions.
- "Cosmogenic" radionuclides which are produced continuously in the upper atmosphere and enter the geosphere through the water cycle.
- "Anthropogenic" radionuclides which are produced by the activities of man. Major sources to the geosphere include general atmospheric input from nuclear weapons testing and nuclear fuel cycle activities/accidents and direct underground input at specific locations due to disposal of radioactive wastes or, again, nuclear weapon testing.

[1] Radioactive waste repository designs vary depending on the precise waste type. This chapter focusses on what is commonly termed ‘deep geological disposal’, meaning the waste is emplaced in a repository constructed at depths of several tens to over one thousand metres deep, so making use of the surrounding geological environment to both protect the waste itself and to retard any radionuclides released from the repository.

It should be emphasised that specific radioisotopes may have more than one source: for example, ^{3}H which can be produced *in situ*, is formed cosmogenically, is a component of anthropogenic atmospheric fall-out and is also a major component in many wastes considered for geological disposal.

5.1.2 Measuring and modelling radionuclide mobility

From a purely academic point of view, the radioactive decay process has two key features which attract interest to the study of subterranean radionuclides:

- The simple first order exponential decay law characterised by a single constant which can, in principle, be measured simply and accurately. This provides a "clock" which is the basis for an enormous range of geochronological techniques (see, for example, Faure, 1998)
- The elemental transmutation and energy release associated with the decay process which can lead to partitioning of parent and daughter isotopes. This provides a valuable tool to assess in-situ elemental mobility / geochemistry over long periods of time (for examples, see Ivanovich and Harmon, 1992).

Additionally, from a practical viewpoint, the radiation produced by the decay process provides a convenient method for quantifying specific isotopes at very low concentrations. Indeed, until the relatively recent development of mass-spectrometric techniques, the sensitivity of radioanalytical methods was an important consideration in the selection of radionuclides as tracers of environmental processes.

The simplest application of radioactive decay considers "closed" systems in which no material exchange occurs. For example, a mineral which incorporates a radionuclide at a known concentration (or activity) or which excludes the daughter resulting from decay of this isotope can be readily dated by quantifying the extent of decay which has occurred. The principles involved and practical aspects of their application to a wide range of systems are discussed in detail elsewhere (*eg* Eicher, 1976, Ivanovich et al., 1992 and Press and Seiver, 1998).

Of more interest for the purposes of this chapter, however, are open systems in which a material flux exists. This chapter will focus exclusively on fluxes of radionuclides in groundwater although many of the basic principles could be applied to other fluids (magma or gas).

For any open system, the parameters of interest could include one or more of the following:

- Rate of radionuclide release from a specific source
- Rate of transport of the fluid or contained solute
- Rate of accumulation of radionuclide at a specific sink.

Either the radionuclide examined could be of direct interest or it could be considered as an analogue for the behaviour of other isotopes of the same element or, indeed, chemically similar elements (see discussion on analogue and homologue elements in Petit, 1991 and Miller et al., 2000).

Such information is clearly important for studies of geochemical cycling of elements, either for purely academic reasons or for commercial application in identification of the key factors influencing the formation and longevity of ore bodies (eg Plant et al., 1999). In other areas of the geological environment, the ability to trace and evaluate groundwater flow rates and provenance has long provided a valuable hydrogeological tool for groundwater exploitation (eg Land, 1966). Indeed, historically, this was the main reason for the development of many groundwater modelling tools earlier last century. These tools dramatically increased in power and definition with the introduction of stable and radioisotopic analytical methods, so allowing a better definition of groundwater ages and temporal evolution (see, for example Osmond and Cowart, 1976 and Loosli et al., 1991, for an overview)

The development and application of such models of radionuclide behaviour in the geological environment in these more 'traditional' areas of study are adequately covered in the literature (see, for example, Faure, 1998, Pearson et al., 1991, and Ivanovich and Harmon, 1992). Increasingly, however, another justification for such modelling studies is associated with the disposal of wastes (both radioactive and chemotoxic) or the remediation of areas already contaminated with such material and this, the modelling of contaminant transport in the geological environment, will form the main focus for this chapter.

5.1.3 Historical overview of radionuclide transport modelling in the geological environment (geosphere)

Although the basic principles involved were established early last century, systematic studies of radioactivity in the geological environment (or geosphere) date from the latter half of the century, associated with considerable advances in analytical methodology. Initial emphasis was very much on the geochronology of closed systems and, in parallel, uranium-series geochemistry (linked, to a large extent, to the expanding military and civil use of nuclear materials).

Quantitative studies of radionuclide mobility in the 1960s and 1970s were primarily associated with quantifying groundwater flow in aquifers or evaluating radionuclide transport from subterranean anthropogenic sources (nuclear weapon explosion cavities, waste disposal sites)[2]. Mathematical models used were, of necessity, rather simplistic, limited by both the available databases and the power of computational tools.

During the 1980s and 1990s, great efforts were made in the characterisation of rock formations previously simply regarded as aquitards and the true complexity of solute transport in a wide range of geological formations became increasingly evident. Key topics of interest during this period included:

- The rôle of geological structure/heterogeneities/anisotropies (limitation of equivalent porous medium models and upscaling problems; see comments in Smith et al., 2001a,b and references therein)
- The diversity of individual processes included in bulk terms such as "sorption" or "retardation" and the problems of measurement (see, for example, Sibley and Myttenaere,1986)
- Complexities added by the presence of colloids, organics and microbes in groundwater (see review of McCarthy and Zacchara,1989)

[2] This was paralleled in marine radiochemistry with the use of isotopic signatures from atmospheric weapons testing and releases from nuclear accidents or reprocessing plants to trace ocean currents and to assess ocean residence times etc.

- Inherent errors and uncertainties associated with models and the need for verification and validation (see, for example, McCombie et al., 1991 and Pescatore, 1995)

Much model development focussed on the development of "coupled" codes which coupled physical (groundwater transport) and chemical (contaminant retardation) processes in one code, so providing computationally-efficient tools to quantify increasingly complicated conceptual representations of radionuclide migration (eg Lichtner et al., 1996).

Now, at the beginning of the new millennium, cheap computational power has changed the perspective on the quantification of radionuclide mobility completely. The limiting factors constraining more detailed quantification of radionuclide mobility in relevant rock formations are now set almost entirely by limitations in our ability to characterise the processes involved in situ and to validate model extrapolations made on the basis of laboratory-scale experiments to the temporal and spatial scales relevant to geological processes (see comments in Smith et al., 2001a,b and references therein).

5.2 Modelling radionuclide transport in the geosphere

5.2.1 Introduction

For most applications of radionuclide transport modelling, the model and data requirements are very similar, namely that all relevant processes and, in the geosphere in particular, structures are represented in an appropriate manner. This is the case whether transport in the rock formation is predominantly controlled by diffusion (eg in a salt diapir or in a consolidated claystone) or by advection in porous media (eg in a sandstone aquifer or fluvial gravel deposit) or in fractured media (eg in a limestone formation or an uplifted granite massif). Here, those features which are basic to most models are listed and described and, where appropriate, comments are made on outstanding data requirements for transport models.

Radionuclides are present in geological media in a variety of forms. These forms may be immobile, such as solutes in stagnant porewater in isolated pore spaces within the rock (and as fluid inclusions in minerals), and forms that are bound (sorbed) in some way to immobile mineral surfaces. They may also be mobile, such as solutes in connected networks of fractures and pores, volatile species in flowing gas[3] and forms that are bound to groundwater colloids (which, for the purposes of this discussion, may be treated as mobile solid surfaces). Radionuclides may also be exchanged between forms: mobile radionuclides may, for example, enter and leave solution many times during migration from a deep source (eg an ore body or a waste repository) and the surface as they pass through various geochemical boundaries in the geosphere. In order to model how radionuclides are transported through geological media, an understanding must be developed of the structure of the pore spaces (including fractures) within the media, the transport processes that operate within these voids and the processes by which radionuclides are exchanged between immobile and mobile forms. The following sections give a general discussion of transport-relevant processes and geosphere structures, although, it should be noted, the actual structures

[3] Gas-mediated radionuclide transport is not discussed further in this chapter (see overview of contaminant transport in gas by Philp and Crisp ,1982).

that are present (and the relative importance of the different transport-relevant processes) vary considerably between different geological media.

5.2.2 Transport processes and geosphere structures

Processes

Advection is the process by which radionuclides, which are associated, for example, with dissolved species or colloids, are conveyed by the bulk motion of flowing groundwater. The pressure gradients driving groundwater flow may arise, for example, from variations in the hydraulic head (due to variations in the height of the water table, for example), glacial rebound and variations in density associated with salinity and temperature contrasts.

Groundwater flow rates vary considerably in most media, due to the heterogeneity in fracture and pore space structure and to friction on fracture or pore space surfaces. There will exist some paths for advection through the media or fractures that are faster than the average and some that are slower. The resulting spreading of transported solutes or colloids is known as **mechanical dispersion**. Longitudinal dispersion, the spread in the direction of bulk flow, is generally much stronger than transverse dispersion perpendicular to the flow.

Diffusion is the process by which radionuclides migrate down gradients in chemical potential. Diffusion causes a spreading of transported solutes or colloids which, when combined with mechanical dispersion, is called hydrodynamic dispersion. The rate of diffusion is determined by the magnitude of the concentration gradient, and also by the diffusion coefficient of each given solute. The diffusion coefficient is itself a function of the properties of the rock, such as the tortuosity of pore spaces, the properties of the groundwater and, in particular, its temperature, and the properties of the diffusing species, such as their charge and size.

In addition to the direct processes of advection, dispersion and diffusion, a range of **coupled processes**, including thermal, chemical and electrical osmosis, thermal diffusion, hyperfiltration and electrophoresis, can transport porewater and radionuclides in solution in response to gradients in temperature, pressure, solute concentration and electrical potential. Although such coupling is generally negligable for most practical applications, some of these processes, including chemical osmosis and hyperfiltration, can be significant in argillaceous sediments, where the overlapping diffuse double layers of clay platelets result in the rock acting as a semi-permeable membrane. The complete Onsager matrix of direct and coupled processes is given in Table 5.1.

Where there is significant groundwater flow, advection and mechanical dispersion are often far more effective transport processes than diffusion or the coupled processes mentioned above. This does not mean, however, that these processes can necessarily be neglected. In many media, significant groundwater flow is confined to discrete water-conducting features, such as fractures, sand lenses, thrust planes, alluvial or glacial channels and rock unconformities. Elsewhere in the rock body, groundwater may effectively be stagnant. In such cases, diffusion may, for example, transport radionuclides from water-conducting features into stagnant porewater "matrix" regions and vice versa. This process of **matrix diffusion** (see Figure 5.1a) is referred to as a retardation process, since it results in slower transport than would be the case if only advection, mechanical dispersion and diffusion along water-conducting features operated. In some media, matrix pores may be accessible to solutes by diffusion, but larger molecules, ions and colloids may be excluded due to size and/or charge effects (Figure 5.1b). The retarding effect of matrix diffusion may thus apply to radionuclides associated with solutes, but not to those associated with colloids (although colloids may be retarded or immobilised in other ways, such as by filtration).

A number of **chemical retardation** mechanisms have been identified in natural systems, as shown diagrammatically in Figure 5.1c and d

- adsorption,
- ion-exchange,
- precipitation,
- mineralisation;

Adsorption and **ion-exchange** are often collectively termed **sorption**, a term used generically to encompass chemical interactions with solids that retard transport. Sorption is generally modelled as a reversible process whereby sorbed radionuclides may be released to solution if solution concentrations or composition change. Desorption kinetics may be slower than sorption kinetics and there may be instances where sorption could be considered irreversible[4].

Precipitation is not a sorption process but it can, on occasions, be difficult to discriminate between the two mechanisms in both field and laboratory studies. Generally, sorption would be expected to be the dominant process at low solution concentrations of radionuclides. As concentrations of radionuclides increase, precipitation of phases in which the radionuclide of interest is a stoichiometric component (ie an essential component of the mineral structure) may occur if saturation is reached in the groundwater. In circumstances where the total amount of dissolved solids in the groundwaters is high enough that precipitation occurs, a very complex chemical environment may develop in which radionuclides may co-precipitate as solid-solutions in a variety of mineral phases, or be scavenged by amorphous precipitates, such as iron oxyhydroxides, and also effectively be co-precipitated. The stability of newly precipitated minerals and amorphous phases depends on saturated concentrations being maintained. If concentrations fall below saturation, perhaps after a pulse of contaminated groundwater has passed, or as the radionuclide source area (eg the radioactive waste) is flushed with fresh groundwater, these minerals will begin to dissolve and release any radionuclides they contain back to solution. The situation can become even more complicated in microporous media, where particular solutes may be concentrated enough close to mineral surfaces to allow precipitation to occur - even when bulk solution is undersaturated.

