PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Modelling of Migration of Leached Radionuclides by Groundwater

J. Rae and J. H. Black

Phil. Trans. R. Soc. Lond. A 1986 **319**, 97-108
doi: 10.1098/rsta.1986.0088

Phil. Trans. R. Soc. Lond. A **319**, 97–108 (1986) [97]
Printed in Great Britain

Modelling of migration of leached radionuclides by groundwater

By J. Rae[1] and J. H. Black[2]

[1] *Theoretical Physics Division, AERE Harwell, Oxfordshire OX11 0RA, U.K.*
[2] *British Geological Survey, Nicker Hill, Keyworth, Nottinghamshire NG12 5GG, U.K.*

Radionuclides may be carried by groundwater from an underground waste repository to places accessible to man. The central role of mathematical modelling is to predict how their concentration varies in time at positions around the repository. This requires consideration of the physical and chemical processes at work and representation of the most important by mathematical expressions. As this can be done on several levels of detail both for water flow and radionuclide behaviour the resulting overall models can differ greatly in completeness and complexity. We discuss briefly some examples of such models, the data they require, the way these are gathered and how they have been used in practice. It is necessary to examine the validity of proposed models and we describe some approaches to this. We indicate the strengths and weaknesses of some commonly adopted models and outline current and future work aimed at improving our understanding.

1. Introduction

The prime reason for burying radioactive waste is to isolate it in such a way that leakage paths will be slow enough for processes of decay and dilution to render safe any contact with plants or animals. We discuss the topic from the point of view of modelling and should ask first: why do we need such models?

Materials that are buried as a safeguard will undergo various physical and chemical changes while underground. Some of them may become accessible through excavation or mining or may return to the surface in gaseous form. These and many other possibilities have to be addressed in safety assessments. One circumstance, however, is almost certain to occur except in deserts or icecaps, namely that the buried material will encounter groundwater and some of it will dissolve. This paper concerns what happens to radionuclides dissolved in groundwater, be it stagnant or moving under the influence of pressure or heat. In practice this means very long times, from hundreds of years to hundreds of thousands of years, much longer than any feasible experiment could take. There are two main reasons for using models. First we are using data from a wide variety of sources within the framework of a model to make predictions and assess the implications over long times. A second reason is that in the wider appraisal of radioactive waste disposal, migration in groundwater is only one contributing factor. Degradation and leaching of the waste packages, uptake by biological routes, geological disturbances and many other factors must be considered. If these are to be compared or coupled together in any quantitative way, some sort of model is required.

The end product of the groundwater migration modelling is commonly in the form of a spectrum of radionuclides delivered at various rates to man's environment, to the place that is most sensitive to contaminated groundwater. This has been invariably identified as a water supply borehole near to the repository. This end product forms the input for further modelling

in terms of biological uptake of radionuclides and the resulting doses to populations and individuals. Thus groundwater migration modelling forms the central portion of a three-part chain of models: that is near-field (source term), far-field (groundwater migration) and biosphere (dose assessment).

It can be seen that this whole approach predicts outcomes on the basis of extrapolating currently observed processes and flow systems. To take account of effects like glaciation and climatic change, these have to be assessed in terms of how they would change the groundwater flow rates via the hydrogeological boundary conditions. Other circumstances such as meteorite impact are not considered as part of a continuous process and are assessed outside this chain of modelling.

The following sections will discuss the processes which will affect migration of radionuclides in groundwater, the various ways in which these have been modelled in mathematical terms, the choice of model for a particular application, some examples of the use that has been made of models and, finally, the considerations that determine our confidence in applying the model. The subject has such a vast range and the publications and reports are so numerous that our treatment is eclectic, not comprehensive.

2. Processes that matter

Once dissolved in groundwater, radionuclides will undergo a number of well recognized physical processes. By 'dissolved' in this context we usually have in mind ionic species or simple inorganic complexes, but similar considerations apply to anything that moves with the groundwater. Also, we are almost always considering concentrations in trace amounts. The processes considered are:

(i) Motion with any water flow along pathlines. The flow may be driven by natural hydraulic gradients or near some heat-emitting wastes by buoyancy.

(ii) Spreading by molecular diffusion through the water, the free-water diffusion coefficient being modified by the tortuosity of pores and channels in rock.

(iii) Spreading by hydrodynamic dispersion. This comes not only from flow variation across individual rock channels, akin to Taylor dispersion in pipe flow, but from different arrival routes to the same point in the rock via several flow channels. There is no obvious reason why the latter contribution should obey Fick's Law, although this is usually assumed.

(iv) Sorption on solid phases. As part of the geochemical equilibrium, species in solution may attach themselves to rock minerals or other immobile solids by ion exchange, precipitation, chemisorption or other means.

(v) Radioactive decay. This nuclear process of course continues inexorably for the radionuclides irrespective of their local environment.

Most models for migration in groundwater attempt to take some account of these processes, although at many different levels of complexity. Of course, in projecting these processes forward in a model over long periods we must assume that chemical interaction, diffusion and radioactive decay continue to behave as we understand them now. A great body of astrophysical, geological and archaeological evidence implies that this is a reasonable assumption.

