

## CHAPTER 1

# Introduction

Radioecology is still a relatively young science, having originated after the development of nuclear weapons at the end of World War II, and is concerned with both effects and pathways of radionuclides in the environment, particularly the latter. Initially, pathways research was concerned with routes through food chains by which human exposures could arise from radionuclides deposited as fallout from nuclear weapons testing in the atmosphere (Scott Russell, 1966). Following the international moratorium on such tests in 1963, attention was switched to pathways to humans after operational releases or accidental discharges from nuclear installations, particularly power stations. In the case of accidents, an enormous volume of research was carried out following the widespread deposition of radionuclides over Europe as a result of the Chernobyl disaster in 1986 (Savchenko, 1995; Warner & Harrison, 1993; Desmet *et al.*, 1990). Much of this was concerned with pathways in semi-natural ecosystems, including potential routes to humans via food chains (Shaw & Bell, 2001; Desmet *et al.*, 1990; Bell & Shaw, 2005). Most of this research was concerned with radioactivity deposited from the atmosphere onto both native vegetation and crops. The vast amount of data generated permitted a large number of calculations of soil-plant transfer factors, which are defined as radioactivity per dry weight of various plant organs divided by the radioactivity per dry weight of soil within the rooting zone (Shaw & Bell, 2001). Transfer factors have been collated in a data base set up by the International Union of Radioecologists (IUR) (Frissel, 1992) which thus

provides a compilation of the efficiencies of different plant species in taking up individual radionuclides from a range of soil types; in all cases there is a range of values, depending on soil, climate and biological factors. In addition, a large number of models have been developed which predict the distribution and transport of radionuclides in air-soil-plant systems (Thiessen *et al.*, 1999).

With the passing of the years, a large inventory of nuclear waste and contaminated areas and artefacts has built up in all developed and some developing countries. This ranges from low level waste, consisting of items such as contaminated gloves, laboratory coats and laboratory equipment, to intermediate level waste which largely consists of components of fuel elements from nuclear reactors, to high level waste which comprises both spent fuel and some reprocessing wastes. Currently in the UK, low level waste is disposed of by placing it in sealed containers in engineered vaults at Drigg on the Cumbrian coast, in a facility operated by the British Nuclear Group (BNG) (formerly British Nuclear Fuels). However, the capacity of this facility is limited and it will be filled in the foreseeable future. In the case of high level waste, this is stored in a concentrated vitrified form in special buildings at the BNG site at Sellafield, Cumbria pending a national policy decision on disposal. Currently there is also no disposal route for intermediate level waste in the UK. Radioactive waste management has been a major government policy issue for over 20 years. In the 1970s considerable use was made of deep sea disposal, but this ceased when an international voluntary moratorium on such action was agreed in 1983, under the London Dumping Convention. In the 1980s the government initially investigated additional shallow disposal methods, but this provoked widespread opposition, which resulted in plans for the development of such a sites for low level waste being abandoned in 1987. This was accompanied by a policy change in favour of a deep repository for both intermediate and low level waste. Initially, some 500 possible sites for this were considered, but this was reduced to a shortlist of 12 and subsequently to just two viz. the nuclear sites at Sellafield and at Dounreay on the north coast of Scotland.

In 1982, UK Nirex was set up by the government to implement a strategy for disposal of low and intermediate level wastes. Since 1988, Nirex has funded extensive research into the deep disposal of such wastes under its Geosphere and Biosphere Research Programmes. The latter concerns the study of processes in the near-surface and surface environment that might affect radiation doses to humans over very long time periods as a result of radionuclides migrating upwards by various routes to reach the surface and entering into food chains. The aim of this research has been to develop appropriate tools and data-bases for the safety assessment of a deep repository.

Currently (2005) in the UK the issue of radioactive waste management is being investigated by the government's recently established Committee on Radioactive Waste Management (CoRWM). This has reviewed all possible options, including some as bizarre as shooting packages of waste into space and transmutating certain radionuclides to other elements. At the time of writing, these options have been whittled down to thirteen, three of which involve deep geological disposal, which is the route favoured by most experts around the world, but not necessarily by the environmental movement and the general public.

Since 1988 Imperial College has had a multi-disciplinary team contracted to Nirex and working on the Biosphere Programme. This team has brought together experts with a range of skills and experience, which has facilitated the essential multi-disciplinary approach to this complex problem. It has involved a number of College departments, these currently being the Department of Civil and Environmental Engineering, the Centre for Environmental Policy and the Division of Biology. In Civil Engineering, the research has been conducted in the Environmental and Water Resource Engineering Section (Professor H. Wheater and Dr. A. P. Butler), which has worked for many years on a whole range of hydrological studies, with strong emphasis on modelling groundwater flow. In both Biology and Environmental Policy (Professor J. N. B. Bell, Professor G. Shaw and Dr. D. Ashworth) experimental research has been

conducted since 1980 on the migration of radionuclides into soil from surface deposition and subsequent uptake into crops, representing situations following operational or accidental releases from nuclear installations.