The kinetics of sorption and dissolution and precipitation processes are clearly important to long-term predictions of radionuclide behaviour. Irreversible sorption (slow desorption) and precipitation processes are obviously beneficial in terms of radionuclide retardation because they immobilise radionuclides very effectively. They are, however, difficult to demonstrate, and,

[4] Strictly speaking and according to thermodynamics, no chemical process, such as the sorption of radionuclides by mineral surfaces, can ever be truly irreversible. All chemical reactions are reversible, the significance is in the time taken and the conditions necessary for the reaction to be reversed. When irreversible sorption is discussed in terms of repository PA, it is generally meant that the kinetics of desorption are slow compared to the time period of interest to the assessment, assuming the physico-chemical conditions are constant.

consequently, many PA representations of radionuclide retardation make the assumption that all chemical retardation processes are instantaneously reversible[5]

Sorption processes generally occur sufficiently rapidly (relative to transport rates) in the geosphere to allow their kinetics to be ignored in transport models, although precipitation and mineralisation kinetics would clearly be relevant if these processes were to be included in an model. Where groundwater flows are relatively rapid (as may occur, for example, in a major fracture zone or in a shallow aquifer), then retardation mechanisms become increasingly less significant in affecting the rates of transport of radionuclides.

Radioactive decay and ingrowth are, of course, important processes to be taken into account in evaluating radionuclide transport. Radionuclides may decay, or be created by ingrowth, during transport through a geological medium. If the time taken for a radionuclide to be transported across the medium is significantly longer than the radionuclide half life (and no ingrowth from a parent radionuclide occurs, including that of recoil from radionuclides already present in the rock), then very little of that radionuclide will emerge. If, however, some transport paths exist for which the transport times are less than the half life, then these can result in some radionuclide migration to the surface, even though a large proportion of the radionuclide mass may decay during transport along other, slower paths.

It was noted above that some radionuclides could be retarded and then released again to solution due to changes in the geochemical conditions of the geosphere. There are two other processes which contribute to changes in retardation potential of radionuclides in the geosphere, namely recoil and chemical transformation. **Recoil** is the process whereby a daughter radionuclide may be physically ejected from the site of the mother by the decay process (see Osmond and Cowart, 1976, for a detailed discussion). This may lead to direct transfer of the daughter from a mineral surface into solution or transfer of the daughter into another site within the mineral which may cause subsequent loss of the daughter to solution as the lattice is now damaged and so the daughter may be more readily leached from the mineral. In addition to the above chemical transformations, a change in speciation can occur (eg U^{4+} to U^{6+}) either during the recoil process itself or due to a different oxidation potential between the displaced site (eg in the groundwater) and the original mineral lattice site[6].

In **chemical transformation** the decay process can produce a daughter whose geochemical characteristics differ markedly from the mother, so changing the retardation properties of the daughter. A very good example is that of ^{226}Ra and its daughter and granddaughter. In a sulphate rich groundwater, for example, ^{226}Ra can be present as $^{226}RaSO_4$ and so may be co-precipitated in gypsum (as $Ca^{226}RaSO_4$) or barite ($Ba^{226}RaSO_4$). Upon decay, however, the daughter ^{222}Rn, in gaseous form, usually diffuses out of the mineral structure where it can be transported in the groundwater as it decays through a series of short lived daughter radionuclides to the relatively

[5] This aspect of PAs is often difficult for many people to understand: an important premise of a PA is to show that, despite assuming the worse case scenario for each process or mechanism (for example, ignoring geosphere retardation or irreversible sorption), a repository can be shown to satisfy the safety criteria set out by the regulatory authorities. Expressed in another way, the repository is deliberately 'over-engineered' to provide large margins of safety (ie boots and braces).

[6] Ku et al., (1992) noted that, in models of radionuclide transport in the geosphere, "The most appropriate formulation of recoil input remains an open question requiring further study." They further noted that their formulation "...should provide an upper limit for input by recoil...". Krishnaswami et al., (1982) and Scott et al., (1992) argue for much lower recoil-induced input on mathematical and chemical grounds, respectively. Indeed, Scott et al., (1992) further note that, in many cases, recoil loss *per se* will be minimal. See also the discussion in Short et al., (1988).

insoluble ^{210}Pb (which will sorb onto mineral surfaces, co-precipitate with iron and manganese oxyhydroxides or precipitate as lead sulphide).

Structures

Radionuclide transport is sensitive to the geometrical, hydrogeological, mineralogical and geochemical properties of a geological medium. In many rock types, water-conducting features provide the dominant pathways for radionuclide transport. These features may be structural or sedimentary in nature and include, for example, joints, mineralised veins, complex shear zones, regional faults, sedimentary lenses and various forms of alluvial channels. As illustrated in Figure 5.2, advection and dispersion are usually the dominant transport processes in deep geological media containing water-conducting features, with retardation by matrix diffusion, filtration and sorption[7].

The processes that dominate radionuclide transport often vary according to these properties, which are typically heterogeneous over a range of spatial scales. Even if it is the transport of radionuclides over a scale of tens or hundreds of metres that is of interest to the long-term performance of a deep geological repository, this does not necessarily mean that small scale structures and property variations can be neglected or "averaged over" (see comments in Smith et al., 2001a,b). Small-scale structures may, for example, provide relatively fast paths for advective transport, particularly if, say, they are lined with minerals that prevent or limit retardation by matrix diffusion and sorption. Alternatively, fracture infill material and altered zones adjacent to fractures may have porosities and mineralogical properties that favour retardation (see examples in Smellie et al., 1986;Moeri et al., 2001a and Ota et al., 2002).

The characterisation of a geological medium for transport modelling purposes typically, therefore, has to examine features at the scale of decimetres or less (*eg* Moeri et al., 2002, Ota et al., 2002), as well as those at a larger scale (*eg* Mazurek et al., 1997). Figure 5.3 shows an example, based on limestone beds in the Palfris Formation at Wellenberg in the Central Swiss Alps (a candidate location for a repository for low and intermediate-level radioactive waste), which illustrates the way in which information from hydraulic testing and geological characterisation, on scales ranging from 500 m to less than 0.1 m, provides input to radionuclide transport modelling.

In other geological media, argillaceous sediments for example, the low hydraulic conductivity, lack of water-conducting features and immobility of colloids means that transport is dominated by diffusion of solutes, with retardation by sorption (*eg* Gautschi, 2001). Coupled processes may also play a role where, for example, strong thermal gradients are present (see case study by Soler, 1999 and the overview of Horseman et al., 1996, for example).

5.2.3 Modelling approaches in a repository PA

5.2.3.1 Introduction

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[7] Although not included in Figure 5.2, transport of radionuclides sorbed on natural groundwater colloids may also have to be taken into account in such deep geological media. In shallower geological media, transport in association with microbes and colloids must also be taken into account as should geochemical fronts. For example, redox fronts are well-known sites of radionuclide retardation (causing, for example, U roll-front ore bodies). Unfortunately, the current understanding of radionuclide entrapment at such fronts is limited to a few (economic) elements and, even then, the radionuclide trapping efficiency of such systems is unknown (see review in Hofmann, 1999).

The approaches outlined above are appropriate to all types of radionuclide transport modelling in the geosphere. From this point, however, the modelling approaches employed in a repository PA will be described (for modelling approaches in more standard applications, see, for example, Osmond and Cowart, 1992, and Bruines and Genske, 2001) with particular reference to several Swiss studies on fractured rocks.

It must be emphasised that all models and databases are simplifications of reality.The processes that are incorporated and their representation in the model, and the geometry (structures in the rock) within which these processes operate, must be simplified because of the complexity of natural systems and the impossibility of complete characterisation of the geosphere. Within a repository PA, it is necessary to have confidence either that the simplifications have a negligible impact on the results or, if bounding estimates are acceptable, that the simplifications are conservative, leading to an overestimation of radionuclide releases and consequently doses[8]. It is also necessary to show that the parameters used in the PA are realistically (or, again, at least conservatively) assigned.

In accordance with current understanding of geosphere transport processes (see, for example, NEA, 1999), the dominant processes governing solute transport in fractured rocks are generally assumed to be:

- advection and dispersion within the water-conducting features and
- retardation due to matrix diffusion into the rock matrix and sorption onto mineral surfaces.

In PA, these processes are assumed to operate in extensive heterogeneous networks of water-conducting features, although a detailed, small-scale understanding of the structure of the features is also required in order to model matrix diffusion (see, for example, Nagra, 1994). As illustrated in Figure 5.3, the processes (including the geometrical parameters and the parameters that are used to define the rates and spatial extent of processes) are derived from a broad base of information. which includes the results of a range of characterisation techniques and general scientific understanding. The information is interpreted, in terms of transport-model parameters, by means of various supporting hypotheses and models (eg measured transmissivities are converted to advection parameters via groundwater flow models; sorption measurements are converted to transport model parameters via a Kd or sorption isotherm model: see, for example, Mazurek et al.,1992; McKinley and Alexander, 1992).

It should be noted that there may be many specific differences in the structures that are relevant and the parameter values used depending on whether a model is applied in performance assessment or to the modelling of a field experiment. If, for example, the aim is to interpret experimental results or field observations, or to forecast the outcome of a future experiment, a model prediction is likely to correspond to a "best guess" of the evolution of radionuclide concentrations or fluxes. If, on the other hand, the aim is to demonstrate the safety of a radioactive waste repository, a model prediction is more likely to correspond to a pessimistic "upper bound" of the fluxes to, or concentrations in, the near-surface environment. Uncertainties arising from limited understanding of transport-relevant processes and from incomplete characterisation of geosphere properties and structure will inevitably limit the accuracy of "best

[8] _ In the jargon of the radioactive waste disposal industry, this is called ‘conservative’ behaviour and may be compared with the boot and braces approach noted above.

guess" predictions, or necessitate the use of pessimistic model assumptions and data in making bounding predictions. The following sections discuss the modelling of transport-relevant processes and geosphere structures, including the influence of uncertainty on model formulation and parameter values.

5.2.3.2 Modelling transport-relevant processes

Some of the processes relevant to radionuclide transport are well understood and can be modelled using fairly simple relationships based on fundamental physical and chemical principles (such as Darcy's Law for advection, Fick's Laws for diffusion and the Bateman Equations for decay and ingrowth) and these are incorporated, in some form, into most radionuclide transport models. Other processes are more complex to model and, in some cases, less well understood, examples being advection in flowing groundwater in highly heterogeneous geological media and the range of processes that are grouped together as "sorption".

Two distinct approaches have been adopted for incorporating such processes in transport models:

(i) Incorporate detailed models for these processes in the transport models themselves.

(ii) Incorporate the processes in a relatively simple form in the transport models ("model abstraction" or simplification). Develop separate detailed models for the processes and use these to derive input parameters for the transport models.

The first approach is often favoured if the aim is to make best-guess predictions of radionuclide migration in, for example, aquifer studies. The second is frequently used for bounding estimates in a repository PA. The relative merits of the two approaches are discussed further in Section 5.3.

Examples of the first approach are the computer codes that have been developed for both groundwater flow and radionuclide transport modelling. Typically, the groundwater flow part of the model is first set up and calibrated against field measurements. Radionuclide transport is then modelled, for example, by assessing radionuclide advection through the calculated groundwater flow field, while taking into account other transport-relevant processes such as retardation by matrix diffusion and sorption. Numerous such codes exist for porous media rocks and research is currently ongoing to integrate mechanistic sorption models in such codes for fractured media (see, for example, NEA 1997, 1998).

Examples of the second approach are the computer codes that require, as input, the results of groundwater flow modelling (eg in the form of specific flow rates along discrete transport paths) and parameters describing the relationship between the amount of a radionuclide sorbed on solid surfaces and the amount in solution (eg the sorption (or distribution) coefficient, Kd[9], or sorption isotherms). Sorption coefficients, or sorption isotherms, are generally based on laboratory experimental observations. In theory, mechanistic sorption models (see review in Mangold and

[9] Simply put, Cr = Kd.Cw, where Cw = the solute concentration in the aqueous phase (mol m^{-3}), Cr = the concentration 'sorbed' on the solid phase (mol kg^{-1}) and Kd = the distribution coefficient (m3kg-1). In a simple system, such a Kd may be thermodynamically based, but the values used in transport models are usually an empirical representation of kinetically fast, concentration independent, reversible uptake. Unfortunately, many transport modellers do not understand this distinction and consequently employ the Kd concept in totally inappropriate circumstances (see discussion in McKinley and Alexander, 1992, 1993a,b, 1996, 2001, Alexander and McKinley, 1994 and Ku et al., 1998 for examples and critical discussion).

Tsang, 1991) can provide a tool for extrapolating such laboratory-derived sorption data into a representation of in situ radionuclide retardation in the geosphere. Unfortunately, the radionuclide sorption literature is, according to McKinley and Alexander (1993a) "....a morass of poorly defined concepts and inconsistent nomenclature." and so this approach must be treated with great care and a pinch of scepticism.

