Preface

Radioactive wastes contain a wide spectrum of radionuclides arising from commercial power production, industry, medical applications and defence-related activities. In the UK today, much of the low-level radioactive waste that is generated as a result of these activities is conditioned and packaged as it arises, prior to transport to a near-surface disposal facility at Drigg, West Cumbria. However, the storage and disposal options for other longer-lived solid low-level, intermediate-level and high-level radioactive wastes, as well as for other materials, such as separated plutonium, that could eventually be declared as wastes are still to be decided.

A leading option for the long-term management of various types of radioactive wastes is deep geological disposal. In the UK the radioactive waste management agency United Kingdom Nirex Limited (Nirex) has been investigating issues concerned with the deep geological disposal for more than 20 years and, as part of its research and development programme, Nirex has developed a Phased Geological Repository Concept. In this concept the wastes are to be emplaced in a repository constructed at several hundred metres depth in an appropriate host geological formation, but importantly the phased repository concept includes the potential for retrieving the wastes and, by relatively straightforward means, having a capability to monitor them for extended periods of time (up to several hundred years).

Over the very long timescales that need to be considered in post-closure radiological safety assessments much of the activity initially associated with the waste will reduce to negligible levels over relatively short periods of time (hundreds to several thousand years). However, due to the long half-lives associated with certain radionuclides some would remain hazardous for very much longer periods of time. Although any releases of radionuclides that might eventually be released from a deep geological repository would be in very low concentrations they would largely be precluded from migrating to the inhabited environment (the biosphere) by a number of physical and chemical barriers. These barriers relate to the engineered facility and to the characteristics of the surrounding host and overlying rock. For example, the engineered facility can provide both physical and chemical containment, e.g. by limiting degradation of the waste form and by inducing a chemical environment that suppresses solubility and enhances sorption of many of the radionuclides in the waste. Similarly, the host and overlying rock can inhibit radionuclide transport by slow or non-existent groundwater flow, by sorption of radionuclides onto mineral surfaces, and by diffusion of radionuclides into the rock matrix. Any low concentrations of radionuclides that do migrate away from the immediate repository and host rock environment would be further reduced over time by dispersion and mixing with overlying groundwaters. Radiological safety assessments are typically undertaken by both radioactive waste management agencies and by the regulatory authorities to evaluate the potential and consequences for any such discharges.

As a consequence of the effects of solubility limitation and sorption, many of the radionuclides that are of environmental interest in the context of effluent discharges from nuclear power plants, e.g. ^{137}Cs, ^{239}Pu and ^{241}Am, do not generally feature as a concern in safety assessments of deep geological repositories for radioactive wastes. Rather, the emphasis is on the very long-lived radionuclides that are highly soluble in repository conditions and that are adsorbed to only a very limited degree on repository materials or the host and overlying rock. In particular, radionuclides that are released and transported in anionic form tend to dominate the radiological impacts determined in post-closure safety assessments. As a result of the assessment activities undertaken as part of the Nirex research programme and elsewhere, ^{36}Cl, ^{79}Se, ^{99}Tc and ^{129}I in particular have been identified as being of interest as over the very long timescales that typically need to be investigated (up to and beyond one million years) they have the potential to migrate to the biosphere.

In assessment scenarios where radionuclides do migrate to the biosphere, a further consideration relates to the way in which the accessible environment could become contaminated. In the past, the emphasis has been on releases of radioactive materials to the atmosphere, from weapons' testing in the 1950s and 1960s, the routine operations of the commercial nuclear fuel cycle, and nuclear accidents. Therefore, in considering contamination of soils and subsequent transfers through the food-chain, the emphasis has been on radionuclide deposition from the atmosphere. However, in the context of the geological disposal of radioactive wastes, the routes by which the accessible environment may become contaminated include abstraction of groundwater, for use in irrigation and for other purposes, and the natural discharge of groundwater from depth to soils and the aquatic environment. Contamination by irrigation may be treated somewhat analogously to deposition from the atmosphere. However, contamination by natural discharge presents substantially different considerations and it is those considerations, as they apply to the radionuclides of particular interest in solid radioactive waste disposal, that Imperial College has explored in an extensive experimental programme over the last eighteen years.

