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12 The role of academic environmental geoscientists in radioactive waste disposal assessment

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Summary

- Academic environmental geoscientists can provide an independent scientific view on the issues involved in the disposal of nuclear waste, as illustrated by the proposed site at Sellafield, Cumbria.
- UK NIREX Ltd had identified Sellafield as a possible site for the disposal of low and intermediate level nuclear waste, but following an extensive Public Inquiry, it was judged to be unsuitable.
- Here we discuss some of the contentious issues which played a role in the Inquiry, and use computer simulations to better understand the geosphere processes involving hydrogeology and geochemical interactions.
- An open forum needs to be established within which the academic community, together with commercial geoscientists, can reach an understanding on the best approaches, and on the sites which appear best suited, to solve the pressing environmental problem of radioactive waste disposal.

During the last half-century of the 'nuclear age', significant quantities of radioactive waste materials have been generated (in all the major nuclear countries); nearly all of these wastes are presently in storage awaiting a decision on how best to achieve their permanent disposal. The volumes of this waste are not large in comparison with domestic or industrial waste: 257 000 m^3 of Intermediate Level Waste (ILW), forming 26% of the total radioactive waste volume, will have been produced in the UK by AD 2030 (NIREX 1992*a*). As an aid to visualizing the quantity of waste, this volume is similar to that occupied by the 1×10^6 tonnes

of metal in a small, commercial mine although a mine will have a substantial amount of gangue materials which will contribute to the void space created during its extraction.

Deep underground burial is presently considered to offer the greatest chance for long-term isolation of the waste (Bredehoeft & Maini 1981; Chapman & McKinley 1987; Billington *et al.* 1989; Nuclear Energy Agency 1989; Chapman 1994; Royal Society 1994) and is currently favoured in several countries (Hooper 1995; Karlsson 1995; Langmuir 1995; Horseman 1996). Assessment of the failure potential of such a disposal scheme is usually divided into two parts: the engineered system (waste form, containers and their surrounding packing); and that of the geosphere, which includes the hydrogeological system and the host rock. The disposal industry refers to containment in terms of an engineered barrier, which is surrounded by a geosphere which results in dilution and dispersion of escaped wastes. Although the failure probabilities of the engineered systems are relevant, most safety debates focus on the geosphere. In this chapter, we consider the role of the geosphere in retarding the return of radionuclides to the surface of the Earth. In doing so we emphasize the behaviour of the hydrogeological system, including the interaction of groundwaters with their natural and engineered settings.

Our purpose is to describe the ways in which environmental geoscientists can be involved in evaluating the expected performance of a nuclear waste disposal scheme – that is, the performance of the geosphere in slowing or preventing the migration of escaped wastes. Geoscientists are uniquely qualified to participate in such an assessment because of our breadth of knowledge concerning a wide range of interacting earth processes, and because of our appreciation of the complexity of earth processes across a spectrum of scales. Central to the involvement of environmental geoscientists is a recognition of the ways that mass can be transported through a body of rocks. Such concerns arise in all areas of geoscience (Wood 1997). Groundwaters are a readily-identified potential agent of transport, but gas-phase or additional liquid-phase mechanisms cannot be ignored. Within the groundwater domain, there is also a need to consider if the materials being transported (e.g. radionuclides) are retarded by physical and

chemical interactions with the rock mass, or by changes in the chemical and/or electro-chemical state of the waters as they migrate to other crustal locations. Each of these concerns is relevant to some of the radioactive species which might escape from a disposal site. Many of the topics addressed in this chapter are applicable to other forms of waste which could be disposed of in the geosphere. We use the issue of nuclear waste disposal to illustrate general points that we seek to make concerning the involvement of academic environmental geoscientists in the broader topic of geosphere pollution assessment, prevention and remediation.

Context of geoscience involvement

We consider it useful to contrast two situations which arise in the effort to develop a safe disposal scheme for nuclear waste materials: (1) the exploration for potential sites; and (2) the evaluation of proposed sites. In the exploration phase, it is possible that academic and commercial geoscientists act similarly, but once a potential site has been identified and the safety of the site is being evaluated, the two groups certainly have differing agendas, as we explain below.

The content of this chapter refers specifically to the disposal site for radioactive waste that was proposed to be constructed at Sellafield, in Cumbria, UK, since the scientific and political issues raised prove particularly instructive. Other views on the geology and hydrogeology of the Sellafield site are summarized by Bath *et al.* (1996), Black & Brightman (1996), Heathcote *et al.* (1996) and Michie (1996). The disposal site at Sellafield was planned to be located in the Borrowdale Volcanic Group (BVG) at 650 m depth, at a location between the Cumbrian mountains and the coastline. These rocks are overlain (from 450 m up to the present land surface) by Carboniferous–Triassic marine deposits, terrestrial sandstones, and mudrocks, along with a veneer of Recent deposits.

It is important to point out that on 17 March 1997, the UK Secretary of State for the Environment upheld the decision by Cumbria County Council to reject a planning application to construct an underground Rock Characterization Facility (RCF) at the Sellafield site. His decision was based on the recommendations of the Planning Inquiry Inspector, who stated that: 'The indications

are, in my judgement, still overwhelmingly that this site is not suitable for the proposed repository, and that investigations should now be moved to one of the more promising sites elsewhere' (McDonald *et al.* 1996). Since NIREX have stated that they will not challenge this decision the Sellafield repository site has, in effect, been abandoned.

Exploration

In common with disposal organizations overseas (Karlsson 1995; Horseman 1996), it is proposed that the entire UK inventory of ILW should be disposed of in a deep, geological repository (NIREX 1993*b*). Such repositories are subject to regulatory guidelines (Holmes 1995; Hooper 1995) such that for the period after closure of the disposal facility the radiological risk to the human population will be 1×10^{-6} (i.e. 1 death in 1 million per year). Given that ILW contains a significant inventory of very long-lived radioactive constituents (NIREX 1992*c*; Royal Society 1994), it is UK policy that the disposal concept provides adequate containment for greater than 10^8 years.