Other processes are also relatively complex or poorly understood and have, up to now, also invariably been modelled in a highly simplified manner, justified by empirical observation, by arguments that suggest the process can be neglected or by arguments that suggest the approach, though unrealistic, at least leads PA modellers to err on the side of repository safety. Examples are the modelling of mechanical dispersion, coupled processes and colloid-facilitated radionuclide transport. In the case of mechanical dispersion, spreading is generally modelled empirically as a diffusion-like process, calibrated against field-scale tracer tests. Although this empirical approach may be adequate for fitting radionuclide breakthrough curves (*ie* the plot of the radiotracer concentration observed at the output borehole versus time since injection at the input borehole), it should be noted that it breaks down if used to model radionuclide fluxes at different points within the medium. Indeed, field observations suggest that the dispersion coefficient that must be used to fit breakthrough curves is a function of the mean travel distance (Gelhar et al. 1992). In spite of this concern, the modelling of dispersion as a diffusion-like process is common in repository PA, and justified, in part, by the fact that calculated radionuclide releases from the geosphere to the near-surface environment tend to be quite insensitive to the value assigned to the dispersion coefficient (*eg* Nagra,1994).

5.2.3.3 Modelling geosphere structures: an example from fractured rocks

In any fractured rock, there exists a continuum of discrete, water conducting features at all scales, from dekamtres to millimetres. However, in PA modelling, there are two types of heterogeneity that, in general, need to be considered for transport-modelling purposes:

- The larger-scale heterogeneity, arising from the fact that a typical transport path through a rock body often consists of a network of features with different properties.

- The smaller-scale heterogeneity corresponding to the internal structural of the features, which may arise, for example, from a multi-phase history of deformation and mineralisation.

In both cases, the structural complexity of most geological media, and the limited data to characterise this complexity, means that geosphere transport models almost invariably include a high degree of simplification.

In modelling groundwater flow, smaller-scale heterogeneity can often be neglected. This heterogeneity determines the transmissivities of water-conducting features, but transmissivities are usually measured directly (encompasing relatively large sections of a borehole and, therefore, usually including numerous water-conducting features), rather than derived from the direct characterisation of smaller-scale structure. It is also common in groundwater flow modelling to simplify larger-scale structures and to treat geological media either as continua or as discrete fracture networks. In the continuum approach, the actual medium, with its structural heterogeneity, is conceptually replaced by a more homogeneous "equivalent porous medium (EPM)", characterised by macroscopic parameters, such as the hydraulic conductivity. In the discrete fracture network approach, flow in the volumes of rock between fractures is assumed to be negligibly small and the fractures themselves are treated as, often homogeneous, planar features characterised by macroscopic parameters, such as the transmissivity. In both cases, macroscopic empirical laws, such as Darcy's Law, are used to describe the average groundwater flow through the medium as a whole or the flow through the discrete fractures in response to pressure gradients.

Figure 5.5 shows an example of how a conceptual model of larger-scale heterogeneity in a geological medium can be simplified for the purposes of modelling groundwater flow. In this example from crystalline basement of Northern Switzerland, a potential repository host rock, a set of three nested models is used. A regional-scale EPM model provides boundary conditions for a more detailed local-scale hybrid model, in which some of the larger linear structures ("major water-conducting faults (MWCF)") are modelled explicitly. The local-scale model in turn provides boundary conditions for a block-scale discrete fracture network model, which is still more detailed. It is only on the relatively small block scale that the network of individual water-conducting features is modelled explicitly.

In the context of transport modelling, it is important to emphasise that the groundwater flow modelling techniques outlined above yield average information about flow, either within the medium as a whole or within discrete features. A knowledge of the average flow within a water-conducting feature is necessary, but not sufficient, to model radionuclide transport. Channelling of the flow (which determines the actual rates of advection and gives rise to dispersion) and the effectiveness of matrix diffusion and sorption as retardation processes are strongly influenced by the internal structure of the features. Figure 5.6 gives an example of the internal structure of one type of water-conducting feature observed in the crystalline basement of Northern Switzerland, and the simplification of this structure that is made for transport modelling purposes. In this example, advection/dispersion occurs in channels, embedded within fractures that are themselves contained within the water-conducting features.

Many paths for advective/dispersive radionuclide transport through a geological medium may exist, consisting, for example, of series of connected channels within a larger network of fractures, and these paths may vary widely in their transport-relevant properties, both along their lengths, and between paths (see, for example, Moeri et al., 2002, Ota et al., 2002, for a clear example). Nevertheless, it is common in transport modelling to consider just a few parts (sometimes only one) as representative of the whole range. These representative paths are assigned uniform properties along their lengths, such as transmissivity, cross-section geometry, and properties relevant to retardation. Although an extreme simplification of reality, properties can be assigned such that the final results err on the side of safety (as defined above) and, furthermore, some numerical studies (*eg* Schneider et al.,1997; JNC, 2000) have indicated that, provided this assignment is carefully made, a small number of representative paths can be used to simulate closely radionuclide breakthrough for a more complex network.

In Figure 5.6, matrix diffusion and sorption would be expected to occur with fracture infill material between channels, and within altered and unaltered wallrock. Such supposition has recently been shown to be the case (Figure 5.7) by an unequivocal data set from an *in situ* study by Moeri et al. (2002, Ota et al., 2002). Such "matrix heterogeneity" is a feature of many fractured rocks. Until recently, most radionuclide transport models could not account for this heterogeneous nature of the matrix, and considered only one-dimensional matrix diffusion, in a direction normal to the flow through a fracture or channel, into a homogeneous matrix region. More recently, transport models (*eg* Barten et al., 1998; Vieno and Nordman, 1999) have become available that can account for matrix heterogeneity, thus making fuller use of the type of information on the internal structure of water-conducting features illustrated in Figures 5.6 and 5.7.

5.2.4 Governing equations and solution methods

The governing equations and boundary conditions for radionuclide transport models vary, to some extent, depending on the processes of concern (whether or not is it possible to exclude coupled processes, the transport of radionuclides on groundwater colloids, etc.), and the degree to which detailed models of specific processes are incorporated directly into the transport model. Nevertheless, the starting point in the development of differential equations to describe the transport of radionuclides in geological media is to consider the flux of radionuclides into and out of a fixed fluid volume (or 'element') within the flow domain. A conservation of mass statement for such an elemental volume is:

$$\begin{bmatrix}\textbf{net rate of}\\ \textbf{change of mass}\\ \textbf{of nuclide within}\\ \textbf{the element}\end{bmatrix} = \begin{bmatrix}\textbf{flux of}\\ \textbf{nuclide into}\\ \textbf{the}\\ \textbf{element}\end{bmatrix} - \begin{bmatrix}\textbf{flux of}\\ \textbf{nuclide out}\\ \textbf{of the}\\ \textbf{element}\end{bmatrix} \pm \begin{bmatrix}\textbf{loss or gain}\\ \textbf{due to}\\ \textbf{retardation}\\ \textbf{processes}\end{bmatrix} \pm \begin{bmatrix}\textbf{loss and gain}\\ \textbf{due to}\\ \textbf{radioactive}\\ \textbf{decay and ingrowth}\end{bmatrix}$$

The flux into and out of the element typically arises from advection, mechanical dispersion and diffusion. Retardation processes include sorption and, in the case of fractured media, matrix diffusion.

Where matrix diffusion is considered to provide retardation, two differential equations usually arise, one describing radionuclide transport in the matrix and the other describing radionuclide transport along a fracture or channel itself. A typical form for such equations, in the case of uniformly spaced, parallel fractures in a homogeneous, isotropic matrix, is:

$$\frac{\partial C_f^{(i)}}{\partial t} = \frac{1}{R_f^{(i)}}\left[-u\frac{\partial C_f^{(i)}}{\partial z} + a_L u \frac{\partial^2 C_f^{(i)}}{\partial z^2} + R_f^{(i-1)}\lambda^{(i-1)}C_f^{(i-1)} - R_f^{(i)}\lambda^{(i)}C_f^{(i)} + \frac{1}{b}\varepsilon_p D_p \left.\frac{\partial C_p^{(i)}}{\partial y}\right|_{y=b} \right]$$

for transport in the z-direction along fractures, where, on the right-hand side, the terms within square brackets represent, respectively, advection, hydrodynamic dispersion, radioactive ingrowth and decay, and diffusion into the porous matrix, and

$$\frac{\partial C_p^{(i)}}{\partial t} = \frac{1}{R_p^{(i)}}\left[D_p \left(\frac{\partial^2 C_p^{(i)}}{\partial z^2} + \frac{\partial^2 C_p^{(i)}}{\partial y^2} \right) + R_p^{(i-1)} \lambda^{(i-1)} C_p^{(i-1)} - R_p^{(i)} \lambda^{(i)} C_p^{(i)} \right]$$

for transport in the matrix, where the terms on the right-hand side represent matrix diffusion, and ingrowth and decay.

The parameters are defined as follows:

$C_f^{(i)}$ and $C_p^{(i)}$	are the concentrations in solution, within the fracture and the matrix porewater, respectively, of the ith radionuclide in the decay chain 1 → 2 → 3 → .... → i
$\lambda^{(i)}$	is the decay constant for radionuclide *i*,
$R_f^{(i)}$ and $R_p^{(i)}$	are the retardation factors for advection and dispersion within the fracture and for diffusion within the matrix, respectively,
u	is the water velocity within the fracture,
a_L	is the longitudinal dispersion length within the fracture,
ε_p	is the matrix porosity,
D_p	is the pore diffusion constant within the matrix,
b	is the fracture half aperture,
y_p	is the half distance between parallel fractures.

In general, such equations are solved by numerical techniques, such as the finite element and finite difference methods. Many computer codes have been developed for this purpose. There are, however, a number of analytical solutions for special cases, such as steady-state conditions, which are useful for code verification and for performing simple scoping calculations (see for example, Nagra,1994; Vieno and Nordmann,1999).

5.3 Testing radionuclide transport models: an example from the Swiss radioactive waste management programme

5.3.1 Introduction

Altough the aspect of testing is fundamental to all modelling activities (eg Whicker et al., 1999), special emphasis is put on clear and transparent model testing in repository PA. This is because repositories have to isolate radioactive waste (and some chemotoxic wastes) from the environment for hundreds to hundreds of thousands of years and, for many scientists and engineers, and especially the general public, such time spans are beyond comprehension and, consequently, they have grave doubts as to the safety of any such waste repositories. That repository performance over these long time scales is primarily assessed by the use of complex mathematical PA models only adds to the mistrust of many. How then can people be convinced that it is possible to assess the performance (and thus ensure the safety) of a repository over the long timescales of interest? One way is to address the robustness of the PA models, by clearly indicating the form and extent of model testing carried out within the repository PA. Not only can this show that the individual component parts (such as the models of radionuclide migration in the geosphere) of the complex structure which constitutes most PA models have been checked, but also that the 'mathematical black boxes' constitute an acceptable representation of the repository system.

Part of the problem undoubtedly lies in the unusual nature of radioactive waste disposal: in most major engineering projects, such as bridge construction or aerospace engineering, the designs are tested against a range of laboratory experiments backed up by expert judgement based on experience with the same or similar systems. Here repository design deviates from standard engineering practice in that no high-level waste (and only a few low- and intermediate-level waste) repositories yet exist and, even when they do, testing their compliance to design limits will be, to put it mildly, somewhat difficult due to the time scales involved. In addition, the irrational fear of most things radioactive means that most people require some greater form of 'proof' that a repository is safe than they are willing to accept for other engineered systems such as bridges or aircraft[10]. This being the case, significant additional effort must be expended within the radwaste industry to make it completely clear that the PA models can adequately predict the long-term behaviour (and consequent safety) of a repository. Here, a short overview is presented on the approach to testing geosphere radionuclide migration models in the radioactive waste industry using the Swiss national programme as an example.

5.3.2 Just what does 'testing' imply?

Model testing in the academic world tends to be a relatively relaxed affair. After all, if your model is shown to be 'wrong' or inconsistent with new data, it is hardly the end of the world. Indeed, many would argue that this is simply what good science is all about: puting forward a thesis (in this case your model of radionuclide migration in the geosphere) and having it proven or challenged and replaced by a better, more appropriate, thesis is how science progresses. Unfortunately, in the field of radioactive waste disposal, the ability to make mistakes in the name of scientific progress is simply not acceptable. Not only are there the public fears and misconceptions (noted above) to be dealt with but, because of the seriousness of the job in hand, there are, inevitably, legal requirements to be met. Apart from general legal requirements pertaining to the use of predictive models in environmental protection (*eg* Hagenah, 1999), there are also specific national radioactivity dose requirements to be met (*eg* Frank, 1999). Consequently, a more rigid set of testing guidelines has developed and these are outlined below.