This ongoing work consists of both experimental and numerical modelling work on the upward movement in the unsaturated zone of a suite of radionuclides that are of radiological concern in terms of what might reach the surface from a deep repository.

The context for this work is the assumption that on the time-scales of relevance to radionuclide safety assessment, physical containment and chemical containment in an engineered repository will break down. Groundwater then becomes a potential transport pathway of radionuclides to the near-surface environment. Risk to humans is affected by direct ingestion of contaminated plant material, or indirectly, through ingestion by animals, hence an important element of the safety assessment of subsurface disposal of radioactive wastes is the movement of radionuclides through soils and their uptake by vegetation.

At the outset of the Biosphere research programme it was recognised that this was an area of considerable process complexity. Upwards migration of contaminants in soils is determined by the interactions of the soil-plant-atmosphere system that determine the flux of water in response to plant root uptake. Solute movement can occur by advection, or by root uptake and translocation, and is strongly influenced by geochemical interactions with the soil. In turn, these geochemical interactions are strongly influenced for certain radionuclides by the redox status of the soil, in a zone where strong redox gradients occur across the interface between the saturated zone of groundwater, and the unsaturated zone of the soil water system.

Given this context, it was felt that the representation in safety assessment procedures, largely based on soil-plant transfer factors derived from atmospheric deposition, was simplistic, and that appropriate data to support the safety assessment (in particular concerning upward migration of radionuclides) did not exist. A programme of research was therefore developed at Imperial College with the

following strategic goals:

- (a) To develop an appropriate level of scientific understanding of soil migration and soil-plant transfer processes to support a credible safety assessment case for subsurface disposal of radioactive wastes.
- (b) Development of models to transfer results from the research to safety assessment.
- (c) Provision of guidance on appropriate parameter ranges and parameter uncertainty for safety assessment.

The experimental work has involved outdoor lysimeters and columns in a controlled environment in which the radionuclides are injected into a simulated water-table towards the base of these containers and upward movement measured over a time course. A generic crop is grown in the soils which fill the containers, this being either winter wheat (*Triticum aestivum* L.) or perennial ryegrass (*Lolium perenne* L.). A key element of the experimental programme is the measurement of uptake of the radionuclides into the roots and aerial parts of these crops, this being the most likely route for human detriment. The second part of the programme is a series of numerical modelling exercises designed to simulate the movement upwards of radionuclides through the soil/plant system, on a time course basis, with a root uptake model being central to this part of the exercise. Such modelling is applied to both the lysimeter and column experiments, as an essential tool in the analysis of complex data sets, and as a means of generalisation of the results for safety assessment.

The experimental programme has been carried out continuously since 1990 in eight phases. The first two phases were conducted in lysimeters from 1990 to 1993, followed by a single phase II lysimeter experiment in 1995/6 (the data from the Phase 1 experiments were subsequently used as the basis of an IAEA model intercomparison project (Butler *et al.*, 1998)). After the end of the first year of the latter phase there was a policy change at Nirex, which resulted in support for the complex and expensive lysimeter studies being withdrawn. Instead future work was conducted in columns in a controlled environment cabinet, which had the same basic experimental set-up

as the lysimeters, but on a much reduced scale. The column studies covered Phases III to VIII, with some ancillary research, as well as pot and mini-column studies. Phase II involved the comparison of radionuclide migration in two soil types. However, the columns provided a greater degree of flexibility. This permitted examination of a wider range of soil types, including French soils, as well as use of intact cores on occasion. The French soils were included because Phase VIII, which studied the behaviour of  $^{75}\text{Se}$ , was supported by both Nirex and Andra, the equivalent radioactive waste management authority in France. The radionuclides studied at different stages throughout the programme were  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{75}\text{Se}$ ,  $^{36}\text{Cl}$ ,  $^{99\text{m}}\text{Tc}$ ,  $^{125}\text{I}$ ,  $^{22}\text{Na}$ , and  $^{60}\text{Co}$ . A summary of the eight phases is given in Table 1.

This book gives an overview of the entire 17 years' period for which Nirex and more recently Andra have funded the programme. To the best of the authors' knowledge, this is a uniquely comprehensive programme and remains the definitive research elucidating the behaviour of the upward migration of radionuclides in the unsaturated zone of soils and their associated vegetation. While it is specifically concerned with radionuclides, the techniques employed and the lessons learned have direct relevance for upward movement of other contaminants from groundwater and are thus of interest to research on contaminated land. Furthermore, a vast amount of information has been generated on plant uptake, which may be applied to the development of phytoremediation as a tool for the management of contaminated sites.