3. Range of models

The range of models available for calculating groundwater flow and solute migration is very large and it is impossible to review them and their relationships in the space available. A recent review for the European Community (Broyd *et al.* 1985) listed about 40 such models and one for the U.S. Nuclear Regulatory Commission (Curtis *et al.* 1983) about 160 more, with very little overlap. Instead, we shall introduce the most commonly used simple model and, having discussed its characteristics, relate the wider discussion to it.

The concentrations of radionuclides as they change in space and time can often be treated by a deterministic model in one space dimension, which consists of a set of partial differential equations of the form:

$$R_i \frac{\partial C_i}{\partial t} + V \frac{\partial C_i}{\partial x} - \frac{\partial}{\partial x} D_i \frac{\partial C_i}{\partial x} = \lambda_{i-1} R_{i-1} C_{i-1} - \lambda_i R_i C_i. \tag{1}$$

This set of equations, in time and one space dimension as independent variables, governs the concentration C_i of radionuclide species i. There is one such equation for each species in a decay chain of species $1, 2, \ldots, i-1, i, \ldots, N$.

The process represented by the terms in (1) are exactly those described in the previous section. In turn from left to right they are:

(i) The change in concentration at a particular place. This term is scaled by a retardation factor R_i, which is greater than 1, sometimes much greater, if species i is sorbed onto surrounding immobile solid. R_i has mostly been taken as a species-dependent constant, corresponding to a linear equilibrium sorption model (Jensen 1982) in which sorbed concentration is always proportional to dissolved concentration.

(ii) Advection with the water flow velocity V, which is often taken as a constant.

(iii) Dispersion and diffusion of species i. Although hydrodynamic dispersion does depend on water velocity (Bear 1979), when V is assumed constant this in effect gives D_i a constant value.

(iv) Creation of species i by decay of its parent nucleus $i-1$ at a rate λ_{i-1}. Radioactive decay operates whether the species is sorbed or not, so this term picks up a factor R_i.

(v) Destruction of species i by decay into its daughter $i+1$ at rate λ_i.

On this simplest level the equations (1) are weakly coupled linear parabolic differential equations (Friedman 1964) and have to be supplemented by appropriate initial and boundary conditions to complete the model. In the linear case it is often possible to write down solutions more or less explicitly (Burkholder & Rosinger 1980; Harada *et al.* 1980; Pigford *et al.* 1980; Herbert 1984) but these are often too cumbersome to use effectively. Numerical methods are often employed, and always in the case of more complex models. Many approaches are available and have been used, for example finite differences, finite elements or numerical integral transforms, but things are not always straightforward and difficulties are encountered. We mention here only the principal two. First, because the retardation factors R_i and decay rates λ_i can differ by many orders of magnitude within a single chain there can be many timescales implicit in (1). If the solutions are to be computed stably and efficiently timestepping methods appropriate to stiff equations have to be used (Byrne & Hindmarsh 1975). Second, in (1) the interplay between the second term, convection, and third term, dispersion, is measured by the dimensionless Péclet number VL/D, where L is an appropriate length. When the partial

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

differential equations are made discrete in space by finite difference or element methods, the local mesh or element size is the appropriate L and one obtains a mesh Péclet number P. For many numerical schemes, including centred differences and the simplest element methods, the solutions can be spatially unstable and exhibit oscillation if P is greater than a critical value. There are remedies to this, for example by refining the mesh to reduce P, by adopting some form of upstreaming (Hughes (ed.) 1979; Roache 1972), or by some Lagrangian approach (Ames 1969). These all have drawbacks in terms of cost, accuracy or complexity so care is needed in the choice of methods.

The model given in (1) is relatively simple and many alternatives and extra complexities have been considered. Within the class of deterministic models some examples of natural extensions to it are:

(i) Flow and dispersion in 2- or 3-dimensional regions. This has important implications for the hydrogeological data needed and for the costs of computation.

(ii) Non-constant velocities obtained from groundwater flow calculations. Most commonly these are 2- or 3-dimensional permeable flow calculations based on much of the available geological and hydrogeological evidence on strata, faults and hydraulic head distribution. Less commonly, the water flow may be calculated for fractured rock where the scale of the fractures is not small enough for them to act as a permeable medium. One approach to this (Robinson 1984*a*, *b*; Long & Witherspoon 1985; Schwartz *et al.* 1982) is to construct computer realizations of fracture networks with parameters such as length, orientation and aperture sampled from probability distributions that have been fitted to experimental data on fracture statistics.

(iii) A better representation of the chemistry of sorption. The linear equilibrium model adopted above is known to be inadequate in many cases. Not only is sorption often nonlinear but kinetic effects can also occur, especially in the shorter laboratory or field experiments. With many dissolved species present simultaneously with many rock minerals and degradation products this geochemistry becomes a formidable problem in its own right.

(iv) More elaborate dispersion models. In layered rock strata, account may have to be taken of the tensorial character of dispersion and its dependence on flow velocity. In general the relation between longitudinal and transverse dispersion is poorly understood and this is especially so for fractured rock (Robinson 1984*b*; Matheron & de Marsily 1980).

(v) Modelling effects of heat. Some radioactive wastes emit appreciable amounts of heat, which may affect the geochemistry, particularly very near to the repository, the rock mechanics and, through buoyancy effects, the groundwater flow (Hodgkinson *et al.* 1983).