5.3.2.1 Testing: just what do we do?

The direct testing of the results of a geosphere radionuclide transport model, as used in PA, is impossible, due to the scales of space and time involved. Rather, confidence is developed through the consistency of the model assumptions, and associated databases, with a large number of diverse observations and experiments. Furthermore, the model should have the capability to make predictions, or at least bounding estimates, that can be tested, even if the scales differ from

[10] Consider the case of a fully loaded Boeing 747-400: despite having some 6 million parts, flying at 10,000m at 900kmh^{-1}, controlled by a crew, several computers and air traffic controllers, it has full acceptance from the public - despite the massive consequences of catastrophic failure (to the passangers).
For a radioactive waste repository: despite having very few main parts, being immobile, needing no control, it has no acceptance from the public - despite the fact that the consequences of even catastrophic failure will be small.

those relevant to PA. In the case of a geosphere radionuclide transport model, testing needs to address:

- the adequacy of the catalogue of relevant processes incorporated in the model;
- the adequacy of data-collection techniques and the transferability of laboratory data to in situ conditions;
- the understanding and modelling of individual processes;
- the modelling of combined processes in the transport model.

In situ field experiments, with supporting laboratory programmes, can be used to address all of these issues. It should, however, be noted that there may be many specific differences in the structures that are relevant and the parameter values used depending on whether a model is applied in PA or to the modelling of a field experiment. For example, in a field experiment, the focus may be on an individual feature rather than on a network of water-conducting features and the scales may differ - most field experiments are carried out over distances of metres to tens of metres whereas distances of 100s to 1000s of metres are more applicable to a repository PA. The timescales of an *in situ* experiment also differ from that of a repository PA (hours to months compared to hundreds to hundreds of thousands of years).

In addition, because of the large number of simplifications which have to be made for the PA calculations, the PA codes themselves cannot be directly tested: rather a sub-model (also known as a research model) of the PA model is used. The research model differs from the PA model only insofar that features and processes relevant to field and laboratory tests are also considered (such as small-scale porosity differences or kinetic effects during sorption) rather than only those relevant to the scales of a repository. Otherwise, the modelling approach outlined above applies equally in both applications. Thus, having achieved confidence in a research model through predictive model testing, an PA model developer is in a strong position to decide where simplifications can be justified in terms of either conservatism or insignificant effects.

As an example of research model testing, a series of experiments carried out in one of Nagra's underground laboratories (the GTS) in the central Swiss Alps (see Figure 5.8) will be examined. Nagra began work on transport modelling at the GTS in 1985 with the hydrogeological characterisation of a water conducting shear zone in granodiorite (marked RRP in Figure 5.8) and has continued, along with JNC (Japan Nuclear Cycle Development Institute), with a large series of *in situ* radionuclide tracer migration experiments. The radiotracers were injected into an artificially controlled flow-field in a complex fracture zone (Figure 5.9) and the characteristic breakthrough curves of the tracers (see Figure 5.10) were modelled in a multi-step process (described below).

The behaviour of the radiotracers used increased in complexity with time from simple, non-sorbing tracers (fluorescein dye, ^{82}Br, ^{123}I, ^{4,3}He and ^{3}H) through various weakly sorbing tracers (^{22}Na, ^{24}Na, ^{85}Sr and ^{86}Rb) to a long-term experiment with strongly sorbing ^{137}Cs (see McKinley et al., 1988, Alexander et al., 1992 and Smith et al., 2001b, for details). Most recently, chemically complex tracers (^{99}Tc, ^{113}Sn, ^{75}Se, ^{234}U, ^{235}U, ^{237}Np, ^{60}Co, ^{152}Eu and stable Mo) have been utilised followed by the physical excavation of a part of the shear zone to recover these strongly retarded radionuclides (Alexander et al., 1996, 2001, Ota et al., 2001, 2002 and Moeri et al., 2002). These radiotracer tests continue at present in the GTS with two ongoing experiments, one examining the effects of colloids produced during the degradation of the man-made barriers on the migration of a suite of radionuclides and a second looking at the interaction of radiotracers with secondary cement phases produced in the geosphere by reaction with hyperalkaline cement leachates (see Kickmaier et al., 2001, for details).

5.3.2.2 The methodology of model testing

The following sequence of tests are applied in the GTS projects to research models:

1. Determination of Parameter Values.

The determination of parameter values describing rates and spatial extents involves an assessment of all the information available from the characterisation of the rock, from field and independent laboratory experiments, such as batch sorption and laboratory diffusion experiments and from natural analogues. These data may need to be interpreted by means of supporting, interpretative models (e*g* a linear or non-linear model of sorption) in order to provide parameter values for a transport model. If all parameters of a transport model can be determined in this way, then the model can be used directly in predictive testing (see below). There may be some parameters, however, that cannot be taken directly from field and laboratory measurements, although it is often possible to place bounds on these parameters by drawing on wider scientific understanding (*eg* from relevant literature or experience in similar rock types or hydrogeological environment). It is then necessary to resort to inverse modelling to "calibrate“ the model, providing parameter values for subsequent application in predictive modelling.

Inverse modelling involves the following steps:

- all „free“ parameters that are not fixed by means of independent field and laboratory measurements are adjusted simultaneously (constrained within certain bounds, if these are known) until the best fit to a tracer-transport breakthrough curve (see Figure 5.10) is obtained,
- the best fit model curves are compared with experimental curves and assessed against success criteria,
- if the comparison is unsuccessful, then either the derivation of fixed parameter values or the model formulation itself must be re-assessed,
- if the comparison is successful, then the fitted parameters are tested for consistency with independent data, *ie* from the characterisation of the rock (*eg* Figure 5.7) and from other field and laboratory experiments
- if there is inconsistency with independent data, then either the derivation of fixed parameter values or the model formulation itself must be re-assessed,
- if the fitted parameters are consistent with independent data, then they can be taken into account when assessing available information:
    - to reduce the number of free parameters in further inverse-modelling exercises (*eg* with more complex tracers, as in the GTS experiments)
    - to determine whether free parameters can be eliminated and predictive modelling (*eg* of new experimental set-ups, with different flow fields, as in Figure 5.10) is possible.

2. Predictive (or blind) modelling

This type of model testing involves the prediction of experimental breakthrough curves **in advance of the experiments** for a range of tracer experiments carried out under different flow conditions (pumping rates, separation of injection and withdrawal points, *etc*) and using different tracers.
For each predicted (model) break-through curve:

- comparison is made of the model and experimental curves, with assessment against success criteria (based on an evaluation of experimental errors),
- if the comparison is unsuccessful, then either the derivation of fixed parameter values or the model formulation itself is re-assessed,

following successful comparison, selection of a new experimental set-up (with different flow conditions and/or tracers with different transport-relevant properties) for further predictions is carried out and the procedure is repeated until it is judged that "sufficient confidence" in the model has been attained.
"Sufficient confidence“ is not precisely defined. The aim, however, is to model a large number of experimental set-ups, since a "good model“ should describe a large class of observations. The ability of the transport model to predict the behaviour of a variety of tracers in a range of different flow fields gives confidence that the model can be applied to the transport of radionuclides in a performance assessment, provided adequate characterisation of the repository system and suitable independent experimental data are available.

Such predictive modelling, in particular, provides a sensitive test of the model concept and the numerical procedures precisely because the modeller is given only limited information. The method of testing is crucial: few people, even those involved in the disposal of radioactive waste, fully appreciate the difference between blind testing of model *predictions* and testing if a model can *simulate* particular observations - as can be clearly seen in the literature. This is a crucial point, as noted by Pate et al. (1994) "This aspect of blind (*ie* predictive) testing is particularly important as, in many cases, the manner in which the simulation is carried out can be very objective and, if the ‘answer’ is known, can be biased either consciously or subconsciously." In a repository PA, simulation of data brings little or no confidence that the models involved can later predict repository evolution: confidence can be much better built by carrying out a series of predictive modelling exercises followed by experimental runs and a final assessment of the accuracy of the predictions (and, where necessary, improvement of the models).

In addition, to ensure that the tests are scrupulously fair (and to further increase confidence in the tested models), it is strongly recommended that the predictions are published before the experimental runs are carried out in the field (or laboratory). In the GTS work, unfortunately, the predictions were only published in internal memos but open publication of predictions (in the scientific literature) has been carried out in two tests of thermodynamic databases for PAs[11].

5.3.2.3 The value of field experiments in model testing

[11] In a study of a natural cementitious system, the modelling teams first predicted the behaviour of the elements of interest in the system and only then was the measured data revealed for comparison. See McKinley et al. (1987, 1988) and Bath et al. (1987a,b) for the details. This was the first comprehensive blind predictive PA modelling exercise and the procedures developed for this study have formed the basis for most later studies (see Miller et al., 2000 and Bruno et al., 2001,for additional examples). However, in only one other case has the predictive work been published openly before the full analysis was carried out (see Alexander, 1992 and Alexander et al., 1992).

Given the scales of time and space that must be considered in a PA, direct testing of the realism (or the degree to which the models err on the side of safety) of a model in the system of interest is impossible. Field experiments, such as those performed at the GTS, are consequently invaluable in that:

- the fundamental transport processes that operate in the system are expected to be the same or similar to those relevant to any fractured repository host rock;
- the structures present, though differing in detail, are also similar enough to those of potential fractured repository host rocks;
- the scales of time and space over which the experiments operate, though often considerably shorter than those of a PA, are often larger than those achievable in the laboratory;
- the degree of characterisation of the system that is possible is greater than that achievable at a repository site (due to the smaller spatial scales involved in many field experiments and the need, at a repository site, to avoid perturbing the favourable properties of the site by sticking too many holes in it);
- field experiments, being performed in situ, are less subject to some experimental artefacts than laboratory experiments.

However, it must be emphasised that the successful modelling of radiotracer tests (whether those in the GTS or in other *in situ* tests such as at the Aspö underground rock laboratory in Sweden (*eg* Winberg et al., 2000) or the Whiteshell underground rock laboratory in Canada (*eg* Vilks and Bachinski,1996) gives support to the model representation of structures and processes that exist in, or operate on, spatial and temporal scales that are similar to, or smaller than, the tests themselves. A major difficulty with the tests lies in the extrapolation of this conclusion to the larger scales that are relevant to PA: no information is provided on processes that, though irrelevant on the spatial and temporal scales of field tracer tests, may be important over scales relevant to PA[12]. The tests can, however, add to the body of observations and experiments with which the model is consistent and thus build confidence in its application in PA. This can be further strengthened by carrying out a series of complementary and inter-related experiments as has been the case at the GTS (see Kickmaier et al., 2001, for details) and backing these up with additional work in the laboratory and other natural systems (see also the comments on this approach in Alexander et al., 1998).

5.3.3 Model simplification and testing in performance assessment

The issue of model simplification (or 'abstraction' in PA modelling terms) has been widely discussed in the context of confidence in the long-term safety of repositories for the deep geological disposal of radioactive waste (see, for example, NEA 1999, 2000, 2001). In spite of

[12] It is, of course, possible that different model representations of the structures and processes relevant to the GTS radiotracer tests would be similarly successful fitting the results of (at least some) tests and, for this reason, alternative models were developed by three independent research groups. The aim was, by attempting to falsify some of these alternatives, and, in particular, by making independent predictions of the results of tests in advance of the tests being performed, to narrow down the range of conceptual model uncertainty and to identify the processes that are most important in describing solute transport. This aim was only partially fulfilled, since: 1) The differences between the models were rather minor and thus the range of conceptual-model uncertainty was small. All models were built upon the dual-porosity concept, and no attempt was reported, for example, to understand the test results without invoking matrix diffusion. 2) New flow path structural data produced several years after the model testing was completed included several features (multiple flow paths, diffusion into fracture fill material) which had been explicitely excluded from the modelling approach. This shows that the models tested were insensitive to the fine detail in the flow path (which builds confidence in the overall PA approach) but also that the models, to put it bluntly, got the right answer for the wrong reasons (which is less good for confidence building).

recent refinements, the complexity of many geosphere transport models is still less than the state-of-the-art in mathematical modelling and the understanding of fundamental transport processes would permit. It would, for example, be possible in principle to incorporate a great deal of geometrical and process detail into these models. The main reasons why this is not more widely done in practice are:

- Lack of data: characterisation of the geological environment is a stepwise process, and understanding evolves as characterisation moves from surface-based studies to more detailed studies underground. PAs are carried out periodically throughout this process, and inevitably have to cope with uncertainties associated with incomplete characterisation and so have to utilise models which, in terms of their degree of realism, are often a step behind the cutting edge of geological characterisation. It should also be noted that there are practical limits to the degree to which characterisation can be carried out without disrupting the favourable characteristics of a rock by simply drilling too many holes in the repository host rock.
- A desire for clarity in performance assessment: a single, complex model that attempts to include all features and processes runs the risk of being used as a "black box", which produces results in a manner that few people understand and which is difficult or impossible to check. Many parameters of an "all-inclusive" model might be poorly supported by site-characterisation and other evidence and the fact that a result could nevertheless be obtained may give some people a false confidence in its meaning.
- Lack of communication: whilst initiatives such as those of the NEA (1997, 1998, 1999, 2001), which have attempted to involve as wide a range of disciplines and promote as much discussion as possible, are to be applauded, it remains a sad fact of life that inter-disciplinary communication remains problematic. Even within a single national waste programme, it is always possible that simply too much information exists and so dichotomies arise. For example, in the Swiss programme in the 1990s, while the field exploration groups and the PA modellers were working well to keep each other abreast of new developments in geological and modelling efforts (see Mazurek et al., 1992), there was simultaneaously a break down in communication between the above groups and those working in the GTS, leading to lost chances to employ the most up-to-date geological data in the ongoing PA (see comments in Alexander et al., 1997).