While the experiments and modelling described in this book are aimed primarily at the safety case for British, and to a lesser extent French, waste disposal, the results are applicable to other countries where in all cases some form of burial will be the ultimate radioactive waste management solution. Of particular importance is the decision by the USA Federal Government to propose Yucca Mountain in Nevada for a massive subterranean vault for waste disposal in the unsaturated zone (Umstadter & Baciak, 2002). This has generated its own research programmes, including on transport of fission products and actinides (e.g. Patera *et al.*, 1990). Other research into sub-surface disposal has been carried out in Sweden, (S.K.B., 1998,

**Table 1.** Summary of experimental programme, 1990–2004.

Phase	Year	Container	Soil	Radio-nuclide	Crop	Water-table
I	1990/1 1991/2 1992/3	Lysimeters	Silwood	<sup>99</sup> Tc, <sup>36</sup> Cl, <sup>22</sup> Na, <sup>137</sup> Cs, <sup>60</sup> Co	Winter wheat	Fixed — 2 levels
II	1995/6	Lysimeters	Silwood; Longlands Farm	<sup>22</sup> Na, <sup>137</sup> Cs, <sup>36</sup> Cl	Ryegrass	Fixed
III	1997/8	Columns	Silwood; Wellesbourne; Robertgate (undisturbed cores)	<sup>22</sup> Na, <sup>134</sup> Cs, <sup>36</sup> Cl	Ryegrass	Fixed
IV	1999/2000	Columns	Silwood	<sup>36</sup> Cl + Stable Cl.	Ryegrass	Fixed
V	2000/01	Columns	Silwood	<sup>125</sup> I	Ryegrass	Fixed and fluctuating
VI	2001	Columns	Silwood	<sup>99m</sup> Tc	Ryegrass	Fluctuating
VII	2002/3	Columns	Silwood	<sup>36</sup> Cl, <sup>125</sup> I	Ryegrass	Fixed and fluctuating
VIII	2003/4	Columns, mini-columns, pots	Silwood; Meuse/Haute- Marne clay loam and sandy loam	<sup>75</sup> Se	Ryegrass	Fixed

1999a, b), Switzerland (NAGRA, 2002; Nuclear Energy Agency, 2004) and Finland (Posiva, 1999). Some more general references on this topic are Chapman & McCombie (2003), Nuclear Energy Agency (1999) and Savage (1995). There is no centralised information on studies of this type, with the results of the studies being scattered in reports and published papers. In fact the most credible programmes are largely focussed on the geosphere. A limited number of studies have examined to some extent upward migration. Examples of these are the work of Torok *et al.* (1990) at Chalk River, Ontario, who carried out laboratory scale lysimeter experiments with simulated waste forms placed in potential materials for

a low-level radioactive waste repository, with development of associated transport models. At least 2 studies have examined the role of burrowing animals (Arthur & Markham, 1983) and earthworms (Muller-Lemans & Van Dorp, 1996) in the upward movement of radionuclides. However, very little research has been conducted on upward movement into vegetation, a notable exception being that reported by Murphy & Johnson (1993), examining the uptake of  $^{99}\text{Tc}$  and  $^{137}\text{Cs}$  into grass, crops and trees from a planned low level waste repository at Savannah River, South Carolina. An important development in recent years has been the BIOMASS project, which has developed a set of “Reference Biospheres” for solid radioactive waste disposal, for use in safety assessments of repositories (Crossland *et al.*, 2005). This was established under the auspices of the International Atomic Energy Agency, and involved regulators, waste disposers and independent experts from around the world.

It will be seen that the information on soil-plant transfer of radionuclides due to upwards migration is limited and diverse, and hopefully the work presented here, despite its UK origins, will be seen as a unified and significant contribution to the international literature.

Following this introductory chapter, the book consists of two chapters in which the methods used are discussed and defined, namely Chapter 2 — Experimental design and protocols, including the physical and chemical characteristics of the soils; Chapter 3 — Modelling. Experimental and modelling results are integrated by radionuclide in Chapters 4 to 9; Chapter 4 — Radiochlorine; Chapter 5 — Radioiodine; Chapter 6 — Technetium; Chapter 7 — Radioselenium; Chapter 8 — Radiocaesium, radiocobalt and radiosodium. Finally Chapter 9 presents the conclusions and recommendations. An Appendix provides information on web-based access to data files from key parts of the work.