(iv) Diffusion of solute from flowing groundwater into pores or fissures filled with stagnant water. This has a retardation effect similar to that of sorption but does not depend on the local chemistry (Lever *et al.* 1983; Grisak & Pickens 1980; Neretnieks 1980).

As well as these reasonably obvious extensions to deterministic models there are other effects being studied, such as the role of colloids or organic complexing agents or, indeed, of microbes in the groundwater, which have not yet been included in models (Chapman & Sargent (eds) 1984).

Sometimes deterministic models are simplified back to (1) or even simpler forms. In cases where transfer processes are poorly understood but overall rates are available, models of the compartment type have been used (see, for example, Helton & Kaestner 1981; Smith *et al.* 1982). In other cases the migration models are only a small part of an overall coherent system of assessment, the SYVAC program is an example (Wuschke *et al.* 1981; Thompson 1984), in

which the individual parts must be simple to be manageable. In some models that are run for many choices of parameters, the methods of statistical sampling and processing become an integral and even dominant part of the method (Harper 1983; Kocher *et al.* 1982).

Finally, in some studies where unforeseen natural events or human actions are important, probabilistic models are the most appropriate (D'Alessandro & Bonne 1981).

4. Choice of model

The range of possible models of migration within groundwater is wide. The choice of a particular combination of processes and geometry is based on:

(i) the proposed use of the model (level of accuracy required, number of times the model is to be used, etc.);

(ii) the physical understanding of the expected processes;

(iii) available data and means of acquiring it;

(iv) the ability to verify and validate the model.

We amplify the first three points here; point (iv) is discussed in §6.

Whatever the choice of model it will of necessity be an approximation to reality. The selection of the geometrical representation of the likely flow system is a crucial part and illustrates the approximation procedure. If the model is two-dimensional rather than three-dimensional then the cross section has to be representative of the region perpendicular to the plane of the model. This is easily conceivable in models of regular sedimentary sequences such as at Harwell (Brightman & Noy 1984) or at Mol in Belgium (Bonne *et al.* 1985). Figure 1 shows cross sections of both sites with the planes of the model vertical and parallel to the dip at the formations. In contrast to water resource models where the plane of a 2-D model is invariably horizontal, in migration modelling the plane is always vertical. This results from the overriding importance of vertical flow in the safety assessment of a potential repository site. Both models in figure 1 illustrate the translation of actual rocks into modelled formations. In the Harwell example the central clay formation is in reality two clay formations separated by a thin sand formation (the Lower Greensand). For the purpose of the model this complexity was simplified. Similarly, in Belgium the overlying Neogene aquifer consists of at least four geological units (all sands) considered inseparable for the purposes of modelling. The 'massaging' of the real geometry to be accommodated into a model is perhaps at its most extreme in 2-D analytical models such as the 'wedge and step' approach (figure 2; see later) which has been applied as a reconnaissance tool to the Worcester Basin (Black & Barker 1981). This arises from the need to simplify to the point of having only straight boundaries. As computers become even larger the models are able to grow in geometrical complexity and almost all the geological details (where known) can be included. Most of these considerations apply only to repositories in sedimentary formations. In crystalline rocks flow is controlled by the orientation of major shear zones, faults, crush zones and the like, and 2-D models are inadequate. This is due to the impossibility of choosing one representative cross section because there is no basic symmetry in the system. An example of this, in figure 3, is discussed later. Translating geological systems into a form suitable for modelling can be seen to be very much a matter of judgement.

Like the basic geometry, a relevant set of boundaries is a question of interpretation. Since the models aim to account for the circulation of groundwater within the modelling region it is apparent that all boundaries must be defined either in terms of flow or of