However, for PA modellers, these points may not be such a great problem due to the inherent simplifications necessary in carrying out PA calculations. The fundamental problem is that, to be acceptable in a repository PA, all models (and databases) employed must be 'validated' (in the IAEA sense of demonstrating that they are appropriate for the use envisaged). The whole process of validation remains highly controversial and here is not the place to discuss it in detail (instead, see Pescatore, 1995, and references therein), rather accept that there are often regulatory requirements to address (addressed, for example, in Frank, 1999, for the Swiss radioactive waste programme and defined in Luiten, 1999, for the European Union's integrated environmental assessment).

What is important to note is that validation is inherently subjective due to the difficulties in quantifying all the uncertainties in a slowly evolving repository and the host rock over geological time scales. This means that it is important that the validation procedures are carried out in a transparent and logical manner (hence the development of the blind testing approach detailed in section 5.3.2.1). Thus for any PA model, the testing scheme can be focussed on answering the following questions:

- is the application clearly defined and scientifically reasonable?
- are the theoretical arguments/models used defensible?
- what extent of validation is required to show that the models err on the side of (repository) safety by over-estimating radionuclide transport in the geosphere?
- are the selected data consistent with laboratory experiments, in situ studies and data from natural systems?

It is clear that some aspects are too detailed to include in the models (see comments above on the representation of sorption or small-scale structural information) and so, where simplification is necessary, it must be demonstrated that the models will over-estimate radionuclide migration.

Thus this model simplification (although, arguably, not scientifically rigorous in an academic sense), when it can be justified on the grounds of erring on the side of safety, is widely accepted by repository developers and by regulatory authorities. It is also generally accepted that numerical models that can be demonstrated to be either capable of at least setting upper limits on processes (such as estimating the maximum likely distance a given radionuclide will migrate through the geosphere) or to be erring on the side of safety (by using the worse case scenarios in any calculation) can yield results that are suitable for decision making (*ie* is the likelihood of substantial radionuclide migration in the geosphere high?). It may also be possible to demonstrate, using independent calculations or qualitative arguments, that certain features or processes have a negligible impact on radionuclide transport. Such arguments might, for example, be used to evaluate the importance or otherwise of coupled phenomena in argillaceous media (see, for example, Soler, 1999) or, as noted above, the relevance of matrix diffusion in a particular fractured rock.

It has been pointed out that, although a degree of model simplification is inevitable and may even be desirable, simplification that leads to a clear loss of realism should be avoided (NEA, 1999, 2000). Confidence requires that PA models incorporate the essential features of the system, based on site understanding and conceptual models that are consistent with wide-ranging observations and evidence, even though all such information cannot always be incorporated directly into the models and databases (see comments above). Models of groundwater flow, for example, can be tested for consistency with the distributions of various natural tracers (salinity, heat, etc.), even though these distributions are not direct input to any given PA model for a repository site (NEA, 2000).

5.4 Overview of the state-of-the-art and directions for the future

5.4.1 Interpretation of radionuclide distributions

Despite extensive efforts over the last three decades, the "Holy Grail" of a model which allows observed distributions of radionuclides in geological systems to be interpreted quantitatively in terms of the processes influencing their transport and immobilisation seems far from reach in most cases. Acknowledgement of the difficulties involved and rejection of overly-simplistic models must, however, be recognised as a significant advance.

Only rarely now are natural radionuclide concentrations in groundwaters interpreted directly in terms of a "groundwater age". Instead, information from a wide range of radioisotopes is combined with that from hydrochemistry, mineralogy, stable isotopes, structural geology etc. to derive one or more scenarios of groundwater evolution (see, for example, Edmunds and Smedley, 2000 and Castro et al., 2000). The radioisotope data themselves can be extensively refined to quantify potential perturbations from sampling, diversity of sources and rock / water interaction history (see Pearson et al., 1991, for examples). This requires not only more sophisticated models, but much more extensive analysis of very carefully obtained sets of samples.

This trend can also be seen in the interpretation of tracer tests – carried out in laboratories, drillholes or underground test facilities such as the GTS. Simple advective flow/Kd models have been replaced by interpretations which emphasise detailed characterisation of both the flow system on a micro scale and the diverse range of processes which can influence retardation (various types of sorption, matrix diffusion, colloids, organics, reaction fronts, etc.). Very simplistic "tracer tests" have evolved into integrated, multi-disciplinary migration studies which include laboratory, in-situ, modelling and analogue components and may run on a timescale of decades at costs of tens of millions US$ (see information in Alexander et al., 1997).

One specific area which seems to have come to a dead end involves attempts to derive key radionuclide transport parameters by interpretation of ratios of several natural series radionuclide isotopes (so-called "in-situ Kds"). Despite grandiose claims for this methodology and extensive, often acrimonius, debate in the literature, there is no evidence that any such approach provides sensible results. Indeed, "reality checks" on such models often clearly demonstrate that they are incorrect – for example, when they predict orders of magnitude differences in sorption for different isotopes of the same element (McKinley and Alexander, 1996, 2002 and Ku et al., 1998).

5.4.2 Prediction of radionuclide migration

As discussed in 5.2, for specific applications, the requirements in terms of accuracy/precision may be so limited that prediction of future radionuclide migration which err on the side of safety may be easier to meet than those for the interpretative studies considered in 5.4.1 above. In particular, many concepts for deep disposal of radioactive waste are so over-designed that uncertainties of several orders of magnitude in performance of the geosphere barrier (*ie* how well the geosphere retards radionuclides) are quite acceptable. For example, as shown in Figure 5.12, different representations of retardation of a key radionuclide from high-level radioactive waste (^{135}Cs) change its calculated release by more than 5 orders of magnitude. Nevertheless, even the worst model representation meets regulatory requirements and hence there is no driving need to develop a more realistic model which would undoubtedly give better performance.

Although performance assessment of such deep geological repositories has been a driving force for development of the methodology for radionuclide transport prediction, such over-conservatism is not acceptable for many other applications. The need for greater realism requires that uncertainties and complexities need to be addressed directly, rather than compensated for by simplification which errs on the side of safety. As yet, progress in this area has been very limited – constrained both by poor availability of the databases needed to apply more sophisticated models and the lack of systems which allow resulting models/databases to be adequately validated (see, for example, comments in Bruno et al., 2001). At present, it must be acknowledged that the models used to support near-surface waste disposal or contaminated site remediation are generally very simplistic in their treatment of solute release while transport processes are often based on purely empirical models.

5.4.3 Conclusions

Understanding of the processes of mobilisation and migration of radionuclides in deep geological environments has advanced considerably over the last couple of decades – even if most progress has been in the recognition that transport processes are much more complex than previously thought. Advances in computer hardware/software have effectively removed the inability to simulate defined systems as a constraint – the main stumbling block is characterisation of the system of interest in sufficient detail (especially if it is the site of a deep geological repository).

For specific applications, fundamental concepts are already well established and practical methodologies are available – *eg* groundwater dating, prediction of the migration of simple solutes in saturated, porous media. In other cases, it may be acceptable to cover for uncertainties by always erring on the side of safety if the requirement is only to put bounds on maximum possible extent of mobilisation.

Despite what is written above, it could be argued from an academic viewpoint that it is necessary to develop more realistic models for both interpretation of existing radionuclide distributions or prediction of migration in more complex solutes in more complicated and heterogeneous rocks. Some advances in conceptual understanding might be needed (*eg* for partitioning of natural series isotopes, treatment of colloids, description of reaction fronts, *etc*) but the main rate determining step will probably be provision of better databases from sophisticated experimental projects. Practical applications also require rigorous testing of the model/databases used – which becomes increasingly difficult as the systems represented become more complex.

5.5 References

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	POTENTIAL GRADIENT			
FLUX	**Temperature**	**Hydraulic**	**Chemical**	**Electrical**
Heat	Thermal conduction (Fourrier's Law)	Thermal filtration	Dufour effect	Peltier effect
Fluid	Thermal osmosis	Advection (Darcy's Law)	Chemical osmosis	Electrical osmosis
Solute	Thermal diffusion or Soret effect	Hyperfiltration	Diffusion (Fick's Law)	Electrophoresis
Current	Seebeck or Thompson effect	Rouss effect	Diffusion and membrane potential	Electical conduction (Ohm's Law)

Table 1: The Onsager matrix of direct (diagonal) and coupled (off-diagonal) transport processes (from Horseman et al., 1996)

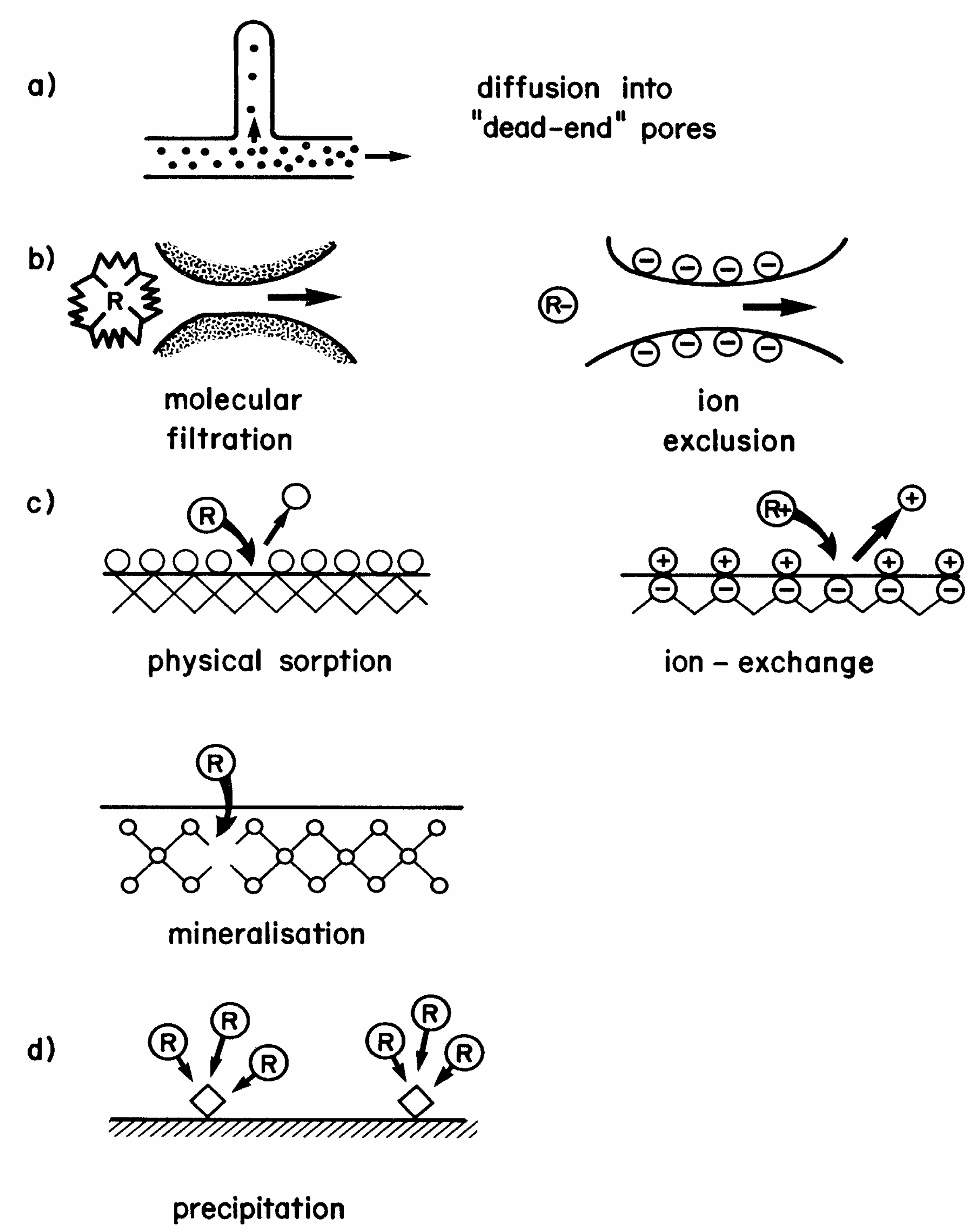

Figure 5.1 The retardation mechanisms that may affect radionuclides in the geosphere (after McKinley and Hadermann, 1984)