A sub-surface disposal site needs to meet certain basic characteristics. In terms of geosphere criteria, these have historically been paraphrased as:

(1) stable and workable rock types, to permit the safe and easy excavation of an open cavern;
(2) suitable hydrogeology;
(3) suitable geochemical environment.

Additionally, for the long timescales of radioactive waste duration, consideration must be given to:

(4) past and future tectonic activity;
(5) possible future intervention by humans seeking to retrieve the material, or to exploit a resource;
(6) climate change;
(7) meeting the present and future regulations on permitted dose rates (Chapman & McKinley 1987).

There has been an emphasis around the world to identify potential sites in 'homogeneous' crystalline (hard) rock masses, although other rock types (such as rock salt or mudrocks), and other situations (such as former mine workings) are also considered. Crucial to meeting the first characteristic is a lack of, or minimal number of, discontinuities (joints, faults) – since these have a major impact on rockmass behaviour. The matter of 'suitable hydrogeology' is less clear. The fact that groundwater is at all important results from the possibility that contaminants could be transported by the movement of groundwaters (in the worst case, to the Earth's surface, or perhaps into the shallow sub-surface). The pattern and rate of undergroundwater flow must, therefore, be both predictable and safe for geologically long times into the future.

The diverse geological factors affecting such predictions, and some of their difficulties of measurement and forecasting, have been reviewed elsewhere (e.g. Chapman 1994). Fundamentally, the groundwater flow prediction must be most concerned with the path taken by any moving water after it leaves a repository and the rate of its movement. If the motion is such that any escaped waste gets carried to depth, and that the water mixes there (i.e. is diluted), then these conditions are favourable, and even better is the case where these movements are slow. Upwards flow paths, and the possibility of mixing with and polluting shallow aquifers – or even of discharge to the surface – are much less favourable, and rapid flows are undesired. It would obviously be ideal if groundwaters were static and any dissolved waste would not move by advective flows, but only by extremely slow diffusion.

Such static groundwaters might conceivably occur in situations with no hydraulic energy, although we are doubtful as to the existence of such circumstances. A variant on this theme is the case at Yucca Mountain, in the USA, where the intended disposal site is located well above a deep water table; here the hydrogeological concerns focus on transient phenomena (such as seismic pumping, climate change or hydrothermal activity) which conceivably might cause a drastic change in the groundwater system. A more typical view of hydrogeology is one in which groundwaters are present in a dynamic, connected system with an ever-present potential for

movement depending on a range of parameters (Neuzil 1995; Toth 1995). Accepting this paradigm, the exploration exercise becomes one of locating sites where, as groundwaters move away from the disposal site, they flow either to deeper parts of the crust, or flow only very slowly back to the Earth's surface. In simple terms, it is essential that a repository be sited in an area of very slow local and regional groundwater movements (i.e. in an area with low regional hydraulic gradients; Chapman & McEwen 1986). This could mean that an ideal site would be in the recharge zone of a major groundwater system, whose flow paths away from the site were long in both time and distance, and directed to greater depth (Bredehoeft & Maini 1981; Toth & Sheng 1996).

Even though a groundwater system may be sluggish, consideration needs to be given to perturbations which could be induced as a consequence of the creation of a repository. For example, although ILW does not, by definition, produce much heat, this energy, and that from exothermic cement reactions (combined with slow, diffusive heat dispersion), could result in the temperature of a repository volume reaching 80°C (NIREX 1995*d*). Consequently, even a naturally static water system may be induced to circulation or convection following the emplacement of waste. This point is revisited below in the context of permeability changes brought about by construction works.

Exploration for a potential disposal site needs to consider the hydrogeological system, and how aspects of the geosphere, or the engineered system, can influence it. This approach was taken during the 1980s in the UK when a number of 'types' of site were identified by the British Geological Survey starting with the identification of general principles (Gray 1976). The reasoning behind each type of generic site is summarized elsewhere (Robins 1980; Chapman & McEwen 1986; NIREX 1989). All these generic sites share the characteristic of inferred slow groundwater flow directed away from the biosphere. Very general hydrogeological characteristics were described for each such type of site, along with comments on other selection factors, such as the rock types, the infrastructure, the local economy and political matters. Of the site types which have been described, none have a specific, 'real' example location identified, and so these sites remain, in the UK, merely hypothetical.

Using a range of research results (Garven 1995; Person *et al.* 1996; Toth & Sheng 1996), it is now possible to predict the general nature of a hydrogeological system based on its geological setting. The 'setting' includes the geometries of the land surface, and the configuration of the sub-surface rocks, along with information of fluid-type distribution, heat sources, or other factors which may influence the hydraulic energy. Such hydrogeological predictions are made using numerical simulations, and this approach requires the specification of material-property distributions, and other parameters, which are not 'known' for a hypothetical site. Nevertheless, geoscientists can make reasonable assumptions about such hypothetical cases and thereby provide a means to assess the general characteristics of each such system.

A principal benefit from such a numerical simulation approach is the identification of those aspects of each system which are most important in controlling the critical behaviour (e.g. the groundwater movement, geochemistry or rock mechanics). Once these key geosphere behaviours are understood, the final stage of exploration is for the geoscientist to seek real-world settings which meet the primary geosphere criteria established through the analyses noted above. If, for example, it is concluded that the 'ideal' site is a granitic upland area which has not suffered extension, then geoscientists can set about the task of identifying potential sites, or discovering that this type of site does not exist in a given country.