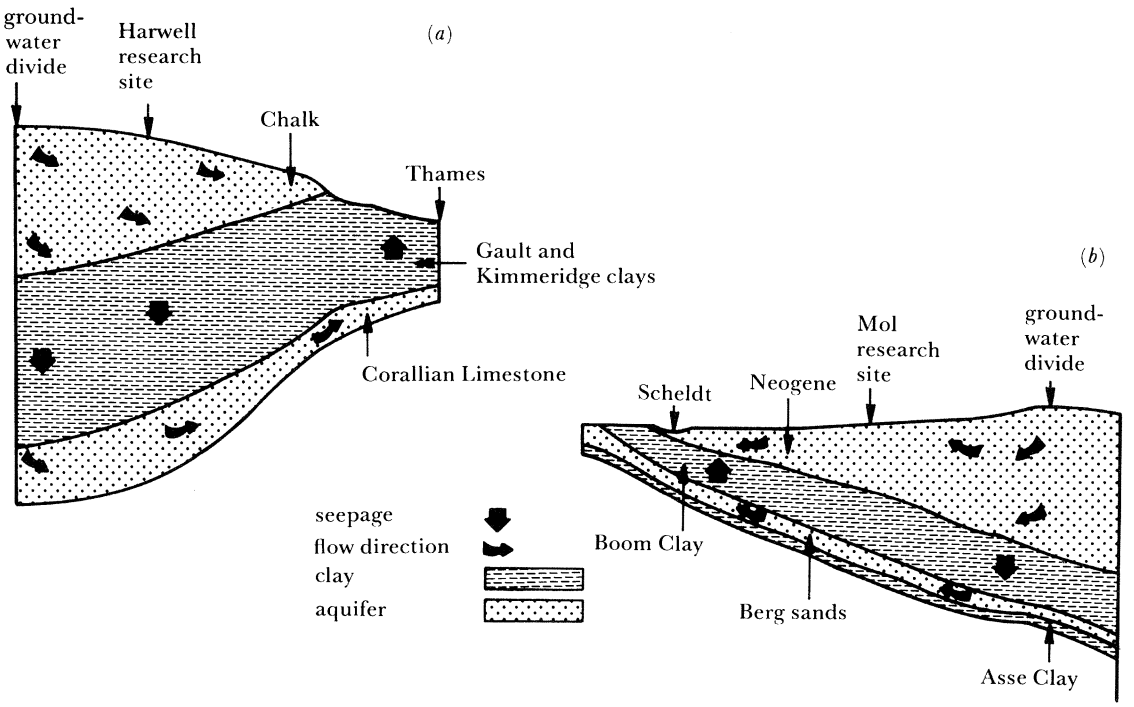

FIGURE 1. (*a*) Model configuration of the Harwell site; (*b*) model configuration of the Mol site.

head. In most models (including those in figure 1) the bottom boundary is an impermeable base where a zero-flow boundary is assumed. This may not always be the last time the sedimentary pile can be shown to have accommodated measurable fluxes from consolidating rocks over geological time. Lateral boundaries have different problems and usually consist of vertical zero-flow boundaries beneath either groundwater divides or rivers (towards which groundwater flow converges). A problem with this approach is that the identified divide may actually be a surface-water divide and it may not coincide with the groundwater divide. Additionally in multilayer sequences the position of the groundwater divides in individual layers may be different. Also where very large systems are being modelled it is sometimes difficult to identify a major flow system boundary because the system may extend to some considerable depth. Even the existence of the coast within a modelled region may not be a satisfactory choice since experience indicates offshore undersea flow in an unsettling number of cases. The top surface of the model is usually assumed to be a fixed head boundary although most models take this to be the water table. Recharge or infiltration rates are then adjusted along this surface to keep it in position. No account is taken of the presence of the unsaturated zone or of the manner in which water tables oscillate on a seasonal basis. It also results in an unrealistic apportioning at the top surface into areas of recharge and discharge. In crystalline rock models the same reservations apply concerning boundary conditions but if anything the top surface is more complex and less predictable. This is because the smoothness of a water table is related to the homogeneity of the rocks in which it is situated. A common assumption in all models where rocks with low hydraulic conductivity intersect the surface is that the water table is coincident with the land surface. This assumption is commonly invalid.

Data gathering and choice of model go closely together, for all variables in the model need

to be supplied somehow and experimental evidence will suggest whether some processes, for example kinetic sorption or rock matrix diffusion, need to be included in the model. Laboratory experiments are usually done on small samples, 1 cm to 1 m, of rock core taken from representative rocks. Permeability and diffusion measurements are made in a variety of experimental cells (see, for example, Bradbury 1983; Hemingway *et al.* 1983) and sorption of specific chemical species can be assessed in a number of ways: batch sorption on crushed or monolithic specimens, sorption in diffusion cells or sorption in flow-through experiments (Nuclear Energy Agency 1983). There are some difficulties of principle in such laboratory experiments. The samples may be too small to be representative, they will have been considerably disturbed during the coring operation, and the short timescales of the experiments, perhaps months, may emphasize transient effects that would not matter in disposal conditions. There are also practical difficulties of holding the samples in an appropriate environment, avoiding contamination particularly of surfaces and using low enough concentrations of tracers to approach the extremely low levels expected in migration from a repository. Field experiments suffer from a similar range of problems. They generally involve drilling a pattern of boreholes and conducting a range of pumping and tracer tests that can provide information on flow, dispersion and sorption (Nuclear Energy Agency 1979*a*, *b*). Measurements are required of the hydraulic conductivity of the rock (if it is porous) or the transmissivities of the fissures if it is fissured. In practice the same equipment would be used for both measurements. Porosity is the other basic hydrogeological property but again it needs to be relevant to the type of flow. Lastly hydraulic head requires measurement so that the model can be calibrated. In this aspect many models are ambiguous because all heads are in terms of fresh water, yet the regions modelled often contain saline water of varying density. Many models can accommodate the effects of heating inducing density differences, though few cope well with salinity-related differences. This is a particular problem in models associated with salt domes.

In tracer tests the scales of distance and time are greater than those in the laboratory experiments but still well short of realistic, and, of course, the circumstances of the experiment are now not fully under control.

5. Use of models

The models described above have been used in several ways: in the design and analysis of experiments (Hodgkinson & Lever 1983; Hodgkinson 1984), in 'generic' safety studies with made-up scenarios (Hill & Grimwood 1978; Burkholder 1980; Wusche *et al.* 1981; Hill *et al.* 1981), in sensitivity studies (Hill 1979; Hodgkinson *et al.* 1984), and in studies of real sites that are not proposed as repository sites, as in the European Community MIRAGE programme (CEC 1984, 1985). Published applications to proposed real repository sites are rare though there are examples such as the Environmental Impact Statement for the Waste Isolation Pilot Plant in New Mexico (U.S. Department of Energy 1980), the Swedish KBS-3 study on disposal of spent fuel (Swedish Nuclear Fuel Supply Company 1983) and the recently published reports on 'Project Gewähr', which examined nuclear waste disposal in Switzerland (NAGRA 1985). Rather than attempting to review these we give just two examples.