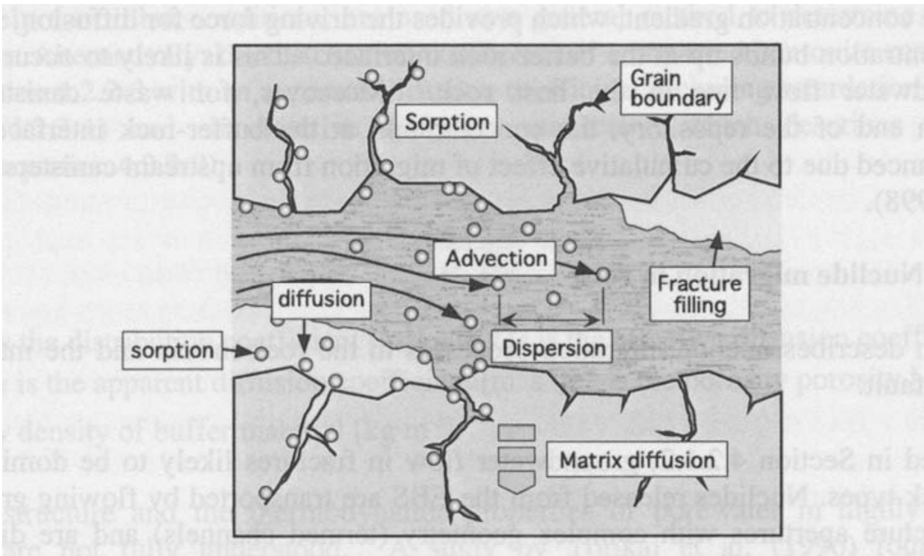

Figure 5.2: Schematic illustration of transport processes in and around a water-conducting feature (after Figure 4.2.2-1 in JNC, 2000)

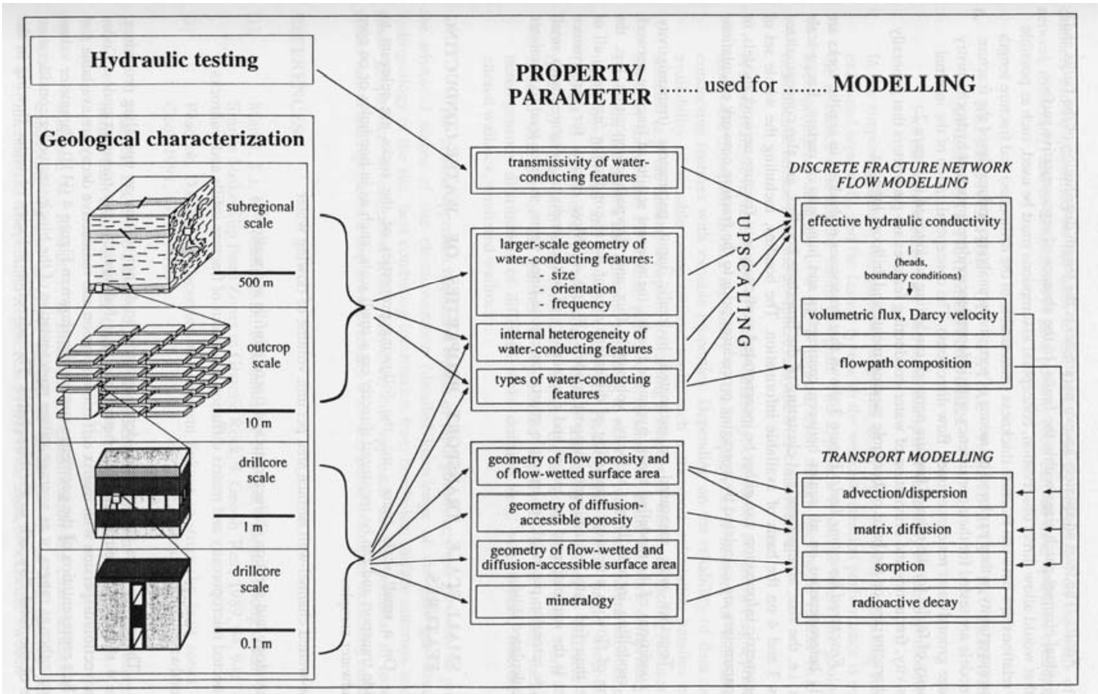

Figure 5.3: Overview of field-derived parameters used for radionuclide transport modelling and flowchart of data derived on different scales, using limestone beds of the Palfris Formation, Wellenberg, Central Swiss Alps, as an example (from Mazurek et al., 1997)

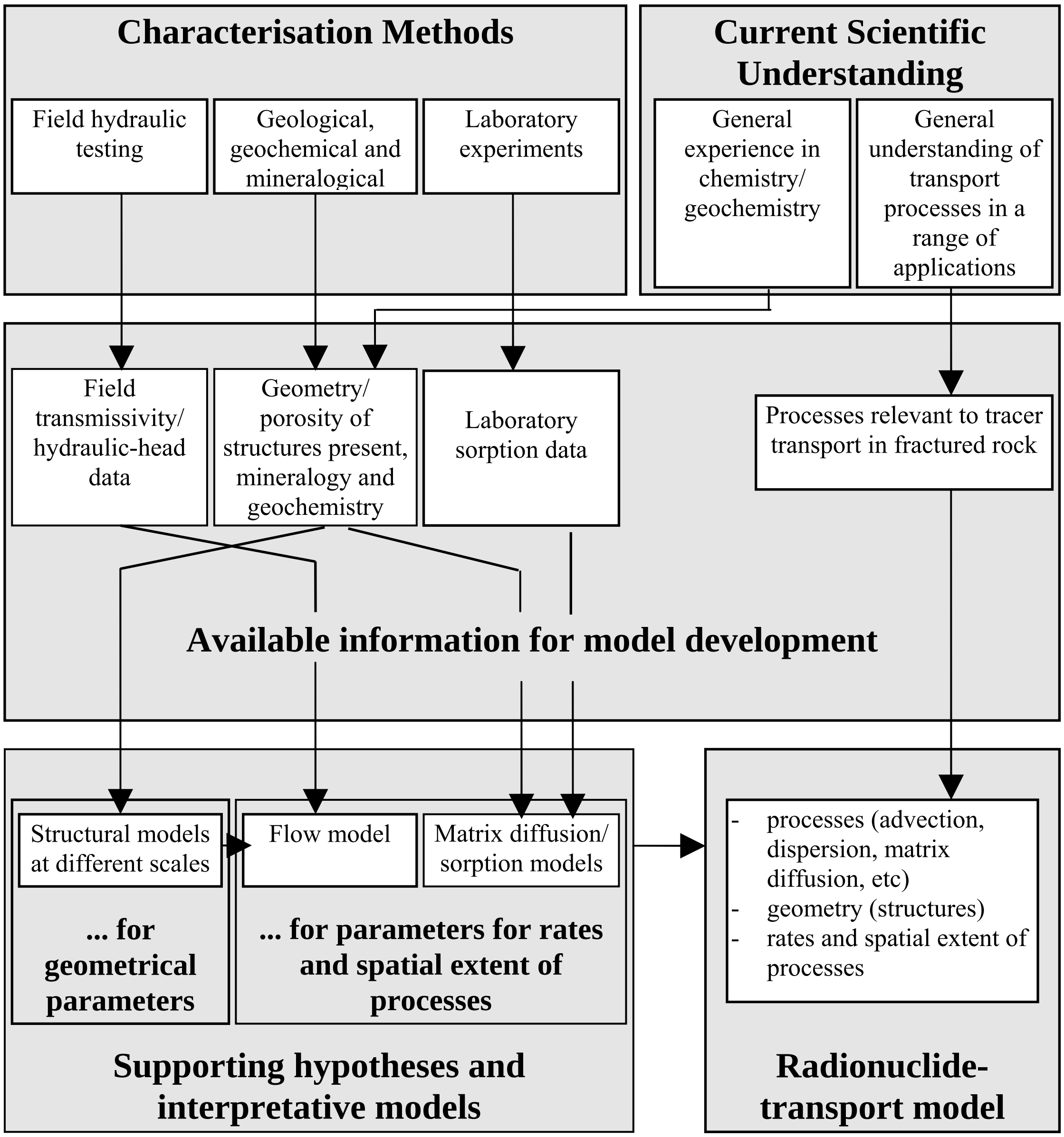

Figure 5.4: The use of supporting hypotheses and interpretative models to assess field and laboratory data in terms of input parameters of a transport model (from Smith et al., 2001b).

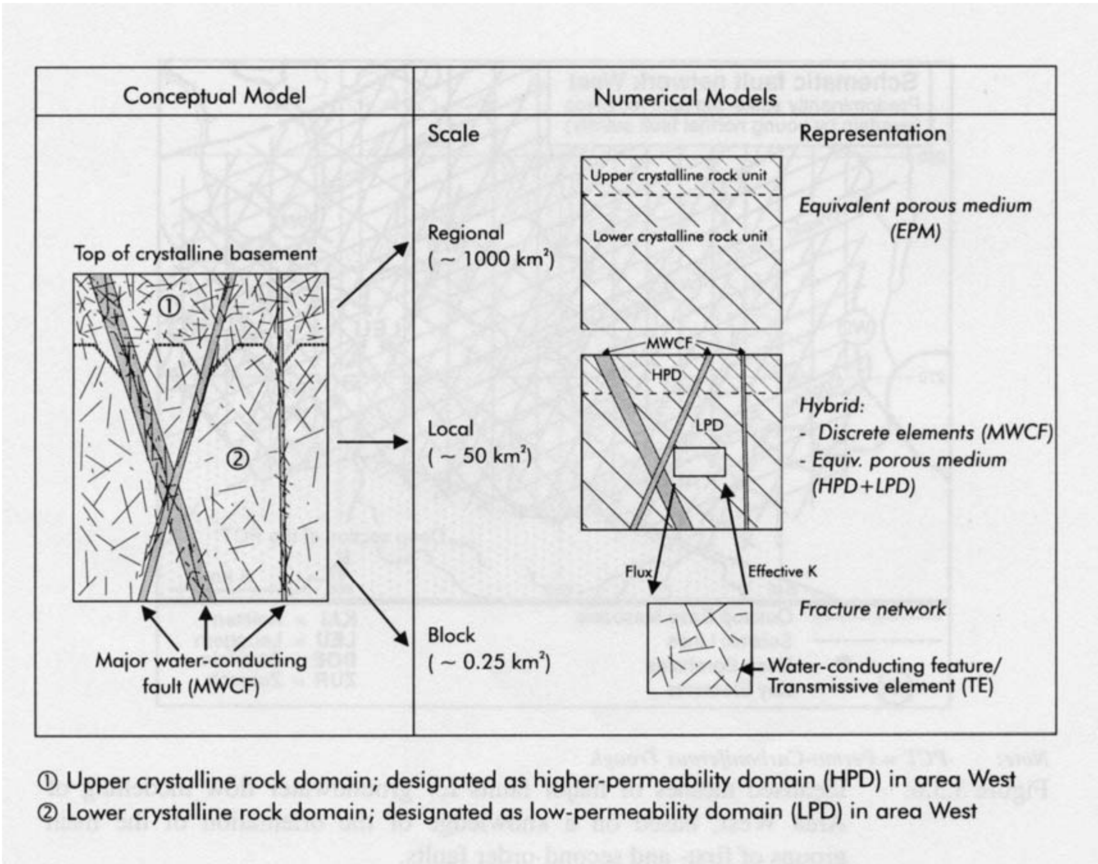

Figure 5.5: Schematic representation of the relationship between a conceptual model of geological structures (in the crystalline basement of Northern Switzerland) and numerical models of groundwater flow at regional, local and block scales (from Figure 3.5.5 in Nagra, 1994).