It is at the stage of site selection that the different pressures acting upon geoscientists start to become apparent. The costs of site evaluation are large; consequently there is a commercial pressure to investigate those sites which are known to be politically feasible – should they prove to be geologically suitable. It is now known (NIREX 1989; RWMAC 1995) that the present site at Sellafield was not in the original short list of geologically suitable sites, but was added later as a 'variant' of the original geological concept. The choice was then made to evaluate only two of the twelve sites: Sellafield, Cumbria, and Dounreay in northern Scotland (the investigation of Dounreay was later suspended). Both of these sites were later admitted by NIREX to be less than ideal geologically, and seem to have been chosen primarily for political considerations. As stated by the Planning Inquiry Inspector: 'It seems that the

process was affected by a strong desire to locate the repository close to Sellafield' (McDonald *et al.* 1996). This strategic choice later proved to greatly complicate and limit the success of the evaluation phase and led ultimately to the site's rejection, although this has not (yet) led to a release of the locations of the other ten sites.

Evaluation

The issue of nuclear waste disposal is one demanding an independent scientific assessment by independent geoscientists. By 'independent' we mean those (usually in the academic world) who are not normally obliged to uphold the views of commercial firms as a consequence of financial support or other commitments. This notion is similar to the restrictions against material interest enjoined against prospective jurors, and the other 'conflict of interest' restrictions with which we are all familiar in our professional codes of practice.

The idea here is one which reaches to the very heart of the scientific endeavour. According to Popper (1963) the scientific method is a process by which new understanding emerges: (1) discovery of a problem (usually as a rebuff to existing theory); (2) bold solution (new theory); (3) deduction of testable propositions; (4) tests (i.e. attempted refutations); (5) preference between competing theories; and (6) back to (1) discovery of a problem. Scientific 'truth' emerges when claims withstand challenges and, especially, when others independently support those claims (Mermin 1996). The process requires the mutual availability of information, and it further requires an established mechanism for discussion and debate. The notion of 'independence' is inherent to the activities of scientists. Although the end result of science is not guaranteed to be universal agreement on interpretations, science does – following debate – produce a consensus on what is agreed, and what is still unknown. This consensus of agreement, and agreement to disagree, extends to interpretations of earth processes as well as of data.

We suggest that the scientific approach is appropriate for the evaluation of a potential nuclear waste disposal site. In terms of the evaluation stage, the 'claim' to be tested is that the site is suitable for nuclear waste disposal. If the claim, that the site is suitable, is shown to be weak, or wrong, and the challenges to the claim are upheld by further independent scrutiny, then the viability of the site falls into serious question. Alternatively, if academic geoscientists are able to independently converge on an interpretation of the geosphere at the site such that these independent interpretations support the site's viability, then the proposal gains credibility.

As is true for most of science, most geoscientists who are interested in the nuclear problem must choose the option of challenging a claim (i.e. that some site is suitable for waste disposal). An attempt to support the claim would require facilities and capabilities which are beyond the scope of academic institutions. Instead, academics most often choose to seek weaknesses in claims – perhaps this is because the scientific method is so intimately linked with challenges. The following section is an example of this approach as taken by academic environmental geoscientists concerning the (now abandoned) site at Sellafield.

Examples from Sellafield

UK NIREX Ltd proposed the construction of a nuclear waste disposal site at a location near the Sellafield reprocessing plant in Cumbria, UK. NIREX scientists evaluated the suitability of this site for many years, and later used this knowledge to apply for planning permission to construct an underground laboratory. A substantial amount of replicate work was carried out by contractors funded by Her Majesty's Inspectorate of Pollution. Along with brief overviews in annual RWMAC (Radioactive Waste Management Advisory Committee) reports (RWMAC 1994), peer reviews have been undertaken the Royal Society (Royal Society, 1994) and other groups commissioned by NIREX. RWMAC advises government on strategy and progress but states that a full peer review of NIREX work is beyond its scope.

Fluid/rock/waste geochemistry

The geochemical containment of radioactive waste in the UK is difficult for several reasons, including its complexity: (1) it contains long-lived radionuclides, such as uranium; (2) it contains radionuclides with contrasting geochemistries; and (3) it comprises a

physical mix of many different elements produced by different 'waste streams', where information on the mineralogical forms, redox states and valences is not readily available. The radionuclide uranium forms an important part of the waste streams which are awaiting disposal. In terms of inventoried mass in the waste, uranium amounts to around 2.5×10^6 kg (NIREX 1992*a,b,c*). The longevity of its radioactivity (Weigel 1986) and the potential risk from the repository (Nuclear Energy Agency 1989) dictate that the retention of uranium forms a major component of the safety assessment. Although radionuclides such as plutonium, caesium and iodine are important contributors to the radioactive waste inventory, uranium is by far the most abundant and better understood, and we emphasize its behaviour in this chapter.

The task of the geoscientist here is to predict the possible concentration of radionuclides throughout the nearby geosphere during the required 10^8 years containment period (NIREX 1992*c*; Royal Society 1994). This formidable task is associated with a requirement to construct industrial-scale systems for containing the waste within the disposal facility. The inevitable slow leakage of waste from a disposal site through time produces a source term to input into models of dispersion in the geosphere. A robust approach to waste isolation also needs to consider the containment of radionuclides by the surrounding geosphere, which requires information on the geochemistry of the natural groundwaters, and on the geochemistry of the rock in contact with (and potentially reactive with) those waters. The success of such retention is measured by a residence time in the surrounding rock. Source term and residence time are seriously degraded by a large flux of natural groundwater through the disposal facility, thus geochemical containment cannot be considered in isolation from hydrogeology.