The first example is a two-dimensional mathematical analysis known as the 'wedge and step' model. It has been used in reconnaissance studies in the U.K. and has simple geometry that can vary between two extreme configurations (figure 2). Essentially it concerns a layer of low

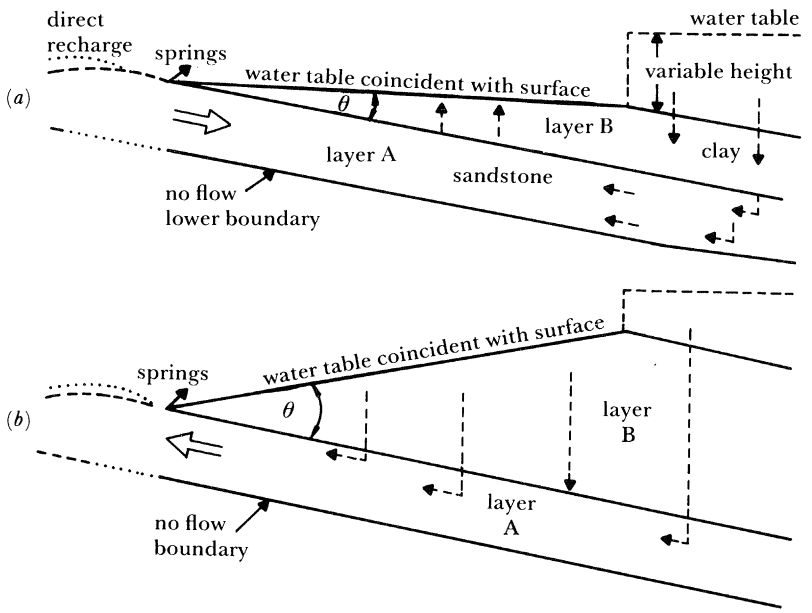

FIGURE 2. Wedge and step model configuration.

hydraulic conductivity (K) between two higher K layers. The underlying low K layer is regarded as having negligible flows and takes no part in the flow system. Layers A and B have parallel boundaries and dip uniformly. Although the boundary conditions are fixed the geometry and the properties are variable. The thickness of layers A and B and the length of the systems are totally variable whereas the angle θ varies between commonsense limits. As regards the boundary conditions, it is first assumed that layer B has a low enough hydraulic conductivity for the average U.K. rainfall to more than saturate it (i.e. $K < 2 \times 10^{-8}$ m s^{-1}). This results in the water table's coinciding with the top surface of layer B. Next it is assumed that rainfall recharging the outcrop part of layer A forms a water table within A and that excess head discharges in the form of springs at the feather edge of the clay. This has the effect of reducing the head at that point in the aquifer to the altitude of the feather edge. The head in the overlying aquifer is completely variable but it is assumed that it applies directly on the top surface of the clay (i.e. there is negligible head loss during vertical flow through the overlying aquifer). Although it is recognized that, in reality, the water table would steepen gradually in association with the scarp-slope and fall away in equilibrium with the dip slope, the simplified model requires that it should rise suddenly and remain at a constant height. This was adopted as the closest approximation to reality with the minimum of assumption. The model does not attempt to predict accurately the groundwater conditions in the immediate vicinity of the base of the scarp.

This model obviously contains a great number of approximations and simplifications but it serves as a powerful tool in reconnaissance. As an analytical model it is easy to see the relations between geometry and the bulk properties of the identified layers. It can be used to predict the likely direction of flow in any of the layers. In the Worcester Basin case the model predicted a flow balance point within layer A. Its position depended on the ratio of the bulk hydraulic conductivities of layers A and B. This flow balance is an important point of this particular system because it strongly influences the interpretation of groundwater ages derived from groundwater

sampling. From a reconnaissance viewpoint perhaps the most important aspect of the use of this model is in determining the likely magnitude of head variations and therefore the measurement sensitivity required in a subsequent site investigation.

For the second example we use the KBS-3 study. Three principal sites in northern Sweden, at Fjälveden, Gidea and Kamlunge, were examined in great detail with many borehole experiments in the granitic rock. The results were processed to produce effective flow parameters such as permeabilities and porosities, but because an assumption of Darcy flow was made here, and the rocks were fractured on a scale of metres, there remains some doubt as to the adequacy of this. The groundwater flows were modelled by using a three-dimensional finite-element model of Darcy flow in a permeable medium, with permeability depending on depth, and some large faults explicitly added. Both regional and local models were used, the former supplying boundary conditions for the latter. An example of such a model is given in figure 3. The results provided typical flows through the repository regions and the most pessimistic of these, that is the highest flows, were used to find essentially one-dimensional paths for radionuclide transport. The KBS-3 design made considerable use of barriers in or near the repository, in particular by having the spent fuel in very thick copper canisters and surrounding them with a layer of absorbent bentonite clay. It was assumptions relating to these barriers and repository chemistry that chiefly influenced the choice of five scenarios to be studied, the flow and migration paths being the same in all cases. The radionuclide transport through the geosphere was calculated along the one-dimensional paths with the species convected, dispersed, sorbed, decaying and diffusing into the rock. Thus the model was a slightly more elaborate version of that given by (1). The arrival rates into the biosphere, in becquerels per time for each radionuclide, became input to further programs calculating doses to man.