When the Imperial College experimental programme began, the approaches used in radiological assessments to relate radionuclide concentrations in soil to uptake by plants were generally highly simplified and empirical. With the emphasis being placed on uptake following surface contamination, the standard approach was to average the concentration of a radionuclide in soil over the depth of the rooting zone and then to estimate the concentration in plants by applying a concentration ratio. This concentration ratio was estimated from experimental studies (often conducted in pots in laboratory conditions) or from observations of stable element concentrations in soils and plants, where it was felt that stable element observations could legitimately be applied to radioisotopes of those elements.

However, with contamination of the soil from below and with the need to utilise results from a moderately extensive experimental programme in a wide range of potential environmental contexts, the existing empirical approach was felt to be inadequate. For this reason and from the outset, the experimental programme at Imperial College was complemented by an extensive, process-based mathematical modelling programme directed first to understanding the processes underlying the experimental results obtained, but subsequently also to determining the implication of those results in an assessment context. Thus, hydrological and contaminant transport models were created that could take into account changes in the hydrological and hydrochemical environment of the soil, both with depth and over the course of time, and the development of the rooting systems of plants over time in that changing hydrological and hydrochemical environment.

In turn, this emphasis on process-based modelling strongly influenced the nature of the experiments that were undertaken. Previous experimental studies had typically paid little regard to either hydrological conditions in the soil or variations in the oxidation-reduction regime with depth. In contrast, in the Imperial College experiments, comprehensive hydrological monitoring was included from the outset, with provision made to measure both soil water content and potential. In addition, techniques were developed to measure the development of root profiles. In later studies, the capability was developed to extract small volumes of soil solution during the course of the experiments, so permitting a full suite of chemical and radiochemical analyses to be conducted.

Initial studies carried out at Imperial College were conducted in ambient conditions in lysimeters with a cross-sectional area of about $1.6\,\mathrm{m}^2$ and a water table that was controlled such that it was maintained at a fixed depth. Subsequently, other experiments were undertaken on substantially smaller soil columns in a laboratory with a controlled environment. Through process-based modelling, it was shown that parameter values determined in the lysimeters could be transferred to the column experiments and *vice versa*. This justified the later stages of the programme exclusively utilising column experiments. The adoption of soil columns as an experimental basis allowed a wider range of conditions to be explored than would have been possible in the lysimeters. Thus, the column studies included comparisons of radionuclide behaviour in different soil types, in intact *versus* repacked soil, with fixed and sinusoidally varying water table heights, and in the presence of different concentrations of stable chlorine isotopes. In the most recent experiments, the system has been further augmented by the inclusion of supplementary experiments in mini-columns, which provide additional insights into the influence of different degrees of water saturation and redox conditions on radionuclide behaviour.

The Imperial College studies should not be seen in isolation. When the Nirex biosphere research programme was initiated in 1987, a structured review was undertaken to identify those areas in which it would be appropriate for Nirex to undertake research to complement that being undertaken for a variety of purposes by other organisations. As described above, one of those areas was determined to be a detailed study of the transport of key radionuclides in soil and uptake by plants when contamination occurred from below. Other areas included the influence of climate change and landform development on the accessible environments into which radionuclides might emerge far in the future, the development and application of 3D catchment models to represent the transport of radionuclides in such environments, and evaluation of the adequacy of existing models of the development of ice-sheets for use in assessment studies. As with the Imperial College experimental and modelling studies, research in these other areas has been consistently funded by Nirex and research in many of these areas continues, both at a national level and through internationally funded programmes.

In a programme extending over eighteen years, it was inevitable that results of the Imperial College work would initially appear in a variety of Imperial College and Nirex reports, as well as in journal articles and conference papers. Fortunately, the team involved in the research has been extremely stable throughout that period and several of the founder members are still involved. Furthermore, from the outset, both the experimental and modelling work was undertaken within a rigorous quality assurance framework, which has meant that all the relevant experimental and modelling results remain readily available for scrutiny. Building on these secure foundations, it has been possible to pull together all the work that has been undertaken into this authoritative monograph. Nirex is proud to have sponsored such an innovative programme of work and hopes that this publication will provide an ongoing useful resource to members of the radioactive waste management and radioecological communities. In addition, it should be of considerable relevance to both research workers and commercial organisations involved in related areas, e.g. to those studying the behaviour of heavy metals in the environment.

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BIOSPHERE IMPLICATIONS OF DEEP DISPOSAL OF NUCLEAR WASTE
- The Upwards Migration of Radionuclides in Vegetated Soils