Repositories must include a number of 'near field' barriers which are designed to prevent or retard the release of radionuclides from the waste into the geosphere, and then to the biosphere (NIREX 1993*b*). In common with ILW repository concepts elsewhere (Miller *et al.* 1994), wastes were to be immobilized in the Sellafield repository, usually in a cementitious material, packaged in concrete or stainless steel/mild steel containers, emplaced in repository vaults and back-filled with hundreds of thousands of cubic metres of

cement-based material (known as NIREX Reference Vault Backfill, or NRVB; Atkinson *et al.* 1988*a,b*; Atkinson & Guppy 1988; Atkinson 1995). NIREX Reference Vault Backfill is mainly composed of hydrated, blended Portland cements (Bennet *et al.* 1992). The detailed constitution of the phases in these cements is complicated but dominantly they are poorly crystalline and represented in modelling studies by the mineral portlandite (Atkinson *et al.* 1993).

The main rationale behind the 'cement and steel' approach is to provide physical containment in the short term (hundreds of years) and chemical containment for radionuclides over longer timescales (hundreds of thousands of years; Hooper 1995). The reducing ambient conditions produced by the corrosion of the steel and the high pH produced by the dissolution of cement were intended to promote precipitation of low-solubility phases and to promote sorption of ionic species onto mineral surfaces. Many questions still remain concerning the roles of: sorption of radionuclides to mineral surfaces; bacterial control; and the formation of organic compounds which can both aid and hinder radionuclide migration.

At the expected 80°C repository temperatures, the ambient fluid pH would be around 10 (Atkinson *et al.* 1991). During construction and operation of the repository, oxygen would be introduced, and when finally backfilled, the cement grout would contain both gaseous and dissolved oxygen in its pores (NIREX 1995*d*). This oxygen would cause the aerobic corrosion of the 10^9 moles of iron present in the repository (from mild and stainless steel packaging containers, construction materials and waste inventory; Naish *et al.* 1990). This mechanism would remove oxygen from the system and it would be expected that repository conditions would become anaerobic within 100 years of closure (Atkinson *et al.* 1993). It is claimed that corrosion of the steel would then proceed anaerobically and produce hydrogen gas and a very reducing Eh (theoretical Eh is as low as -780 mV; Haworth & Sharland 1995), although the long-term duration of this Eh is questionable (McKeown & Haszeldine 1996). However such hydrogen gas along with carbon dioxide and methane exsolving from the wastes would create a pressure inside the repository vault; this energy could force radioactive water to move away from the site, possibly to the

surface (Horseman 1996).

The problems of designing a suitable geochemical containment scheme can be illustrated by two examples: iodine and uranium. The UK waste mix will produce substantial quantities of ^{129}I, which is both highly metabolizable and radioactive; hence, it is potentially carcinogenic, and needs to be excluded from the biosphere for geologically long times. Iodine gas and iodine compounds are very soluble in water, unless combined into compounds such as CuI_2.

Attempting to control the pH and Eh, or to otherwise buffer the near-field geochemistry, are unlikely to be effective at retaining iodine in the vicinity of a disposal site since its solubility is not strongly affected by variations in these characteristics (Bishop *et al.* 1989). The only factor which might have contributed to the local retention of iodine would be the slow flow of groundwater through the alkaline concrete backfill. Once groundwaters containing iodine moved away from the facility, this element would be transported in solution wherever the groundwaters flow. If iodine gas exsolved from the groundwaters, this would be extremely mobile. NIREX (1994*b*) simulations show that iodine can form one of the major components of radionuclide release in the early life of the repository. Some NIREX simulations, which consider the excavation damage related to vertical access shafts (NIREX 1994*b*), showed that radioactive iodine could return to the surface within forty years, in an adverse case.

A full review of the large body of work associated with the geochemistry of uranium is beyond the scope of this chapter. There are many documents giving a broad account of its properties in mineral, fuel and aqueous form (Weigel 1986; Finch & Ewing 1992; Janeczek & Ewing 1992; Harvey 1995). The complexities of uranium thermodynamics are also dealt with in great detail in a wide variety of papers and reports (Langmuir 1978; Morss 1986; Lemire 1988; Bruno *et al.* 1987, 1993; Cross & Ewart 1990; Pearson & Berner 1991; Fuger 1992; Grenthe *et al.* 1992). The most important and fundamental property of the actinide element uranium is that it can exist in more than one oxidation state. This has a direct effect on its solubility in aqueous solution (Weigel 1986). Uranium in aqueous solution is known to exist in oxidation states from U^{2+} to U^{6+} However, only U^{4+} and U^{6+} are

significant in nature (Basham & Kemp 1993). In oxidizing fluids (relatively high Eh) uranium in the hexavalent form (generally agreed to be the uranyl ion (UO^{2+}; Grenthe *et al.* 1992) can complex with a very large number of ligands such as hydroxide, carbonate and sulphate. Such complexes become more soluble in aqueous solution than UO^{2+} (Langmuir 1978; Weigel 1986). These complexes can be mobilized and transported in groundwater. Conversely, reducing conditions (low Eh) encourage the U^{4+} oxidation state to dominate, uranium precipitates from solution and results in very low concentrations in mobile solutions (Nash *et al.* 1981).

Attempting to force a reduced uranium oxidation state within the repository, by means of the 'cement and steel' approach is a sensible option to lower the solubility of uranium-complex ions. However there is a problem – although the presence of iron is likely to produce an initial reduced Eh within the repository, the durability of this condition is open to serious question. The natural groundwater will pervade the repository. NIREX claim this groundwater to be chemically reducing, based on the assumption that the mineral pyrite (FeS_2) is at present coating the walls of fractures in the Borrowdale Volcanic Group. This appears to a be a strange assumption, for the Sellafield site is in the centre of Britain's largest iron oxide (hematite) ore deposit – with no record of pyrite in the ores (Rose & Dunham 1977). When we attempted to recreate these reducing conditions (using rock data from NIREX reports), we discovered that NIREX's own data did not demonstrate any pyrite at the depth of the repository (NIREX 1995*b*; Haszeldine 1996). We found that the Eh of the deep BVG groundwaters is not easily derived, a view shared by the Planning Inquiry Assessor (McDonald *et al.* 1996).

Using a geochemical modelling code, we have shown that the BVG waters – in common with many natural waters (Lindberg & Runnels 1984) – display strong redox disequilibrium, and are most probably oxidizing (McKeown 1997). We have also shown it is highly unrealistic that BVG groundwater Eh would be controlled by equilibrium with pyrite (Haszeldine 1996). Other studies which make the same assumptions as NIREX have also predicted very reducing Eh conditions in the BVG (Metcalfe & Crawford 1994). If

such assumed conditions were carried through to safety case assessments, this would result in gross underestimates of risk.

If the BVG waters are oxidizing – as we maintain – the native iron buffer in the repository (e.g. the steel) would be consumed much more rapidly than NIREX have predicted. Instead of lasting 10 000 years (NIREX 1994*c*), the buffer may only last for hundreds of years. Consequently, there would be no effective long-term engineered barrier against the mobilisation of uranium, nor any natural chemical barrier to the release of uranium from this site (McKeown & Haszeldine 1995). In summary, it can be appreciated that creating a geochemical barrier to prevent the release of radionuclides is not a simple task; this effort needs to reconcile the various reactive chemical behaviours of different radionuclides, and it still needs to consider the fundamental geological setting in which the disposal site is located. The concept of generating suitable geochemical conditions seems to be intended to make virtually any sub-surface site suitable for radioactive waste disposal. We have shown that this does not work at Sellafield, which is geochemically poorly suited for a disposal site, with no inherent natural retention capacity for some of the most problematic radionuclides.

Hydrogeology

A hydrogeological system can be segmented into three component parts – the fluid, the rock, and the energy (Fig. 12.1). Each of these aspects relates to one or more of the parameters in Darcy's porous medium equation:

$$Q = (k \bullet \nabla H)/v,$$

where Q is the flux, k is the permeability, ∇H is the gradient of the hydraulic head, and v is the fluid viscosity. Fluid energy is directly related to the ∇H term. Fluid characteristics can impact the fluid energy (buoyancy variations) as well as the viscosity.

The rock framework is directly related to the permeability distribution, but it additionally affects the fluid energy in several ways:

(1) an irregular water table will occur when the ground surface

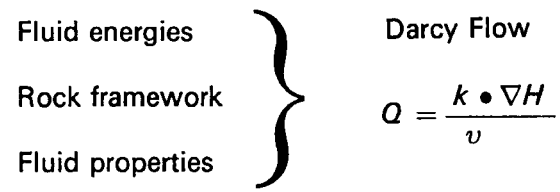

Fig. 12.1. Three of the components of a hydrogeological system in relation to Darcy's flow law.

(top boundary) is both saturated with pore water and topographically uneven;

(2) permeability heterogeneities (faults and fractures, or modern changes to pore geometries due to mineral precipitation) alter the transmission of fluid energy, by acting as flow baffles (pressure drop), or conduits;

(3) any mechanical change in the rock framework (such as neotectonics, or thermal expansion, or volumetric changes associated with construction) will alter the pore volume, and hence will affect the pressures of the contained pore fluids.

Other geosphere factors which may affect the fluid energy include: water-table variations resulting from climate change; brine intrusion; and fluctuations in heat energy. Although design temperatures at Sellafield are stated to be 'low' (80°C), even small amounts of extra heat can be sufficient to initiate convective circulation of groundwaters; such circulations would seriously affect all safety aspects related to water flux.

The simulation of fluid flow through rocks is a subject of great complexity (Huyakorn & Pinder 1983; de Marsily 1986; Cathles 1990; Deming 1994). Flow between the pore space of rocks enables groundwater flow to be described using a porous-medium continuum approach (Bear & Yehuda 1990). The presence of fluid flow in fractures can add further complications. In fractured rocks such as those of the BVG at Sellafield, the interconnected fractures are considered to be the main passages for fluid flow, with the solid rock matrix considered to be almost impermeable (Domenico & Schwartz 1990). This situation results in wide differences in

hydraulic conductivity, with fracture conductivity many orders of magnitude greater than that of the matrix (Brace 1980; Neumann 1990; Clauser 1992). Modelling flow in such a fractured system is obviously problematic (Freeze & Cherry 1979; de Marsily 1986). However, it can be achieved by: (1) modelling the fluid transport through each fracture or network of discrete fractures (Moreno & Neretnieks 1993); and (2) modelling the transport regime in the fractured mass as an equivalent porous and anisotropic continuum (Follin & Thunkin 1994).

The problem with the first approach is that the fractures/fracture networks have to be extensively mapped, with information regarding the hydraulic conductivity, connectivity and geometry exhaustively recorded (Garven 1994). For large-scale regional modelling this is impossibly arduous, although some numerical models use stochastic methods to generate statistical fracture networks (Long *et al.* 1991). In the second approach the fractured medium is viewed as an equivalent porous medium. The obvious advantage is that models of this type are well understood by a wide array of geoscientists and there are numerous implementations available (Domenico & Schwartz 1990). The high density of fracturing in the crystalline rocks of the Sellafield area precludes the need to consider individual fractures within such a regional system. Beyond such practical matters, if the spacing of fractures or fracture zones is less than the scale of numerical discretization (or grid size) in the model, then the equivalent porous-medium approach is justifiable (Garven 1995). The porous-medium approach is widely favoured for regional 2D modelling of fluid movements (e.g. Garven 1995), and it has been applied by others to the Sellafield site (Nicholls 1995; NIREX 1995*c*; Heathcote *et al.* 1996).

To investigate the NIREX proposal for a disposal site at Sellafield site, we initiated a programme of hydrogeological research. We took a very different approach to that used by NIREX in their investigations: we chose to focus on the sensitivities of the whole fluid flow system, and we sought to understand the main processes operating and to identify the key variables affecting the behaviour of the system. From NIREX reports and public, mainly British Geological Survey, information we constructed a

geological cross-section through the site. This was converted into a finite element grid which was used in a 2D computer model which simulated steady state fluid flow (Haszeldine & McKeown 1995). Each element was assigned numerical values representing the equivalent petrophysical properties (porosity and permeability). Simulations of fluid flow were run on this model; these established the likely pathways of fluid flow, and the rates of flow, and they allowed us to investigate the sensitivity of the model to changes in rock properties – both within and outwith the measured ranges of hydraulic conductivity (permeability) of the various rock units.

All steady state simulations showed that topographically-driven groundwaters would descend from the BVG uplands and then pass upwards through the repository site, above which there is less than 200 m of BVG rocks. This 'barrier' of BVG rocks is all that exists to retard the transport of radionuclides carried by the flow issuing from the repository before these materials enter the permeable clastic rocks overlying the BVG. Key parameters in the model include the regional hydraulic conductivities of the BVG, the Permian breccias and conglomerates, and the faults. These regional values are poorly known from borehole tests, with local measurements in the BVG ranging over eight orders of magnitude (NIREX 1993*a*). As noted below, this range of values, or even the lower mode of this range used by NIREX in their modelling, did not in itself assist in choosing the most realistic or 'best' values with which to represent the BVG.

In order that we could define the most appropriate rock parameters to represent the fluid flow system, we attempted to constrain our models to 'reality' by calibrating the sub-surface pressures calculated by the model, against the sub-surface pressures measured by NIREX in their boreholes (expressed as hydraulic heads). We found that the best fit to the borehole data was obtained by using a BVG regional hydraulic conductivity some 500-1000 times greater than that used by NIREX (McKeown 1997). This 'calibrated' BVG permeability was much greater than that which has been used by NIREX in establishing their safety case (NIREX 1995*c*). Consequently, the more rapid water flows we envisage to be occurring in the sub-surface (i.e. as predicted by this model) will permit leaked radionuclides to move more rapidly than has been

suggested by NIREX, and therefore the site would fail to meet safety targets (Wallace 1996; Haszeldine & Smythe 1997).

This concern relating to rapid, upwards groundwater flow through the proposed site was a common thread in evidence given in support of Cumbria County Council at the Inquiry (Haszeldine & Smythe 1996) and, even after NIREX presented all of their hydrogeological evidence, the Inspector was not convinced that the best understanding of groundwater flow rates and paths had been reached (McDonald *et al.* 1996). The hydrogeological work we have undertaken (Haszeldine & McKeown 1995; Haszeldine 1996; McKeown 1997) indicated that the Sellafield site is fundamentally a poor location, being at the outflow end of a regional hydrogeological system, rather than the recharge end (Toth & Sheng 1996). The site is also complex, with flow through rock fractures resulting in wide ranges of measured hydraulic conductivity. We consider that our approach to determining the regional, bulk, 'effective' permeability of the rock units at this site is good practice (i.e. by calibrating the model against measured data) and that BVG hydraulic conductivity values we derived represent 'upscaled' permeabilities incorporating highly-permeable fracture zones. The predictions arising from the model effectively falsify the claim that the site is suitable for nuclear waste disposal.

Although regulatory bodies such as Her Majesty's Inspectorate of Pollution (HMIP) have for many years undertaken parallel research on the suitability of the Sellafield site, it is very interesting to note that two key reports were not obtainable until the Inquiry began. Marked 'commercial-in-confidence' they detailed results from research commissioned by NIREX to look at both fluid flow (Nicholls 1995) and geochemistry (Tyrer *et al.* 1995) at the site. After NIREX ceased funding these projects they remained unpublished until it was discovered by Friends of the Earth that the information was of sufficient public interest to merit free availability. Such problems in obtaining important information highlight the possibility that, without the approach taken by independent earth scientists, there might possibly not have been a freely available alternative view on the site's hydrogeological and geochemical safety to that proposed by NIREX.

Faults and rock structure

NIREX also predicted the structure of the rocks in the area around the proposed disposal site (NIREX 1994*a*, 1995*a*). This entailed mapping the depth and orientation of the bounding surfaces of the different rock units, for example, the top of the BVG; it also required mapping the faults which cut these rocks, including their orientations and their throws. These structural interpretations were essential to the site evaluation because the safety of any disposal site rests on the ability of the geosphere to contain any released radionuclides. As we emphasized above, a critical component of the containment process is the rate of flow of groundwater past the waste, and a crucial element of hydrogeological models is the location and permeability of faults and their associated fractures. Therefore, it is absolutely essential to be able to confidently identify faults within boreholes, and to correlate these faults between individual boreholes. It is also necessary to be able to make similar interpretations of faults from other data (e.g. seismic, structure contour maps).

The first surveys and interpretations of faulting at this site were undertaken by the British Geological Survey; they identified two faults with opposing dips. A further five different interpretations of fault configurations have been published by NIREX in 1990, 1991, 1993, 1993 and 1995. Each interpretation is different, and each shows a varying number of faults, and a range of fault orientations (Smythe 1996; Haszeldine & Smythe 1997). The geological interpretation of the site is apparently subject to substantial revision each year. In a scientific context, such changes of interpretation resulting from new data or further analysis are common; but in the context of assessing a potential nuclear waste repository, these changes are worrying. Consequently, there is no reason to expect that the version of the structural interpretation which NIREX submitted (in 1995) to the Planning Inquiry is anything like the true picture.

As part of the effort to determine fault positions, NIREX funded the acquisition of nine 2D seismic reflection lines. However, NIREX reports do not contain any seismic data, but merely interpretations of those data. This means that it is impossible to make any judgement about the quality of those interpretations, or

to attempt independent analysis. Another approach which is commonly used to evaluate faulting is to correlate faults between boreholes. In the area around the proposed disposal site at Sellafield, approximately twenty boreholes have been drilled. Both from wireline logs, and from cores recovered from these boreholes, it is possible to identify faults. In some cases a fault surface can be identifiable in the core, and often the actual fault plane is surrounded by a large number of brittle fractures associated with the fault 'damage zone' (Knipe *et al.* 1997). However, the identification of a fault in a borehole does not give direct information on inter-borehole correlation of that fault. This is important for assessing the true geometry of the fault plane, and for making use of any information concerning the flow characteristics of the fault.

In order to improve the inter-borehole interpretations, NIREX also acquired seismic tomograms between pairs of boreholes. These surveys image the velocity structure of the rocks between boreholes which, in principle, would enable the changed velocity associated with a fault plane and its damage zone to be mapped, from borehole to borehole. NIREX made such interpretations of these data (NIREX 1994*a*), but there seem to be major problems. An estimate of the reliability of the existing tomograms can be made by the simple method of abutting pairs of tomograms which share a common borehole. This test reveals severe mismatches in the interpretation: that is the same fault is interpreted to cut a single borehole in different places when imaged by different tomograms. An additional problem is that, in some cases, a fault interpreted from the tomogram does not cut the borehole at the depth where core or wireline information suggests the existence of a fault. In addition, the tomogram surveys cannot be matched satisfactorily with the 2D seismic sections. Therefore, the current tomogram surveys are inconsistent and must be considered inconclusive.

In the last decade, the most important and cost-effective breakthrough in sub-surface imaging technology has been the widespread introduction of 3D seismic methods, mainly in oil field settings. This technique provides a means of imaging complex geology, resulting in a depth model of the rocks at resolutions much better than can be obtained with 2D seismic. NIREX did not carry

out a 3D seismic investigation until 1994, when Glasgow University under contract to NIREX undertook the densest such survey in the world over the proposed location of the RCF construction shaft. To date, these seismic data have not been used by NIREX to update their structural interpretation, nor have the data been released to others.

Preliminary interpretation of the 3D seismic data (Smythe *et al.* 1995) indicates that the dips on the top of the BVG (e.g. the unconformity between the BVG and the overlying cover) are consistent with the earlier 2D seismic interpretations, but dips of the lithological units within the BVG differ radically from those suggested by NIREX tomograms or NIREX structural maps. These comparisons suggest that NIREX's present maps are invalid, and that the geological interpretation of the entire site area needs to be revised. Of particular concern is the fact that the positions, orientations, and even very existence of major faults, as depicted in the current NIREX interpretation, may be unreliable. Since faults have a major impact on hydrogeological simulations, the uncertainties concerning the structural interpretation of this site mean that a safety case cannot be reliably supported.

Rock mechanics concerns arising from construction

We have focused in this chapter on hydrogeological aspects and we will not address topics concerning the design and construction of underground cavities. Instead, we address two points which are a consequence of such construction: the first is the increase of permeability in the volume around the disposal facility; and the second concerns perturbations of the water system caused by pumping of inflowing waters away from the works area during the construction phase.

Underground excavation inevitably leads to dilatancy of the rock mass surrounding the works. This is true whether the excavations are achieved through blasting or by tunnelling machine. The increased dilation of the rock mass will result in an increase in the bulk permeability. Simulations of a simple 3D hydrogeological system which is similar to that which applies to the Sellafield site reveal that such a local permeability anomaly leads to locally increased flow rates (Garven & Toptygina 1993). This increase in

groundwater flow will alter the assumptions about the total water in contact with waste containers, as a consequence of the larger number of pore-volume exchanges. More worrying is the possibility that the volume of rock with increased permeability will significantly alter the rock framework in ways that affect the larger hydrogeological system. The specific configuration of the Sellafield site is such that the volume of rock with increased dilatancy may be sufficiently large to affect the 200 m of BVG between the repository and the base of the cover sequence above. This breach could lead to a much enhanced rate of exchange of groundwaters between the 'basement' domain, the proposed disposal site, and that of the layered rocks above, which are hydraulically connected to the surface.

A further hydrogeological concern associated with the construction, and related to rock mechanics matters, is the necessity to draw down the water table during construction. This would certainly occur in the shallow rock sequence, and it might have happened in the BVG if 'open' fractures were encountered since these might have large-volume inflows. This pressure draw down would have some effect on the hydrogeological system – an effect which could be simulated for a range of assumptions about its magnitude. Less easy to simulate are the possible mechanical effects of changes in pore pressure associated with such a draw down. These simulations would require assumptions regarding the detailed state of stress in the entire rock mass – before, and during, the construction. They would also require assumptions concerning the non-linear processes of stress relief. Given that tunnelling experts are still working to understand how to improve such predictions in their work, it goes without saying that there are remaining uncertainties. All of these uncertainties multiply if we also ask whether there will be any changes in the permeability distribution as a consequence of minor structural movements associated with the pore-pressure excursions. Based on the simple situation described by others (Garven & Toptygina 1993), it becomes apparent that this matter of linked processes (e.g. alteration of the groundwater system, and consequent alteration of the rock mechanics leading to changes in the groundwater system) requires considerably more attention.

Discussion

Academic geoscientists are, by and large, supported by the public purse. Their daily task is to engage in the 'science' process which we described above. Until recently, the academic's choice of a topic for research has been largely unconstrained, subject only to the availability of time, and the necessary resources. Political and economic changes have altered this situation, such that funding, and indeed the assessment of research quality, now give strong emphasis to the benefits to society of that research, generally perceived in terms of 'wealth creation'. These circumstances are forcing a major change in the directions taken by academic geologists, and in the way that they approach their vocation.

Many academic geoscientists are adopting the mantle of 'environmental geoscientists'. We applaud this change, and we feel confident that well-trained geoscientists can readily make this transition. However, 'environmental geoscience' demands a proactive approach. Scientists can adopt either of two strategies when assessing a proposition: to seek to support the claim by means of separate studies; or to seek to falsify that claim. Both of these avenues are notionally open to us, but both require access to relevant information, and to the necessary tools. The decision to do something about an environmental issue is what distinguishes an environmental geoscientist from other geoscientists. Some academic geoscientists must continue to be in a position to serve as public 'watchdogs' on issues of importance to the public at large.

In this chapter we have focused on the 'environmental' subject of nuclear waste disposal. We have sought to make the case that there is a role for independent geoscientific analysis of proposals which are made by commercial firms, and that this independent expertise largely resides in the academic community. What is also apparent is that the 'political' issues associated with nuclear waste disposal are difficult to separate from the scientific ones. The challenges brought by independent, academic environmental geoscientists (Haszeldine & Smythe 1996), in conjunction with research by such bodies as HMIP, have highlighted potentially dangerous shortcomings of the Sellafield plans. These challenges have necessarily acknowledged political aspects of the issue while seeking to focus on the scientific

concerns.

It is now known that further site work at Sellafield has been abandoned. In part, this change of plan is related to the scientific arguments made by several parties at the Planning Inquiry, and especially to the challenges raised against the claims made by NIREX. In cost terms, the academic studies we have undertaken, and the other challenges, have consumed less than £200 000 of funding. This total compares favourably with the NIREX budget of some £400 million expended to date on the Sellafield investigation. If one takes into account the large sums which were expected to be spent on the RCF (another £400 million?), but which now will not be wasted on this unsuitable site, it would seem that an argument could be made that environmental geoscientists have proven to be of considerable benefit to society. The benefits arising from health and safety issues further emphasize the importance of having an independent point of view involved in the evaluation process.

What emerges from this argument, in terms of the Sellafield disposal scheme or others which may follow, is that society should demand the establishment of a proper scientific forum in which issues concerning nuclear waste disposal are openly debated. This process of debate has served science extremely well over many centuries, and we submit that it would serve society well in this matter of major environmental concern. The forum should be open to all interested parties. Both the availability of information, and funding, are issues which need to be resolved in order to make this a viable approach.

The discussions in the forum would almost certainly be vigorous, and the issues might not be quickly resolved. Indeed, the debate would be likely to reveal new research questions which are critical to the issue of finding a suitable site. Nevertheless, we all now recognize that something must be done with the existing waste, and the entire community can seek a solution which is the best possible one in the circumstances which exist. As a society we owe our descendants no less than this.

The plea we make concerning a forum to debate nuclear waste disposal will necessitate a supply of qualified academic, and commercial, environmental geoscientists. Other waste and pollution matters – both existing and yet to be uncovered – will likewise

require both knowledge, and the skills to apply that knowledge to solve real problems. We see a solid future for well-trained geoscientists who understand the way the Earth works, its processes, and particularly, how processes interact, and how to do something about environmental issues.

Conclusions

1. Decisions made at the exploration stage when seeking a disposal site can be subject to political considerations which may ultimately jeopardize the technical success of the entire exploration, appraisal and licensing effort.
2. The scientific approach (i.e. refutation or verification) offers the best method of peer review. This process is achieved by means of distinct groups of workers undertaking parallel investigations to evaluate the interpretations and claims made by the companies which wish to develop a waste disposal site. In the early stages, there is likely to be an emphasis on refutation but, as the proposals are modified in light of the debate, it is hoped that there will be a convergence in assessments.
3. A judicial approach in deciding whether a safe site has been selected needs well-informed 'defence' and 'prosecution' cases to be formulated.
4. The waste disposal site at Sellafield was poorly chosen at the exploration stage. Insufficient consideration was given to geological factors, particularly the hydrogeology. Subsequent investigations during the evaluation of the site have proven expensive, and complex. Simulated groundwater flow at this site is upwards, with the potential for carrying radionuclides towards the surface.
5. The geochemistry of the site was not adequately considered at the exploration stage. The natural chemical environment is possibly adverse to the retention of uranium, even in the engineered repository. The potential for retention of other radioactive elements is also unclear (e.g. iodine).
6. Sellafield was a very poor site which, if efforts had continued there, would have been unnecessarily expensive to character-

ize, and would still not have met the regulatory target for safety, due to fundamental flaws in the geological characteristics of this location.

7. Academic earth scientists have had a key role in evaluating this site and in synthesizing and interpreting information from the evaluation investigation. They have been important in upholding the public interest on topics which are beyond the knowledge of much of society.

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13 Airborne particulate characterization for environmental regulation

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Summary

- World-wide concerns over air quality have accelerated research in the growing field of airborne particulate characterization.
- The Earth Resources Centre, University of Exeter has developed novel dust characterization procedures during a major research programme with British Coal Opencast and has subsequently implemented the methodology whilst working with local authorities in England and Wales.
- This chapter describes the implementation of these airborne dust studies for effective regulation and illustrates how the science of environmental geology in air quality is disseminated to both the specialist decision-makers and to the non-specialist.

Airborne particulates reach the Earth's ambient atmosphere from a wide range of sources. A relatively small contribution is derived from the destruction of meteorites whilst terrestrial sources include aerosols from oceanic sea sprays and breaking waves, minerals derived from agriculture and exposed soils, fires and volcanic activity, as well as anthropogenic sources, such as civil engineering, industrial and traffic emissions (Pye 1987).

The size of airborne particulates can range from 0.05 to 1000 μm (microns). Although particles from a few microns to around 100 μm are difficult to resolve by the naked eye, they can cause discoloration of surfaces such as cars, washing and window ledges and are, therefore, categorised as nuisance dust and as such are of ongoing concern to the regulatory authorities (Anon. 1996).