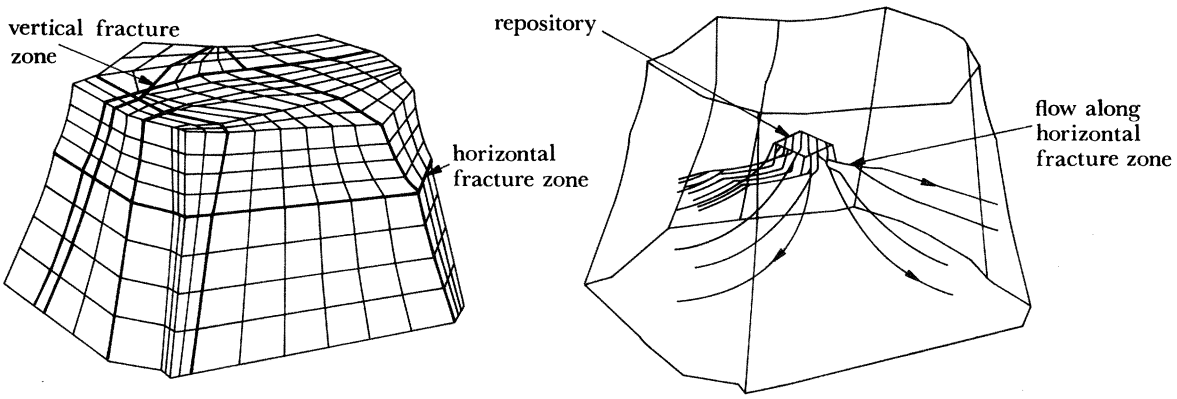

FIGURE 3. Finite-element grid and perspective plot of pathlines for three-dimensional groundwater flow.

6. JUSTIFICATION OF MODELS

It is clearly very important to know to what extent mathematical models are justified, and that we can rely on their predictions. There is no easy or complete way to do this. It is rather a question of applying a whole range of checks and comparisons until enough evidence is accumulated. Some simplification can be found for radioactive waste disposal by erring on the 'pessimistic' side, which is acceptable for a safety case, but care has to be taken that an assumption that is pessimistic for one process alone remains so in a larger context. It is

convenient to distinguish two aspects of justification of models: verification, where we accept the assumptions of the model and check that its calculations and consequences follow correctly; and validation, where we examine whether the model adequately represents the real situation. We touch briefly on both aspects.

In verification of models there are three complementary approaches. First, we can make direct comparisons between independent models that have equivalent capabilities as they perform on a range of problems. This is sometimes done on a one-to-one basis; the Harwell program NAMMU was used to repeat the groundwater flow calculations of KBS-3 (Atkinson *et al.* 1984), and sometimes in a larger, more organized, way as in the international comparison exercises INTRACOIN for migration and HYDROCOIN for groundwater flow (Swedish Nuclear Power Inspectorate 1984; Ross *et al.* 1982). Second, we can compare model performance in certain cases with exact analytic solutions, of which a growing number are being published (Harada *et al.* 1980; Pigford *et al.* 1980; Herbert 1984). And third, we can use a range of software tools to check computational aspects, that data flow through programs is correct, that all paths have been tested, and so on. In these ways we can build confidence that the model is doing what it ought to, although aware that probably no large computer program is ever totally free of bugs.

In validation of models we must compare them with real physical systems and it is useful to distinguish between laboratory experiments, field experiments and use of analogues. Laboratory experiments usually focus on one or two combined processes such as sorption and diffusion (Bradbury *et al.* 1982) but there have been attempts at 'integral' experiments, which attempt to include many of the processes simultaneously (Saltelli *et al.* 1984; Bidoglio *et al.* 1984).

As examples of field experiments on migration we may take the three investigations which are part of the European MIRAGE project (CEC 1984): the Commissariat à l'Énergie Atomique is carrying out a programme of permeability and dispersion measurements in a uranium mine at Fanay-Augères near Limoges: single-fracture tracer experiments are being done at Troon in Cornwall by the U.K. Atomic Energy Authority, and the British Geological Survey operates a tracer test in glacial deposits at Drigg in Cumbria. Other examples are migration in single fracture experiments at Chalk River and at Finnsjön, both of which have recently been used in validation exercises (Lever 1984; Hodgkinson & Lever 1983).

In recent years there has been an increase in interest in the use of natural analogues, that is natural geological migration systems, as a way of extending validation over very long timescales. The method is difficult to use because of the lack of experimental control but well worth investigation. We mention only two examples here. First is the uranium series disequilibrium method (Ivanovich & Harmon 1982) whereby the ratio of the uranium isotopes $^{234}U/^{238}U$, and sometimes thorium or radium isotope ratios, is related to groundwater movement. The method is currently being employed at the Mol site in Belgium and at various uranium mines in Australia. The second analogue is alternating freshwater and marine sediments laid down in Loch Lomond during the last 8000 years. Detailed geochemical profiles of cores taken from these sediments show the effects of diffusion and sorption on various elements over thousands of years (MacKenzie *et al.* 1984).