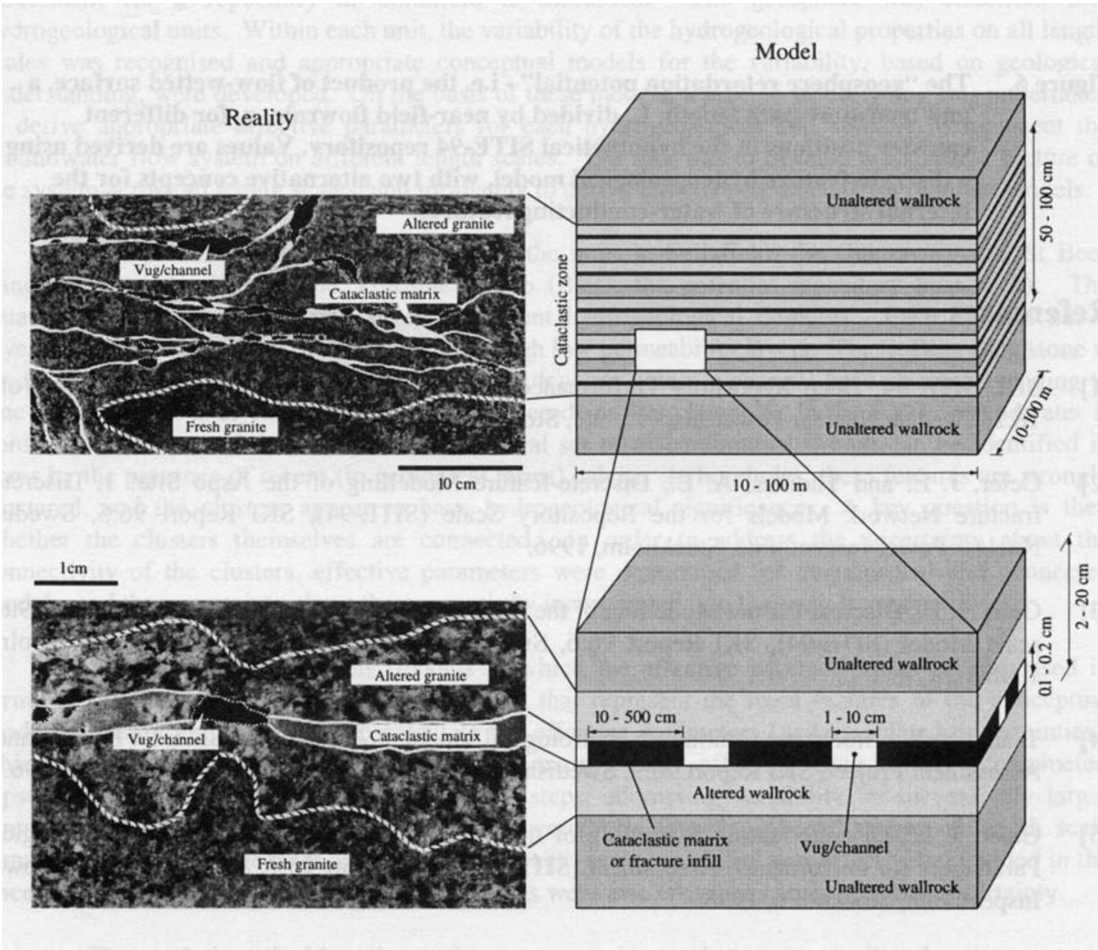

Figure 5.6: Tectonically disturbed zones in the crystalline basement of Northern Switzerland, as observed in drillcores and as simplified for the purposes of transport modelling (from Figure 3.5.4 in Nagra, 1994)

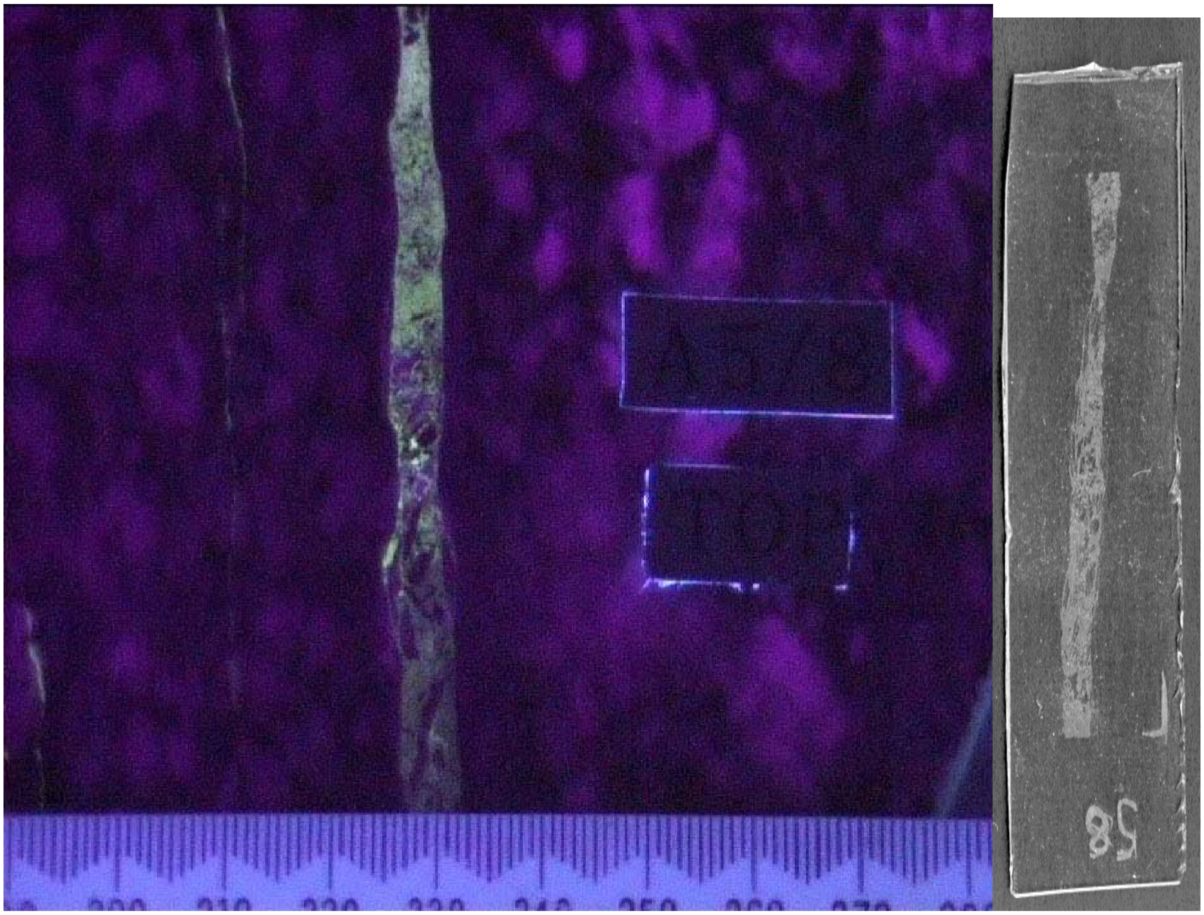

Figure 5.7: Clear examples of radionuclide diffusion into and sorption onto fracture infill and wallrock in granodiorite from Nagra's Grimsel Test Site, Switzerland (from Moeri et al., 2002). The sample has been impregnated *in situ* with fluorescent resin and then over-cored and sawn into slices for examination.
a) On the left: fault filled with fault gouge (yellowish colour) and fragments of wall rock (black) floating in the fault gouge. UV light, partial scale below in mm divisions.
b) On the right: alpha-autoradiograph of part of the sample on the left showing uptake of radionuclides in the fault gouge (white material) and on the fracture surface (or wallrock) and on the surface of the rock fragments (grey areas).

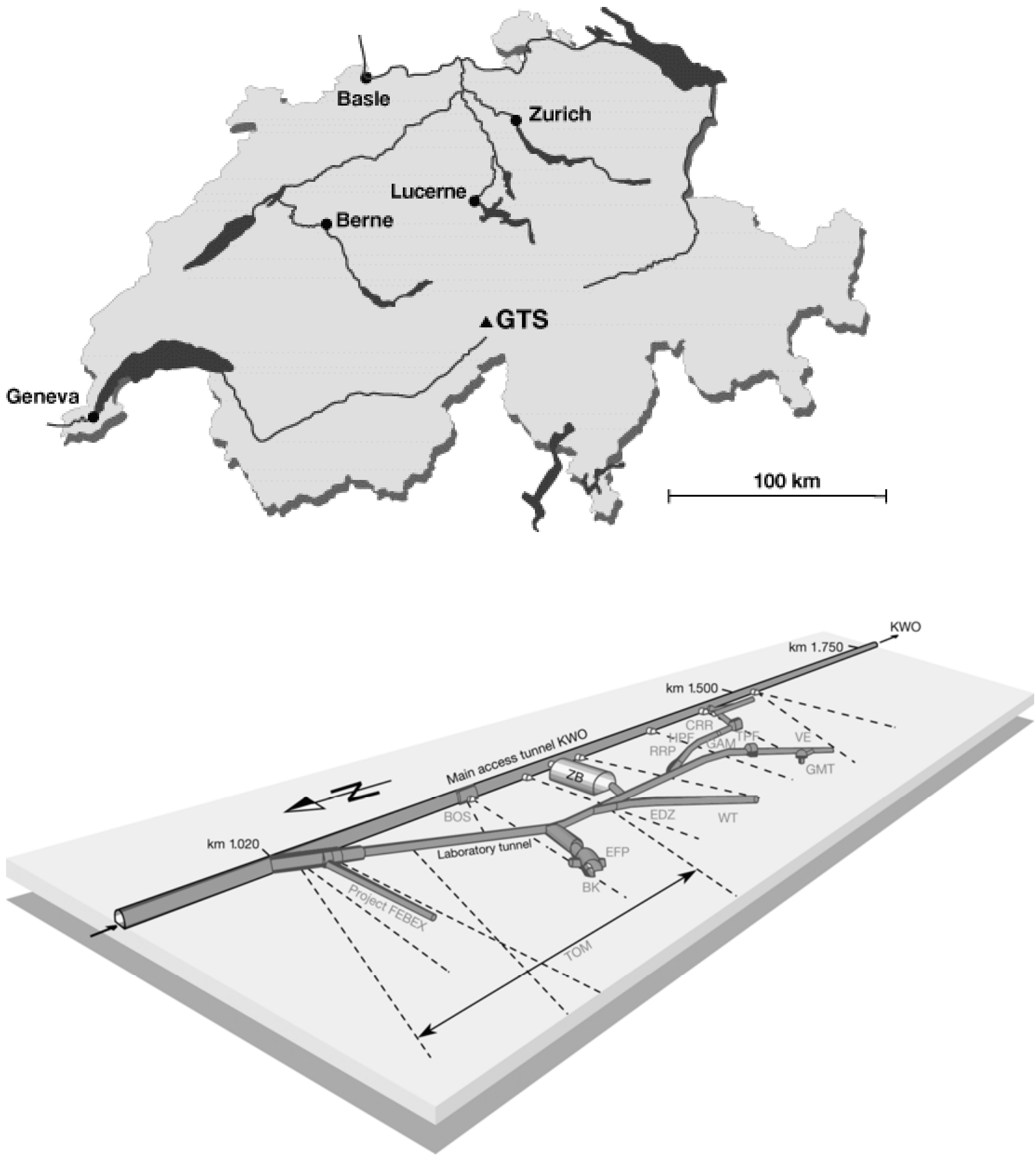

Figure 5.8: a) Map of Switzerland. Nagra's crystalline underground laboratory, the Grimsel Test Site (GTS), lies in the central Swiss Alps.
b) Overview of the GTS tunnel system. The experimental site is marked RRP/HPF/CRR denoting a long series of experiments with radiotracers (for details see Ota et al., 2001)

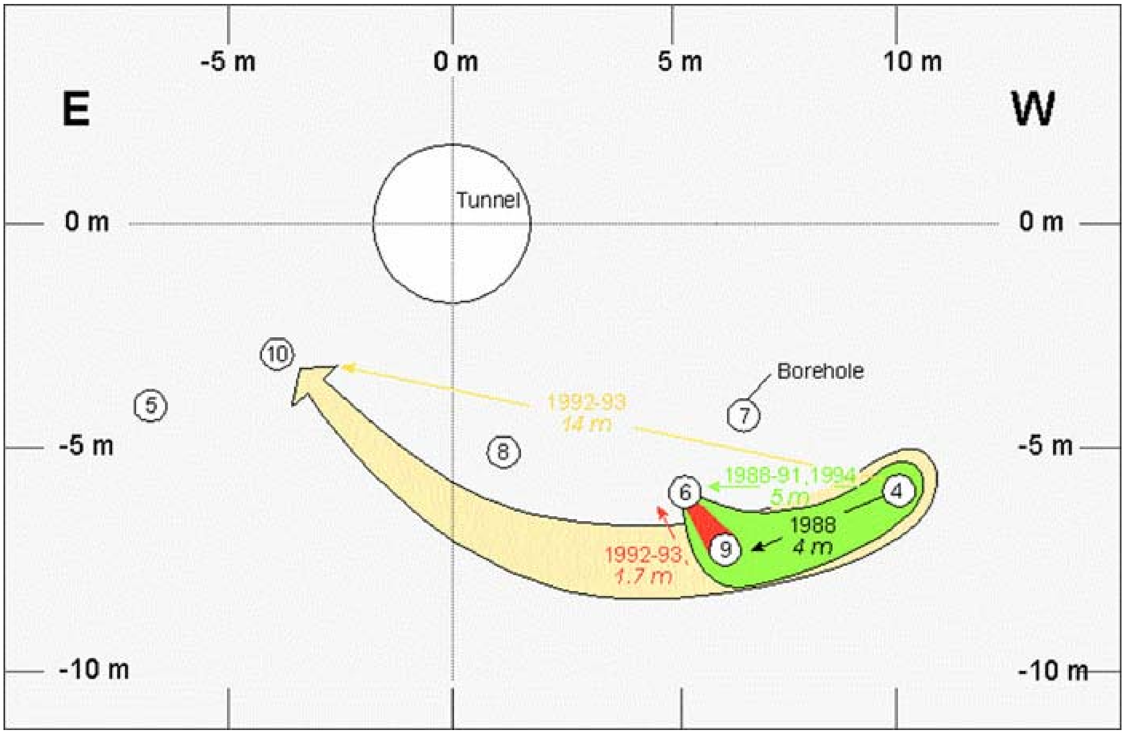

Figure 5.9: Examples of dipole flow-fields in the experimental shear zone in the Grimsel Test Site (the shear zone lies in the plane of the paper). Radiotracers are pumped into the injection borehole (*eg* hole 9) and water (and radiotracer) is pumped out of the outlet borehole (*eg* hole 6) at 10-20 times the injection rate, so ensuring a 'closed' flow system (*ie* the radiotracers remain within the artificially constrained flow system). This has the benefit of simplifying matters for the hydrogeological modellers and ensures full recovery of the radiotracers. Note the deviation from the theoretical straight line flow between boreholes (due to the influence of both the detailed structure of the shear zone and the natural flow gradient).

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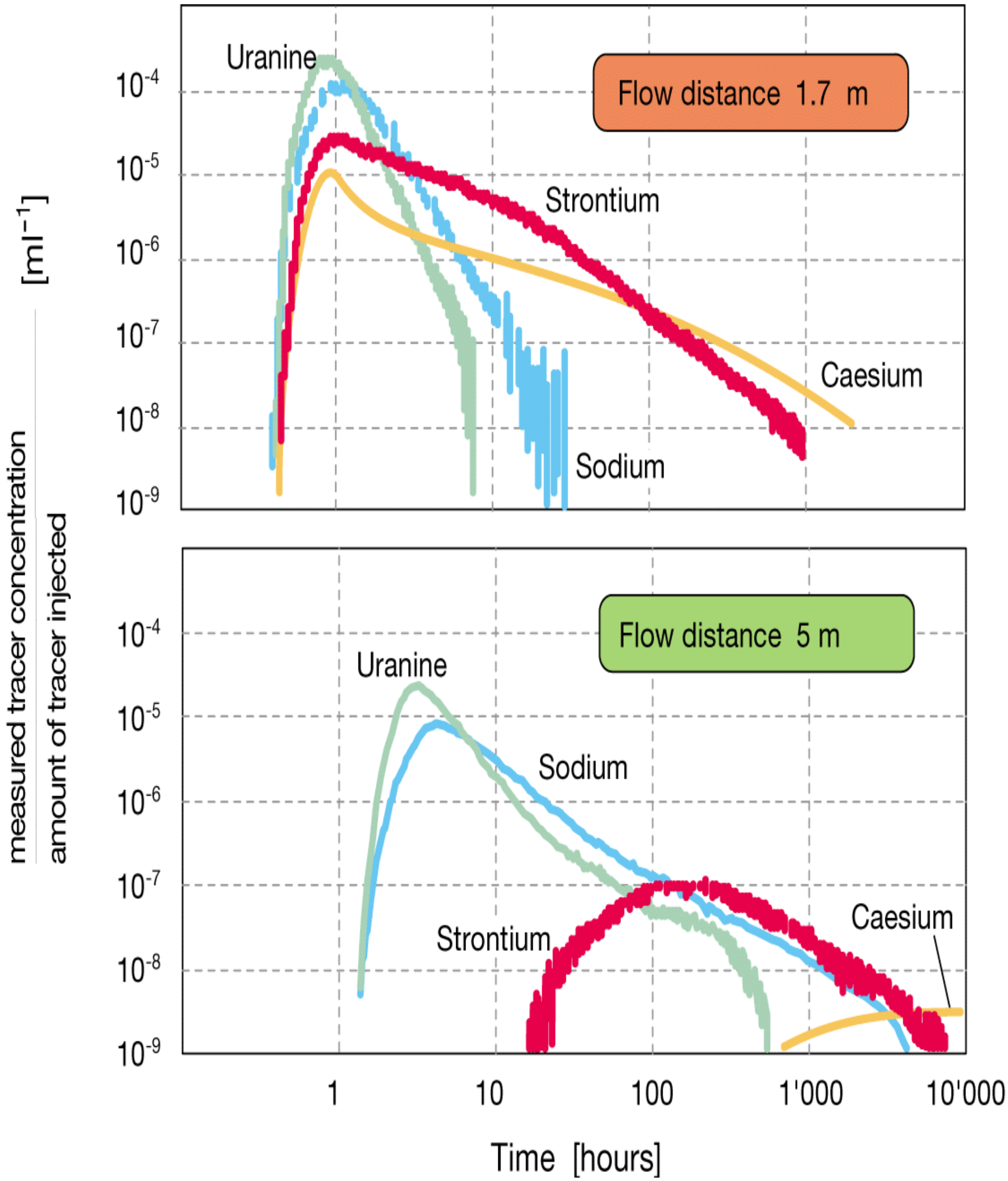

Figure 5.10: Examples of measured breakthrough curves from the MI experiment (*cf* Figure 5.10). The 1.7m flow distance relates to the boreholes 9-6 flow field and the 5m to the 4-6 flow field.

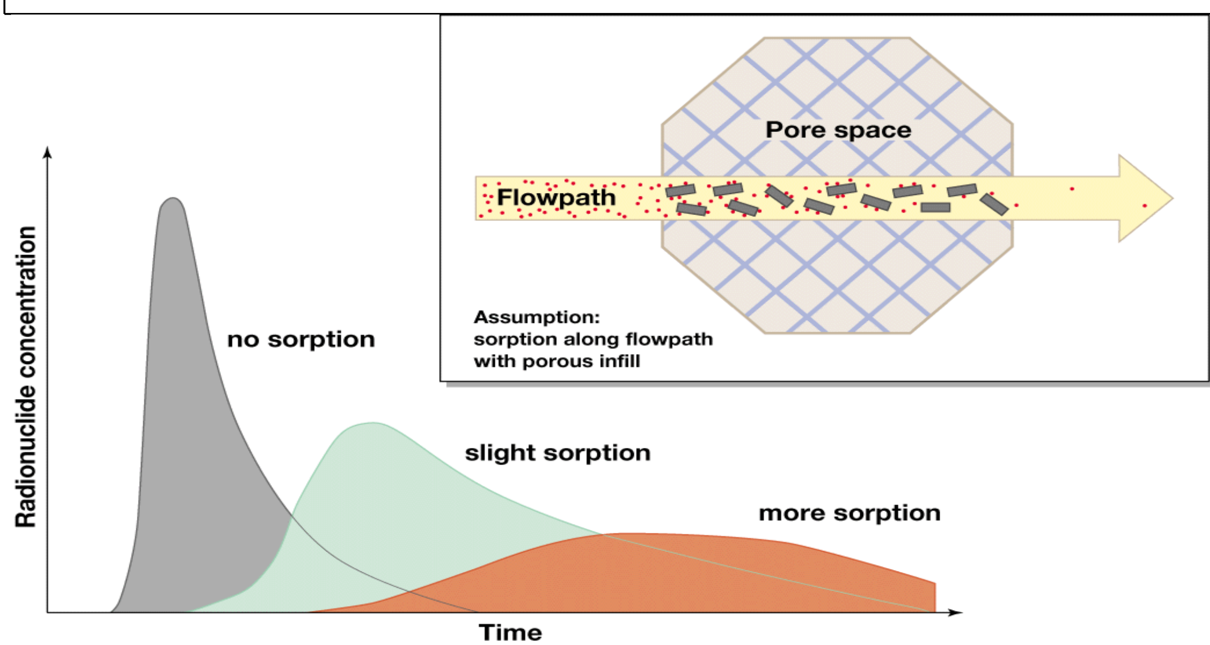

Figure 5.11: Illustration of the effects of different retardation mechanisms on radionuclide tracers in the geosphere (*cf* Figure 5.9). As noted above, the precise form of the breakthrough curves allows some deductions to be made as to the relative influence of the various processes (matrix diffusion, channelled flow etc) occuring in the shear zone. See also Smith et al. (2001a) for comments on the limitations of this methodology.

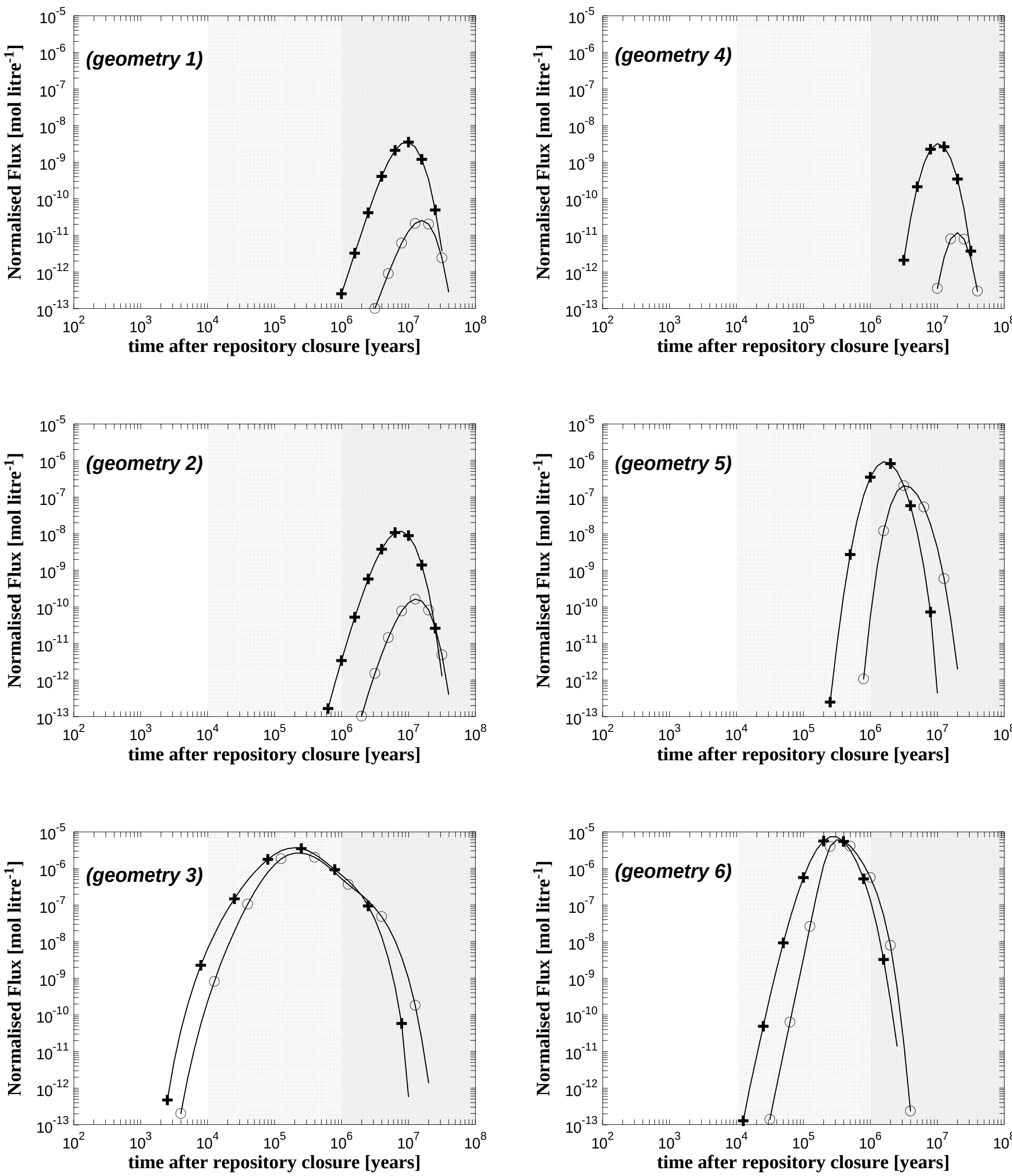

Figure 5.12: An example of the variation in the calculated retardation of ^{135}Cs in the geosphere. Here, six different representations were used to illustrate the sensitivity of the modelling concept to fracture geometry.

Results calculated with non-linear sorption incorporated explicitly ( ) are compared with those calculated with an effective linear isotherm ( ). For an explanation of the significance of the shading used in this plot, see Appendix 1 in Nagra, 1994.