The basic problems with checking the applicability of current models still lies in collecting enough data of different types so that the model can be constructed and predictions checked against an independent measurement. The natural evolution of the composition of ordinary

groundwater within a complete flow system would be a reasonable start. So far no such model has been constructed and all groundwater chemistry data seem to be used in a generally qualitative manner (Nordstrom *et al.* 1985).

7. Conclusions

Although models of the migration of radionuclides in groundwater follow a set of implicit rules they show a great variety. They vary from the straightforward one-dimensional calculation to the complicated three-dimensional finite-element models. It is clear that the choice of model for any given site needs to take into account the purpose of the modelling and the amount and type of data available. Often for reconnaissance purposes a comparatively simple analytical model best suits the density of data available. For detailed assessment the three-dimensional fully coupled model is the most appropriate. Currently the value of such models is reduced by the lack of site specific data to use in the model. Linked with this is the lack of sufficient independent data against which to cross-check the model predictions. At the moment models outstrip the data and it is probably true that there are more models than sites modelled.

Modelling of radionuclide migration is at present a very active area of research. Topics receiving particular attention are flow in fractured rock, the comprehensive coupling of flow and geochemistry, and the associated work on natural analogues. This last illustrates current concern in establishing the validity of models over geological timescales and confirming that all important processes have been identified.

References

Ames, W. F. 1969 *Numerical methods for partial differential equations*. London: Thomas Nelson.

Atkinson, R., Cherrill, T. P., Herbert, A. W., Hodgkinson, D. P., Jackson, C. P., Rae, J. & Robinson, P. C. 1984 *AERE Harwell Report* no. R.11140.

Bear, J. 1979 *Hydraulics of Groundwater*. New York: McGraw-Hill.

Bidoglio, G., de Plano, A., Avogadro, A. & Murray, C. N. 1984 *Inorg. chim. Acta* **95**, 1–3.

Black, J. H. & Barker, J. A. 1981 *Rep. Inst. geol. Sci.* no. ENPU 81-3.

Bonne, A., Black, J. H., Gera, F., Gonze, P., Tassoni, E. & Thimus, J. F. 1985 In *Proc. Conf. on Management and Disposal of Radioactive Waste, Luxembourg 22–26 April 1985*. (In the press.)

Bradbury, M. H. 1983 *AERE Harwell Report* no. R.10836.

Bradbury, M. H., Lever, D. A. & Kinsey, D. V. 1982 In *Scientific basis for waste management*, vol. 5 (ed. W. Lutze), pp. 569–578. Amsterdam: Elsevier.

Brightman, M. A. & Noy, D. J. 1984 *Rep. Br. geol. Surv.* no. FPU 84-1. (In the press.)

Broyd, T. W., Dean, R. B., Hobbs, G. D., Knowles, N. C., Putney, J. M. & Wrigley, J. 1985 *Commission of the European Communities Report* no. EUR 8669 En. Luxembourg: CEC.

Burkholder, H. C. 1980 In *Scientific basis for nuclear waste management*, vol. 2 (ed. C. J. M. Northrup), pp. 689–702. New York: Plenum Press.

Burkholder, H. C. & Rosinger, E. L. J. 1980 *Nucl. Technol.* **49**, 150–158.

Byrne, G. D. & Hindmarsh, A. C. 1975 *ACM Trans. math. Softw.* **1**, 71–96.

Chapman, N. A. & Sargent, F. P. (eds) 1984 *Workshop on the Geochemistry of High-Level Waste Disposal in Granitic Rocks, Atomic Energy of Canada*. Commission of the European Communities Report no. AECL 8361, EUR 9162. Luxembourg: CEC.

Commission of the European Communities 1984 *The CEC Project MIRAGE*. Report no. EUR 9403. Luxembourg: CEC.

Commission of the European Communities 1985 *First Summary Progress Report on MIRAGE*. Report no. EUR 9543. Luxembourg: CEC.

Curtis, R. H., Wart, R. J. & Skiba, E. L. 1983 *U.S. Nuclear Regulatory Commission, Report* no. NUREG/CR-3450.

D'Alessandro, M. & Bonne, A. 1981 *Radioactive waste disposal into plastic clay formations*. Brussels: Harwood Publishers for CEC.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Friedman, A. 1964 *Partial differential equations of parabolic type*. Englewood Cliffs, New Jersey: Prentice-Hall.
Grisak, G. E. & Pickens, J. F. 1980 *Wat. Resourc. Res.* **16**, 719–730.
Harada, M., Chambré, P. L., Foglia, M., Higashi, K., Iwamoto, F., Leang, D., Pigford, T. H. & Ting, D. 1980 *Lawrence Berkeley Laboratory Report* no. LBL-10500.
Harper, W. V. 1983 *U.S. Department of Energy Office of Nuclear Waste Isolation Report* no. ONWI-444.
Helton, J. C. & Kaestner, P. C. 1981 *U.S. Nuclear Regulatory Commission Report* no. NUREG/CR-1636.
Hemingway, S. J., Bradbury, M. H. & Lever, D. A. 1983 *AERE Harwell Report* no. R.10691.
Herbert, A. W. 1984 *AERE Harwell Report* no. TP.1058.
Hill, M. D. 1979 *National Radiological Protection Board Report* no. NRPB-R86.
Hill, M. D. & Grimwood, P. D. 1978 *National Radiological Protection Board Report* no. NRPB-R.69.
Hill, M. D., Mobbs, S. F. & White, F. 1981 *National Radiological Protection Board Report* no. NRPB-R126.
Hodgkinson, D. P. 1984 *AERE Harwell Report* no. R.11287.
Hodgkinson, D. P. & Lever, D. A. 1983 *Radioact. Waste Mgmt nucl. Fuel Cycle* **4**, 129–158.
Hodgkinson, D. P., Lever, D. A. & England, T. H. 1984 *Ann. nucl. Energy* **11**, 111–122.
Hodgkinson, D. P., Lever, D. A. & Rae, J. 1983 *Prog. nucl. Energy* **11**, 183–218.
Hughes, T. J. R. (ed.) 1979 *Finite element methods for convection dominated flows* (AMD vol. 34). New York: ASME.
Ivanovich, M. & Harman, R. S. 1982 *Uranium series disequilibrium: application to Earth sciences*. Oxford University Press.
Jensen, B. S. 1982 *Migration phenomena of radionuclides into the geosphere*. Brussels: Harwood Publishers for CEC.
Kocher, D. C., Sjoreen, A. L., Bard, C. S. & Olsen, C. R. 1982 In *American Nuclear Society Symposium Waste Management '81* (ed. R. G. Post). Tucson: University of Arizona.
Lever, D. A. 1984 *AERE Harwell Report* no. R.11299.
Lever, D. A., Bradbury, M. H. & Hemingway, S. J. 1983 *Prog. nucl. Energy* **12**, 85–117.
Long, J. C. S. & Witherspoon, P. A. 1985 *J. geophys. Res.* **90** (B4), 3087–3098.
MacKenzie, A. B., Scott, R. D., Ridgway, I. M., McKinley, I. G. & West, J. M. 1984 *Rep. Br. geol. Surv.* no. FLPU 84-11. (In the press.)
Matherton, G. & de Marsily, G. 1980 *Wat. Resourc. Res.* **16**, 901–917.
NAGRA 1985 *Gewähr Projektbericht* nos NGB 85–01 to 85-08. Switzerland: National Genossenschaft für die Lagerung radioactiver Abfälle.
Neretnieks, I. 1980 *J. geophys. Res.* **85**, 4379–4397.
Nordstrom, K., Andrews, J., Fontes, J. C., Fritz, P., Michelot, J. L., Moser, H., Olsson, T. & Wolf, M. 1985 *Phase I Internal Report of the Stripa Project, Stockholm.* (In the press.)
Nuclear Energy Agency 1979*a* *Low-flow, low-permeability measurements in largely impermeable rocks*. Paris: OECD.
Nuclear Energy Agency 1979*b* *Use of argillaceous materials for the isolation of radioactive waste*. Paris: OECD.
Nuclear Energy Agency 1983 *Sorption: modelling and measurement of nuclear waste disposal studies*. Paris: OECD.
Pigford, T. H., Chambré, P. L., Albert, M., Foglia, M., Harada, M., Iwamoto, F., Kanki, T. L., Leung, D., Matsuda, S., Muraoka, S. & Ting, D. 1980 *Lawrence Berkeley Laboratory Report* no. LBL-11616.
Roache, P. J. 1972 *Computational fluid dynamics*. Albuquerque: Hermosa Publishers.
Robinson, P. C. 1984*a* *J. Phys.* A **17**, 2823–30.
Robinson, P. C. 1984*b* *AERE Harwell Report* no. T.P.1072 (D.Phil. thesis, St. Catherine's College, Oxford.)
Ross, B., Mercer, J. W., Thomas, S. D. & Lester, B. H. 1982 Report no. NUREG/CR 3097. Washington, D.C.: U.S. Nuclear Regulatory Commission.
Saltelli, A., Avogadro, A. & Bidoglio, G. 1984 *Nucl. Technol.* **67**, 245–254.
Schwartz, F. W., Smith, L. & Crowe, A. S. 1982 In *Scientific basis for nuclear waste management*, vol. 4, pp. 457–463. New York: North-Holland.
Smith, C. B., Egan, D. J., Williams, W. A., Gruhlke, J. M., Hung, G. Y. & Serini, B. L. 1982 *U.S. Environmental Protection Agency Report* no. EPA 520/3-80-006.
Swedish Nuclear Fuel Supply Company 1983 *Report: final storage of spent nuclear fuel KBS-3.*
Swedish Nuclear Power Inspectorate 1984 *Intracoin Final Report level 1*. Stockholm S10252, Box 27106.
Thompson, B. G. J. 1984 In *British Nuclear Energy Society Symposium on Radioactive Waste Management, London* (*Nucl. Energy*). (In the press.)
U.S. Department of Energy 1980 *Final Environmental Impact Statement for Waste Isolation Pilot Plant Report* no. DOE/EIS 0026.
Wuschke, D. M., Mehta, K. K., Dormuth, K. W., Andres, T., Sherman, G. R., Rosinger, E. L. J., Goodwin, B. W., Reid, J. A. K. & Lyon, R. B. 1981 *Atomic Energy of Canada Report* no. AECL-TR-127. (3 volumes.)

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES