THE FOOD CHAIN TRANSFER OF RADIONUCLIDES THROUGH SEMI-NATURAL HABITATS

Thesis submitted in accordance with the requirements of the University of Liverpool for the degree of Doctor of Philosophy by David Copplestone.

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BOOK IN ENCLOSURE AT THE BACK.

The Food Chain Transfer Of Radionuclides Through Semi-Natural Habitats

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The behaviour of ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in food chains in three semi-natural ecosystems: a coniferous woodland, a salt marsh and a sand dune have been investigated. These sites, within the environs of the Sellafield nuclear complex in Cumbria, are subject to both anthropogenic and historic inputs of radioactivity. Inputs to these ecosystems have been assessed, as well as activity in soils, vegetation, litter, invertebrates and small mammals.

Soils exhibited high activity levels for all radionuclides in the order: Sand dunes < Lady Wood < River Esk, with the River Esk containing between 5 and 10 times the activity of Lady Wood soils and 10 times that of the sand dunes. ^{238}Pu:$^{239+240}Pu$ ratios were low in Lady Wood and characteristic of airborne deposition during the early years of operation of Sellafield. Up to 20% of the caesium deposit originates from Chernobyl. The inventory of radionuclides in Lady Wood declines along a linear transect away from Sellafield. Activity in surface soils from the sand dune ecosystem reflects the predominant influence of sea to land transfer involving re-suspension of the sea surface microlayer.

Activity in vegetation from Lady Wood ranged from: 1-5 Bq kg^{-1} (^{134}Cs), 65-280 Bq kg^{-1} (^{137}Cs), 0.3-1.5 Bq kg^{-1} (^{238}Pu), 0.8-8 Bq kg^{-1} ($^{239+240}Pu$) and 0.6-16 Bq kg^{-1} (^{241}Am). Marram grass (*A. arenaria*) and red fescue (*F. rubra*) from the sand dunes showed radionuclide concentrations of 20-70 Bq kg^{-1} (^{137}Cs). Actinide levels were similar at both sites: 1-5 Bq kg^{-1} (^{238}Pu), 10-30 Bq kg^{-1} ($^{239+240}Pu$) and 10-65 Bq kg^{-1} (^{241}Am). Activity levels declined exponentially within a few hundred metres of the shoreline. Activity in vegetation from the salt marsh reflects the intensity and periodicity of tidal inundation and ranged from 10-80 Bq kg^{-1} (^{137}Cs), 2- 21 Bq kg^{-1} ($^{239+240}Pu$) and 3- 54 Bq kg^{-1} (^{241}Am). Senescent material showed higher levels of activity.

Pine needle litter in Lady Wood shows elevated levels of all radionuclides measured, ranging from 130-1,100 Bq kg^{-1} (^{137}Cs), 15-110 Bq kg^{-1} ($^{239+240}Pu$), and 18-105 Bq kg^{-1} (^{241}Am). In the salt marsh, activity levels in strand line debris were: 180-600 Bq kg^{-1} (^{137}Cs), 20-60 Bq kg^{-1} (^{238}Pu), 80-250 Bq kg^{-1} ($^{239+240}Pu$) and 150-450 Bq kg^{-1} (^{241}Am).

The detritivorous fauna showed higher values than for other taxa, for example from Lady Wood: 43- 1,800 Bq kg^{-1} (^{137}Cs), 1-14.5 Bq kg^{-1} (^{238}Pu), 6.5-74 Bq kg^{-1} ($^{239+240}Pu$) and <0.3-400 Bq kg^{-1} (^{241}Am). In the case of ^{137}Cs and ^{241}Am, the highest values were for Oligochaetes and Isopods.

Activity levels in wood mice (*A. sylvaticus*) from the woodland site were 7-150 Bq kg^{-1} (^{137}Cs). At the salt marsh, activity levels for ^{137}Cs in common shrews (*S. araneus*) ranged from 7-81 Bq kg^{-1} compared to 4-48 Bq kg^{-1} for wood mice (*A. sylvaticus*) and field voles (*M. agrestis*). These values were all significantly ($p < 0.01$) greater than for the reference site. Activity levels for ^{137}Cs in all three small mammal species were also significantly higher along the sand dunes than at the reference site ($p<0.01$) and ranged from 13-26 Bq kg^{-1} (*A. sylvaticus*), 11-13 Bq kg^{-1} (*M. agrestis*) and 42-72 Bq kg^{-1} (*S. araneus*). ^{137}Cs concentrations in shrews were significantly higher than those for the other two species ($p<0.01$). Actinide levels in *A. sylvaticus* at all three sites were little different from the reference site.

Pilot studies into tissue distribution of radionuclides in small mammals from Lady Wood showed that the muscle, especially, and pooled viscera (kidneys, liver, spleen, heart and sex organs) were centres of accumulation of ^{137}Cs. Low activity was recorded in the skeleton and gut contents.

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CONTENTS

Chapter One
INTRODUCTION

The Chernobyl accident in April 1986 focused much attention on the effects of environmental releases of radionuclides, following either catastrophic events or through the routine operations of nuclear establishments. The Chernobyl accident has been described as the greatest technological disaster in the world (Savchenko, 1995) and is certainly the worst nuclear incident that has occurred. The failure of No. 4 reactor of the Chernobyl power plant in the Ukraine Republic of the USSR, and the resultant explosions which occurred on the 26 April 1986, permitted the uncontrolled release of radioactivity into the environment. Much of this was deposited locally, within a radius of 30 km of the plant, but a radioactive plume was also released, dispersed under the control of the climatic conditions prevailing at that time, resulting in the widespread deposition of radioactivity throughout many countries in Europe.

The consequences of this disaster are still being determined but current estimates suggest that the eventual toll from cancer and cancer-related deaths will range from 14,000 to 475,000 people (Savchenko, 1995). Moreover, 144,000 hectares of agricultural land and 492,000 hectares of forest were withdrawn from use during the first years after the accident (Savchenko, 1995) as a direct consequence of being contaminated. A total ban on many of the agricultural products produced in the region has been imposed and a number of counter measures implemented to reduce the contamination of the agricultural land (Appleby and Luttrell, 1993). In addition to the human cost, much ecological damage has arisen with the wild animal and plant populations of large tracts of land being destroyed (Savchenko, 1995; Warner and Harrison, 1993).

Prior to the accident at Chernobyl, interest in radioecology had concentrated on the behaviour of radionuclides within particular components of ecosystems, primarily as a consequence of the atomic weapons testing in the 1950s and 1960s but also from the releases surrounding those nuclear establishments producing weapons-grade plutonium; for example the Oak Ridge National laboratory in the USA (Auerbach, 1987). Previous research has examined the transfer of radionuclides through most types of ecosystem, for example, grasslands, woodlands, tundra but was limited to the potential effects on humans. Moreover, it has been demonstrated that some of the conclusions regarding radionuclide transfer derived from weapons testing fallout are not applicable to Chernobyl fallout (Joshi, 1987). Nevertheless, these early studies do provide detailed information on the behaviour of radionuclides within and between components of ecosystems, for example the plant uptake of radionuclides from contaminated soils (Evans and Dekker, 1966; Graham, 1958; Menzel, 1965; Shanks and DeSelm, 1963; Tensho *et al.*, 1961) or from the foliar application of radionuclide aerosols (Merten and Buchheim, 1967; Middleton, 1958; Witherspoon and Taylor, 1970, 1971).

However, the Chernobyl accident emphasised the lack of knowledge about the behaviour of radionuclides within ecosystems as a whole. Savchenko (1995) reported that *'even 10 years after the accident there is still much to learn and understand about the after effects of the accident particularly with regard to the holistic effects of the accident both to the environment and human health'*. Moreover, most of the current research in radioecology has focused upon the consequences of the pulse of environmental radioactivity deposited as a result of the Chernobyl accident (Rudge *et al.*, 1993a, b) and in the development of counter measures to reduce the availability of the radionuclides for uptake and transfer to man (Alexakhin, 1993). For example, counter measures tested and employed around the Chernobyl power plant include changes in land use and species grown, and the mechanical and chemical removal of radioactivity (Alexakhin *et al.*, 1993; Hove, 1993; Jouve *et al.*, 1993; Maubert *et al.*, 1993; Smolders and Merckx, 1993; Voigt, 1993; Wilkins *et al.*, 1988). These measures were also used in forested areas (Guillitte and Willdrodt, 1993) and to reduce the transfer of radionuclides to game and other foodstuffs in areas of semi-natural habitat (Howard, 1993).

Most of the radioecological research and environmental monitoring programmes around nuclear establishments have focused on the impact of released radioactivity on human health, primarily through the study of agricultural food chains and the dispersion of radionuclides throughout the marine and terrestrial environments. In particular, there are a number of studies which have investigated the food chain transfer to man of radionuclides deposited on pasture and crops (Cawse and Turner, 1982; Krieger *et al.*, 1967; Simmonds, 1983; Summerling, 1983), and the uptake of radionuclides released to the marine environment, particularly to shellfish (Hamilton and Clifton, 1980; McDonald *et al.*, 1993). From this work, theoretical models have been derived to estimate population doses to man around nuclear establishments (Crick and Simmonds, 1984; Crout *et al.*, 1990; Jackson *et al.*, 1985; Jones *et al.*, 1995a; Koch and Tadmor, 1986; Nair *et al.*, 1983; Simmonds *et al.*, 1979; Thorne and Coughtrey, 1983, Willans *et al.*, 1994). In addition to direct uptake of radioactivity through agricultural products, several studies have investigated the radionuclide content of 'wild foods' such as berries, mushrooms, grouse, geese, rabbits and deer (Holm and Rioseco, 1987; Liden and Gustafsson, 1967; Lowe and Horrill, 1986, 1988).

In a similar way, much of the research arising from the routine operations and accidents at nuclear power stations has led to the development of models to predict the likely dispersion within- and deposition of- radionuclides from, for example, the atmosphere: (Al-Khayat *et al.*, 1992; ApSimon *et al.*, 1990) and aquatic ecosystems (Bradley *et al.*, 1984; Zheleznyak and Voytsekhovich, 1990). Using these models predictive assessments can be made of the behaviour and, subsequently, the dose arising from the deposition of radionuclides to the environment. Most of these models work with either acute or chronic discharges of radionuclides, that is deposition which occurs following the release of a pulse of radioactivity

to the environment usually as a consequence of an accident or from the low level emissions from routine operations at nuclear establishments.

This type of environmental monitoring and research is in line with the recommendations of the International Commission on Radiological Protection (ICRP), whose principal objective is to ensure that any activities involving human exposure are accompanied by a minimum of risk, and that they are of benefit to society. However, few studies have been concerned with the long term effects on the native flora and fauna of the low level release of radionuclides from nuclear establishments during routine operations. This is mainly due to the belief that '*if man is adequately protected* [from the effects of the radiation] *then other living things are also likely to be sufficiently protected*' (ICRP, 1977). At the time the ICRP made its recommendations, this assumption was based on exposure data for a limited range of wildlife, primarily marine species which are highly fertile and have short life cycles. Consequently, such species are least likely to exhibit any detrimental effects due to radiation exposure (Cassidy and Green, 1993; Thompson, 1988). Furthermore, several studies have shown that the ICRP's assumption is not necessarily true; for example, bioaccumulation and transfer of radionuclides to higher trophic levels have been recorded (Brisbin *et al.*, 1974; Ekman, 1967; French, 1967; Liden and Gustafsson, 1967; Lindner *et al.*, 1992; Pendleton *et al.*, 1964, 1965; Rudge, 1989). Moreover, recent awareness of the possible adverse effects of low levels of radionuclides, in conjunction with the fears and opinions voiced by inhabitants of countries affected by the Chernobyl accident, has led to a backlash against the operations of the nuclear industry in general (Cassidy and Green, 1993; Greenpeace, 1993). This has increased the need to understand the transfer and behaviour of radionuclides within semi-natural habitats. For example, one recommendation made by Cassidy and Green (1993) to improve the knowledge of the behaviour and effects of radionuclides released to the environment is to expand the official monitoring programme (enacted through by the Ministry of Agriculture, Food and Fisheries (MAFF)) to include measurements of the effects of low levels of radiation on wildlife.

1.1 OBJECTIVES

In outline, this project was designed and implemented to investigate the behaviour and transfer of radionuclides released to semi-natural environments, primarily from the ongoing operations at the British Nuclear Fuels Limited (BNFL) nuclear reprocessing plant located at Sellafield, Cumbria. In addition, deposition from the Chernobyl accident, weapons testing fallout, and the accident at Sellafield, then Windscale, in 1957 has been considered (section 1.3). The project provided a unique opportunity to study the enhanced level of radionuclide deposition within three ecosystems, and to assess and determine the availability of radionuclides for uptake and transfer through food chains using small mammals (common

shrew, *Sorex araneus*; field mouse, *Apodemus sylvaticus*; and field vole, *Microtus agrestis*) as bio-monitors.

It was postulated that radionuclide deposition to the three selected field sites is influenced by different mechanisms: aerial/canopy capture, tidal inundation and sea to land transfer, and that this would be expressed as noticeable differences in radionuclide activity and behaviour. The deposition mechanisms are described in detail in the relevant chapters (sections 3.1, 4.1 and 5.1). In addition to differences in the deposition mechanisms, the field sites differ in their ecology, being: a coniferous woodland, a salt marsh and a sand dune system. These are described in more detail in section 1.2 and within the appropriate chapter (sections 3.2, 4.2 and 5.2).

The study followed on from previous food chain work based at the BNFL low level radioactive waste disposal site at Drigg, Cumbria (Rudge, 1989), which assessed the effects and transfer of Chernobyl-derived radiocaesium (^{134}Cs and ^{137}Cs) in small mammal food chains. In addition to investigating the behaviour of radiocaesium, the present study also focused on the transfer of the actinides, plutonium and americium (isotopes: ^{238}Pu, $^{239+240}Pu$ and ^{241}Am). These radionuclides have received little attention as regards natural food chain systems but it is well known that internally deposited actinides are important in determining the dose received by humans due to the high linear energy transfer characteristic of alpha radiation and the consequent high biological effectiveness (National Radiological Protection Board (NRPB), 1989). The presence of these radionuclides internally within the mammalian body can lead to considerable tissue damage and may also contribute to the radiation burden of small mammals.

An assessment of the radionuclide distribution between the various food chain components (soil; vegetation; leaf litter; invertebrates and small mammals) at each site was made during this study. These data then formed the basis of that part of the project in which transfer factors to the higher trophic levels were determined. The primary objectives of this project may be outlined as follows:

1. To study the transfer of isotopes of the radionuclides: caesium; plutonium and americium, (^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am) through food chains in semi-natural habitats.

2. To determine concentration factors between the different food chain components.

3. To determine any differences between radionuclide transfer and bioavailability across three different ecosystems.

4. To make an indicative assessment of the radiation doses received by small mammal species inhabiting ecosystems contaminated with radioactivity.

To achieve these objectives, various experimental strategies were employed and these may be summarised as follows:

1. Quantification of the activity of ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in soils, vegetation, leaf litter, invertebrate and small mammal samples collected from the field sites during the course of a two year programme. ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am are representative of those radionuclides considered to be the most radiologically important.

2. Assessment of the radioactivity accumulated in soil as a consequence of past deposition across each field site. This was intended to provide information on the spatial distribution of the radionuclide deposition and form a baseline against which transfer factors could be calculated.

3. Limited assessment of the vertical migration (or sedimentation history for the salt marsh site) of radionuclides, using sectioned soil cores. This was also thought to be important in determining the concentration of each radionuclide in the rooting zones of the vegetation species located on each site.

4. Characterisation of a few of the major factors controlling the availability of radionuclides in soil and sediment (for example, organic matter content, particle size distribution and pH).

5. Assessment of the temporal and spatial variation within each sample type with associated explanations and/or discussion of the mechanisms involved.

6. Calculation of transfer factors to determine the retention and accumulation of radionuclides through different food chain components discussed in section 1.4.

7. Characterisation of the behaviour and transfer of radionuclides across the three field sites, including an assessment to determine if any differences were related to the ecology of a site or to the deposition mechanism.

8. A comparison of data obtained from the Cumbrian field sites with that from background reference sites to assess the effects of elevated environmental radioactivity on those species representing higher trophic levels.

9. Identification of the radioactivity levels and inventory across the three sites as a first stage in determining the dose received by species representing higher trophic levels, particularly small mammals. It was considered that detailed knowledge of the study sites would allow the relative contributions of radionuclides to the internal and external dose to be estimated in later work.

1.2 SITE LOCATIONS

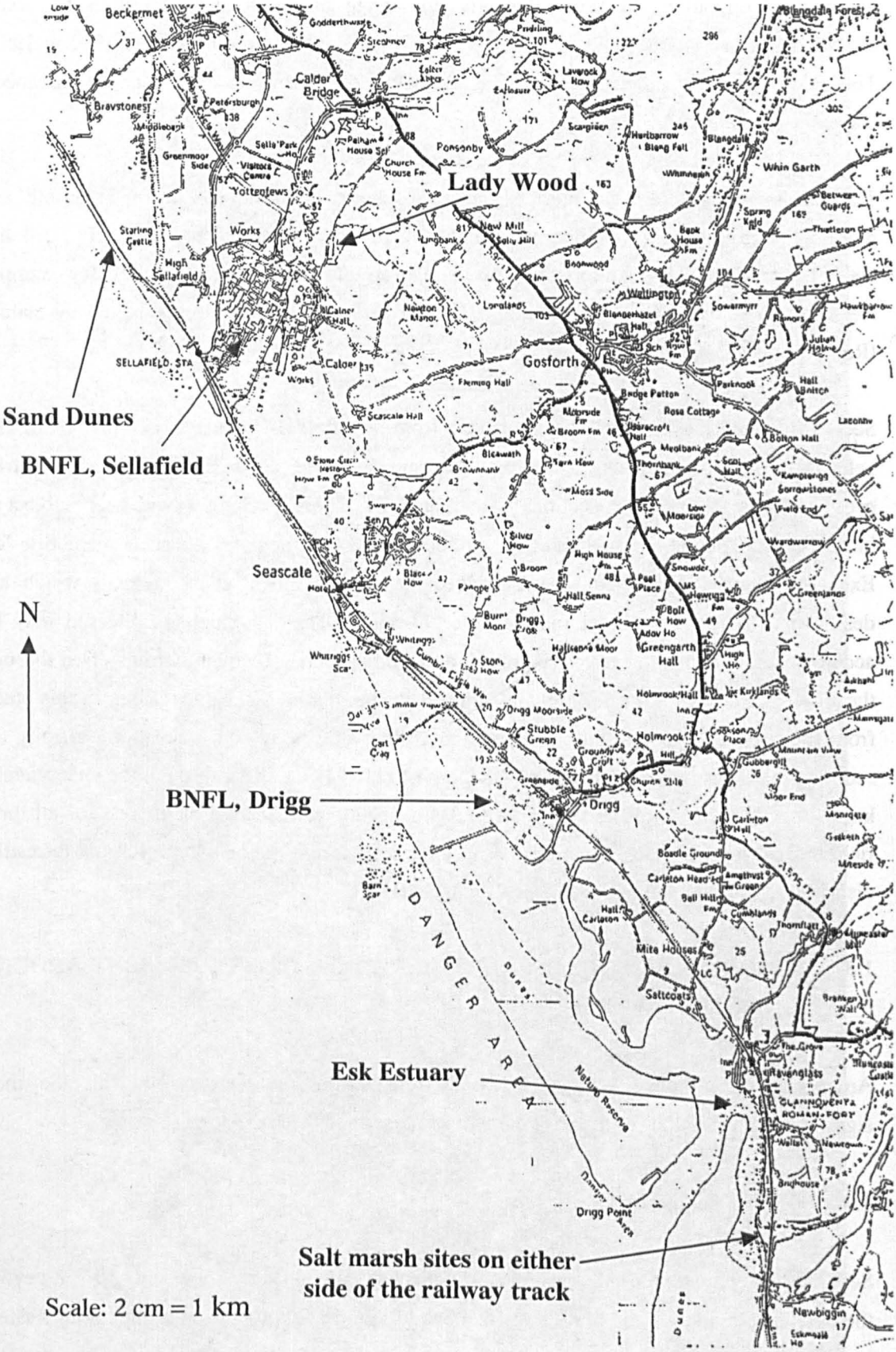
Details of the field sites are provided in Table 1.1. A full site description is given in the relevant chapter (sections 3.2, 4.2 and 5.2). The Cumbrian field sites are all located within 15 km of the BNFL nuclear reprocessing plant at Sellafield, on the north west coast of Cumbria, England (Ordnance Survey Grid Reference (OSGR): NY 025038). Figure 1.1 outlines the position of the field sites relative to the Sellafield site. Plate 1.1 shows how close two of the sites are through an aerial view of the Sellafield site. Lady Wood can be seen in the middle of the plate behind the Sellafield complex and the sand dunes are positioned along the coastline between the left hand side of the photograph and the Sellafield discharge pipeline. Plate 1.2 is a photograph of the Esk estuary which shows a railway viaduct dissecting the salt marsh which was used in the present study. Details of the reference sites used are provided in the relevant chapter (sections 3.2, 4.2 and 5.2).

Table 1.1: Field site details.

	Coniferous Wood	*Salt Marsh*	*Sand Dune*
Site name	Lady Wood	River Esk	Sellafield Dunes
Ordnance Survey Grid Reference	NY 037045	SD 089948	NY 016037
Distance from Sellafield	500 m	<15 km	<500 m
≈ Area	4.7 ha	9.2 ha	0.06 ha
Dominant vegetation in sampling area	Sitka spruce, *Picea sitchensis*	Soft brome, *Bromus mollis* and red fescue, *Festuca rubra*	Marram grass, *Ammophila arenaria* and red fescue, *Festuca rubra*
Deposition mechanism	Aerial/canopy capture	Tidal inundation	Sea to land transfer
Chapter	3	4	5

The Sellafield complex is owned and operated by BNFL and has been reprocessing irradiated nuclear fuel since planning permission was granted in 1952. In addition to the reprocessing division, the complex also contains the world's first industrial-sized nuclear power plant, Calder Hall. This power station presently consists of four small Magnox-type nuclear reactors.

Figure 1.1: **Map of the Cumbrian coastline showing the three field site locations and their proximity to BNFL, Sellafield. Reproduced from the Ordnance Surveys 1:50,000 map of 1987 with the permission of the controller of Her Majesty's Stationary Office. Licence agreement number: ED 275514.**



The mainstay of the reprocessing operations at Sellafield is the separation and extraction of uranium and plutonium from irradiated nuclear fuel from Magnox-type reactors operated by Nuclear Electric in the UK and a few overseas utilities. The current reprocessing plant commenced operations in 1964. Recently, the range of reprocessing facilities has been extended by the commissioning of the Thermal Oxide Reprocessing Plant (THORP) in 1994. This new plant will reprocess the irradiated nuclear fuel from the more modern Advanced Gas-cooled reactors (AGR's) and Light Water reactors (LWR's).

The discharges to both the marine and atmospheric environments from Sellafield are monitored through an environmental monitoring programme conducted by BNFL. All the discharges are subject to authorisation by the appropriate UK regulatory bodies, for example Her Majesty's Inspectorate of Pollution (HMIP) and MAFF, and are controlled by statute (Radioactive Substances Act, 1993).

Section 1.3 describes in detail the discharges from Sellafield, both current and historical. The behaviour of radionuclides in the environment and the deposition back to terrestrial ecosystems are described in sections 3.1, 4.1 and 5.1. These discharges have been subject to increasingly stringent controls and there are new waste treatments plants such as the Site Ion Exchange Plant (SIXEP) and the Enhanced Actinide Removal Plant (EARP) which are designed to reduce the liquid radionuclide emissions. The radionuclides released may be accumulated by aquatic or terrestrial plants and animals and subsequently transferred through the trophic levels within a food chain. Interest in the marine discharges arises in this study from the behaviour of the radionuclides within the water body and their strong affinity for sediment particles through adsorption (section 4.1 and 5.1), and also through the phenomenon known as sea to land transfer (Chapter 5). Atmospheric releases are of interest for all three field sites, because deposition from aerial emissions is the major source for the terrestrial distribution of radionuclides in the locality of Sellafield.

1.3 SIGNIFICANT SOURCES OF THE RADIOACTIVITY PRESENT AROUND BNFL, SELLAFIELD

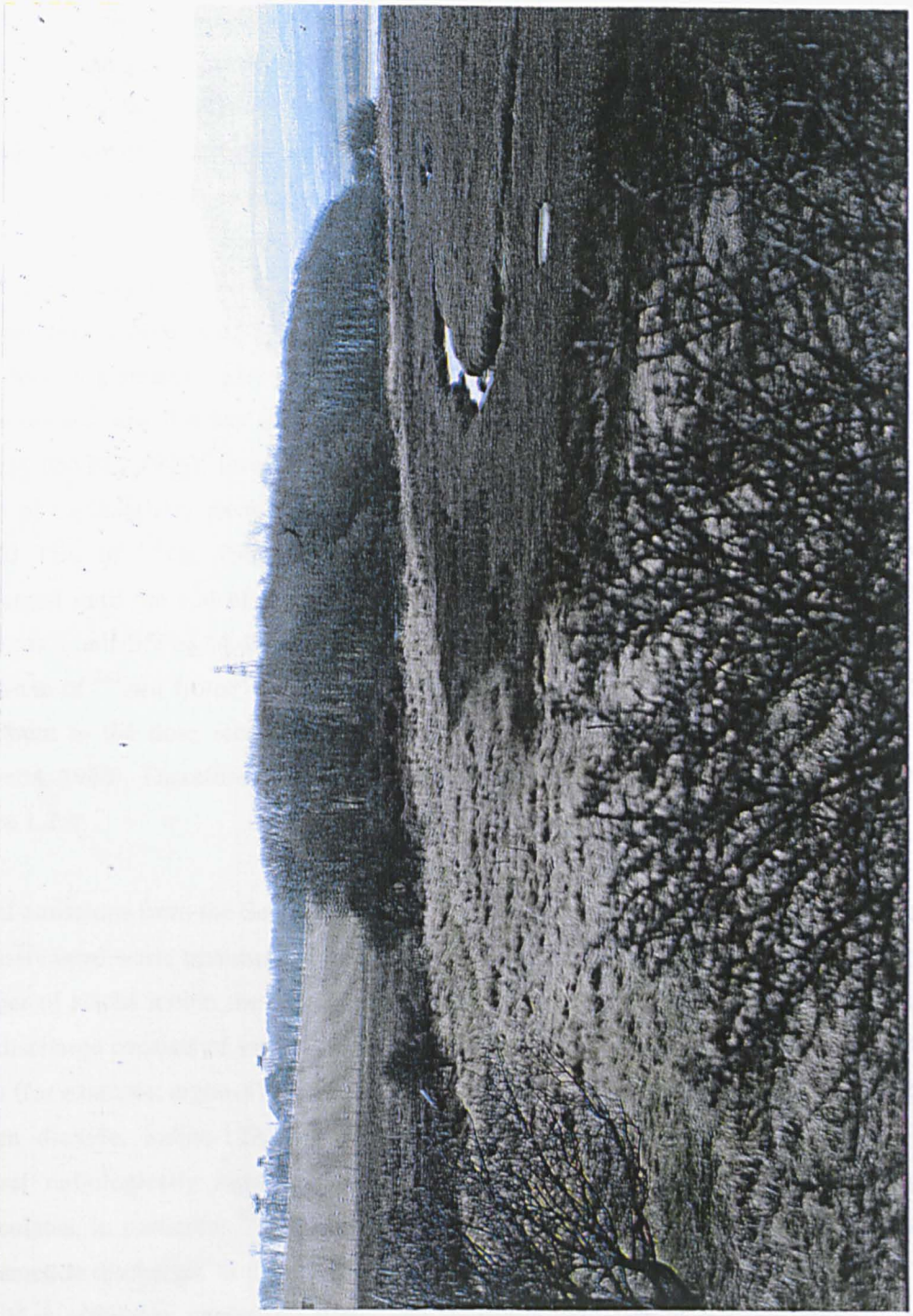
Anthropogenic radionuclides are released from several sources; those with the most significant contributions to the environment are described briefly below.

1.3.1 BNFL, Sellafield, Cumbria

Sellafield discharges into the marine environment via pipelines which extend 2.1 km beyond the low water mark into the north east Irish Sea. A significant proportion of the liquid waste is generated from the Magnox fuel element storage ponds mentioned previously and the

Plate 1.1: Aerial photograph of the BNFL, Sellafield complex showing both the Lady Wood and Sand Dune field sites (see text).

Plate 1.2: **Photograph of the River Esk field site (riverward side of the marsh) taken from the railway viaduct.**



reprocessing plant itself (Appleby and Luttrell, 1993). There was a peak in the releases to the marine environment in the mid 1970s and since then levels have been declining (BNFL, 1971-1995; Gray *et al.*, 1995; Howorth and Eggleton, 1988). ^{137}Cs releases peaked in 1975 with an activity of over 5,000 TBq per year being disposed of into the marine environment. ^{241}Am levels peaked at 120 TBq per year in 1974 and have been declining steadily. The introduction of SIXEP in 1985 has led to a considerable further reduction. It is expected that only a small increase in the emission of radiologically significant radionuclides will result from the commissioning of the THORP plant, mainly due to the stringent operating guidelines and the commencement of EARP operations. Figures 1.2 and 1.3 show the change in marine discharge levels for the radionuclides of interest.

At the present time, releases into the marine environment for ^{137}Cs are in the order of 13.5 TBq per year, 18% of the authorised operational discharge limit for the site. The levels have been declining steadily after peaking at the 5,000 TBq per year described earlier. Current ^{241}Am releases are also low at 30% of the authorised discharge limit, with 0.38 TBq released in 1994 (BNFL, 1995). Over the operational life of the Sellafield complex very significant levels of radioactivity have been released to the marine environment. Totals in excess of 40,100 TBq of ^{137}Cs, 750 TBq of plutonium α-activity and 525 TBq of ^{241}Am were discharged until the end of 1994. In addition, over 21,200 TBq of ^{241}Pu has been released. ^{241}Pu has a half-life of 14.4 years and decays into ^{241}Am. Therefore, there will be significant in-growth of ^{241}Am from ^{241}Pu in the future. It has been recognised that this in-growth will contribute to the dose received from ^{241}Am around the Sellafield complex (Howorth and Eggleton, 1988). Therefore the historical discharge of ^{241}Pu to the Irish sea is also shown in Figure 1.2.

Aerial emissions from the Sellafield plant arise from the operations of the reprocessing plants and associated waste treatment and storage facilities, and the Calder Hall reactors. There are a number of stacks within the Sellafield complex which emit radionuclides to the atmosphere. The discharge consists of ventilation air contaminated with radioactivity in the form of noble gases (for example, argon-41 and krypton-85), other gases (for example, tritium, carbon-14 as carbon dioxide, iodine-129, and iodine-131) and suspended particulates (BNFL, 1995). Several radiologically significant radionuclides can be adsorbed on to these suspended particulates, in particular ^{137}Cs, plutonium-α, ^{241}Pu and ^{241}Am. Again, efforts to reduce the radionuclide discharges to the atmosphere have been undertaken and the stacks are now fitted with high efficiency particulate filters and chemical scrubbers to remove radioactive gases. Aerial emissions from the complex peaked slightly later than those to the marine environment, in around 1980 (Figures 1.4 and 1.5). Gray *et al.* (1995) report revised aerial discharge estimates for the period 1952 to 1992 and these have been used with the more recent data from BNFL (1993 to 1995) to produce Figures 1.4 and 1.5. For the radionuclides of interest,

Figure 1.2: The historic marine discharges of ^{134}Cs, ^{137}Cs and ^{241}Pu from BNFL Sellafield, Cumbria.

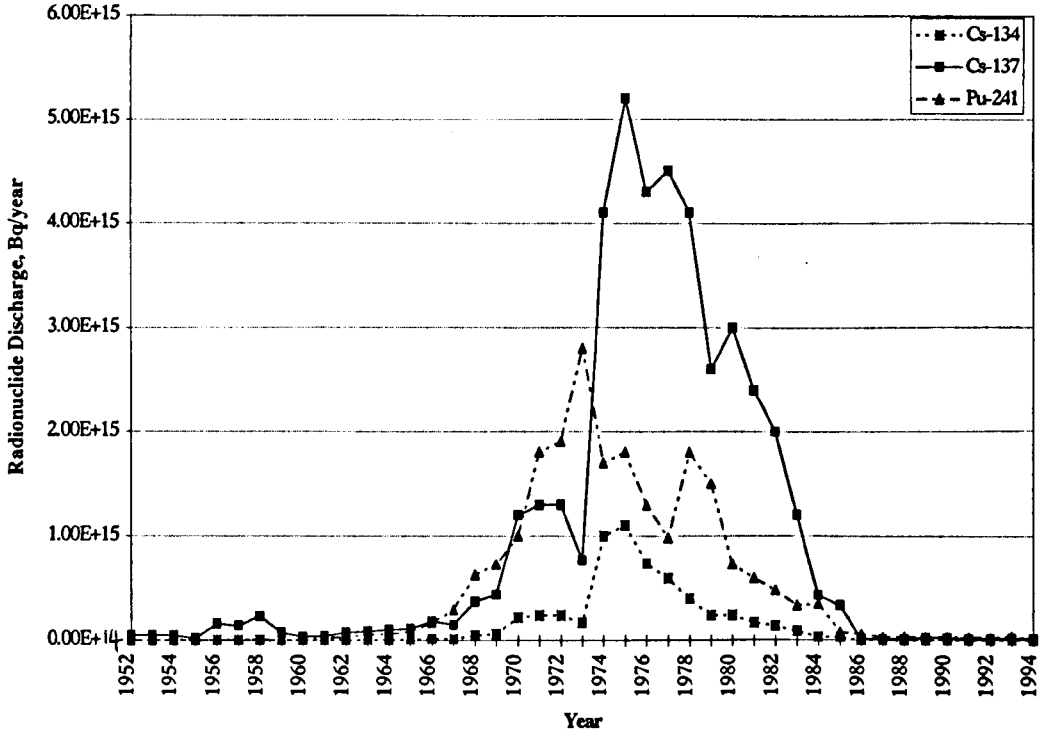


Figure 1.3: The historic marine discharges of the actinides, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am from BNFL, Sellafield, Cumbria.

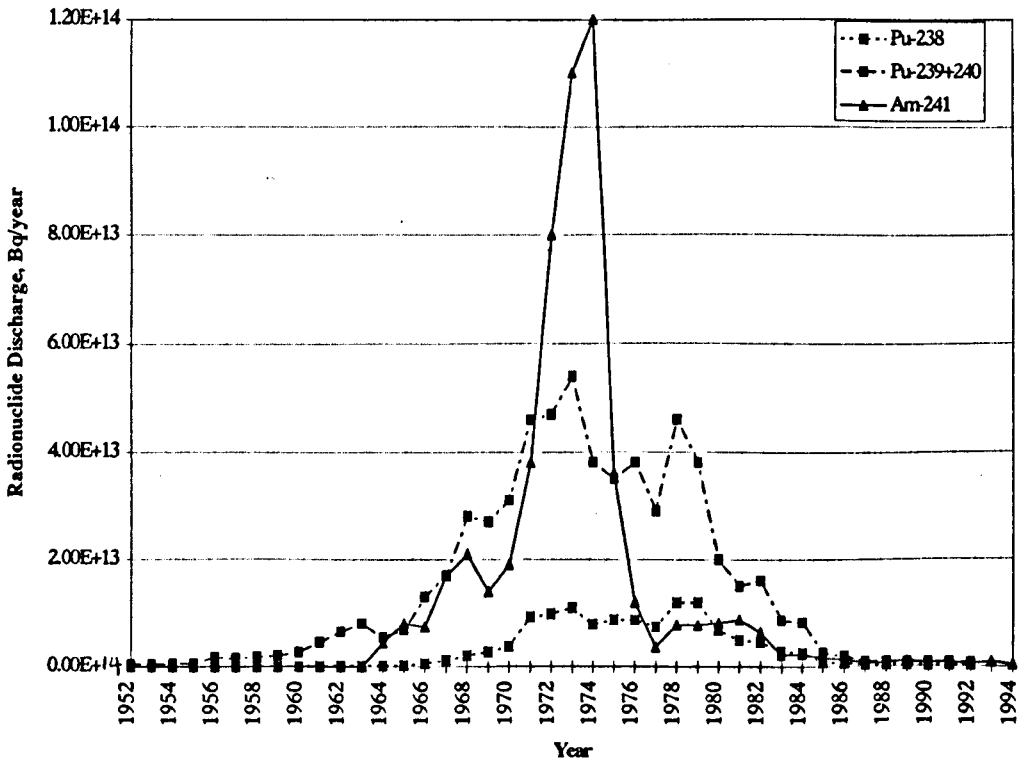


Figure 1.4: **Historic aerial discharges from the high stacks at BNFL, Sellafield, Cumbria, emission height equivalent to 80 m.**

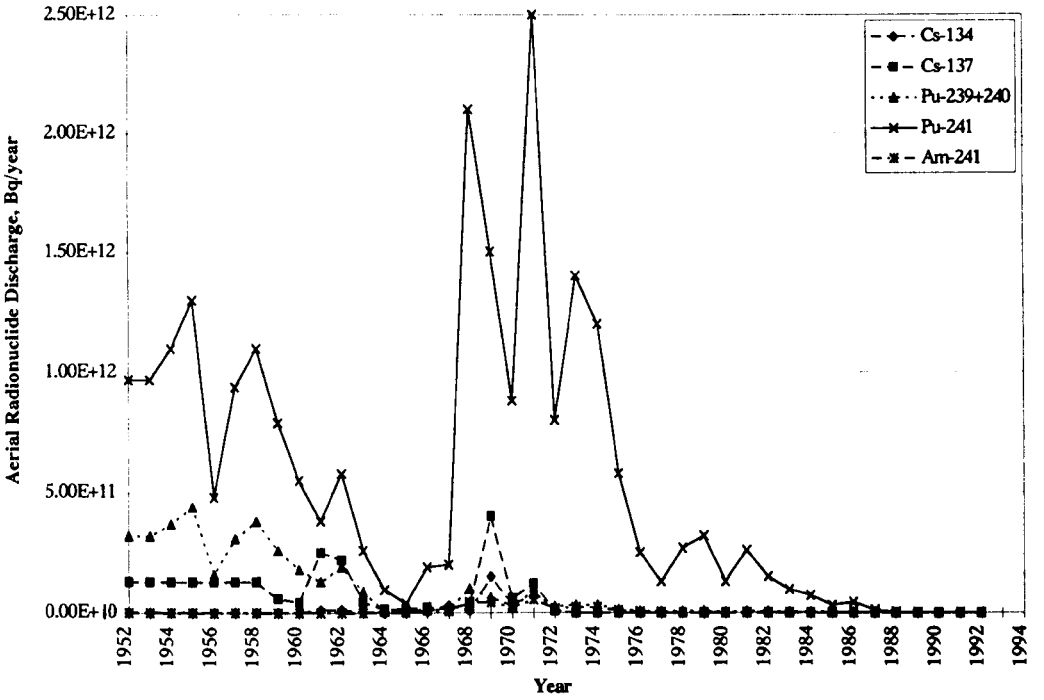
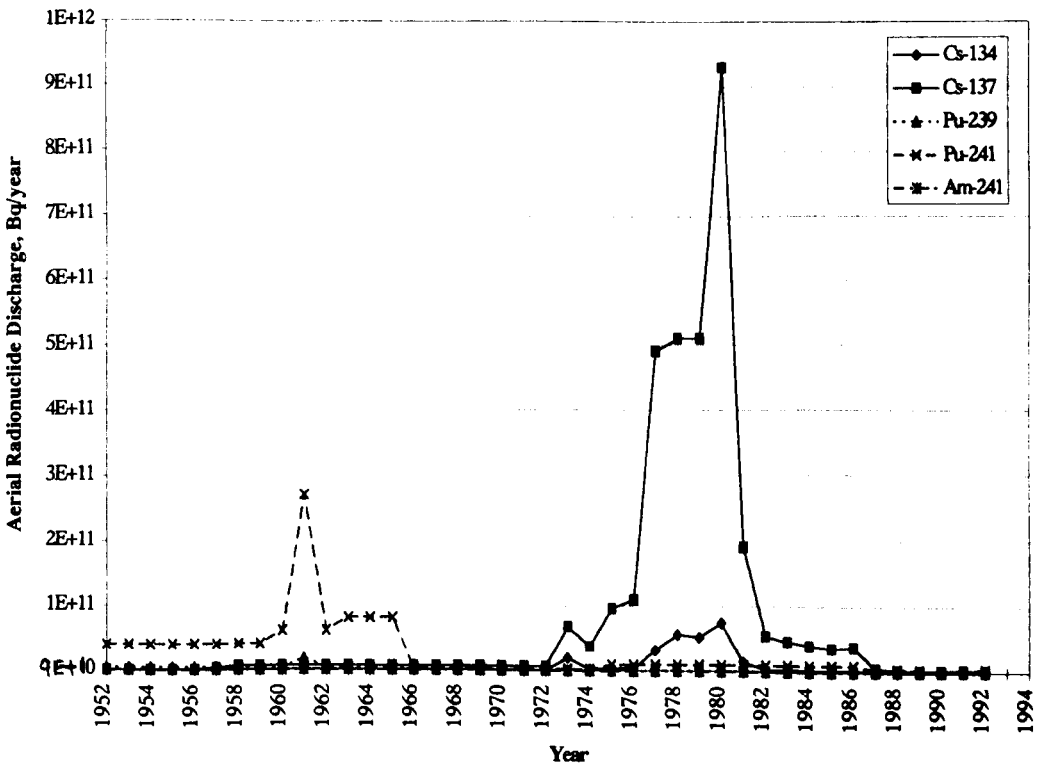


Figure 1.5: **Historic aerial discharges from the low stacks at BNFL, Sellafield, Cumbria, emission height equivalent to 10 m.**



0.68 TBq of ^{137}Cs were released from the stacks in 1994, although the dispersal of the ^{137}Cs is dependent upon which stacks are involved. This represented <4% of the discharge limit. Of the actinides, 0.11 TBq of plutonium-α and 0.97 TBq of ^{241}Pu and ^{241}Am were released in 1994 (BNFL, 1995). Confirmation of the aerial discharge chronology has been obtained from the analysis of lake core sediments from Ponsonby Tarn (OSGR: NY 046045), located on a transect from Sellafield which passes over the Lady Wood site (Bonnett and Cambray, 1991; Eakins and Cambray, 1985). The plutonium data from the cores havebeen compared to model predictions of the aerial discharges and provide a reasonable account of these releases (Gray *et al.*, 1995).

Both the marine and aerial discharges are released on the principle of 'dilute and disperse', a practice commonly used to reduce levels of pollutants within the environment. The practice began at Sellafield when the site was first converted to radioactive purposes and at that time, during the late 1940s and early 1950s, disposal of waste products to the marine environment was commonplace and believed to be a highly safe and effective way of diluting and dispersing pollutants. At the time, the environmental behaviour of radioactive substances was not understood, although it was recognised that any discharges would result in an increase in the dose received by members of the public (Dunster, 1958). Therefore, before any radioactive waste was discharged, several experiments were undertaken at the Sellafield complex to investigate the likely fate of radionuclides in the marine environment (Dunster, 1955; 1958; Seligman, 1955). These recognised some of the critical pathways resulting in human exposure but failed to investigate the effects on wildlife. Furthermore, although the discharge levels have declined significantly, there is still considerable environmental contamination to which wildlife is continually exposed. Therefore Sellafield, not surprisingly, forms the most important source of radionuclides for the locality.

1.3.2 Windscale Accident

The first substantial publicised release of radioactivity to the environment occurred in October 1957 at Sellafield, then known as Windscale, under the control of the UK Atomic Energy Authority. The accident was caused by overheating during a procedure within the Windscale No. 1 reactor designed for the production of military grade plutonium. The overheating caused a containment failure, the oxidation of the uranium fuel and subsequently a graphite fire but no explosion (Chamberlain, 1990). Initial attempts to douse the fire using carbon dioxide failed and so water was applied until the reactor core was cold. This accident resulted in the release of uncontrolled radioactivity to the environment, mainly of gaseous or volatile elements, over a period of 24 hours. This led to the estimated release of 2.2×10^{13} Bq of ^{137}Cs and 1.6×10^{9} Bq of $^{239+240}Pu$ of the radionuclides of interest to the present study. Other principal radionuclides released from the reactor include: ^{131}I, polonium (^{210}Po), ruthenium

(^{106}Ru), strontium (^{90}Sr), technetium (^{132}Te) and xenon (^{133}Xe) (Appleby and Luttrell, 1993; Crick and Linsley, 1982). The release of ^{131}I actually masked the presence of several of the other volatile fission products using the gamma spectroscopy systems available at the time of the accident (Booker, 1958).

The accident-released radioactivity entering the environment was in a predominantly gaseous form, the plume of which was monitored passing over the UK and was detected in Belgium, Germany and Norway (Stewart and Crooks, 1958). Stewart and Crooks (1958) also indicated that the radioactivity underwent dry deposition by turbulent diffusion and impaction because little rainfall fell as the plume passed over the UK. Ground surveys within the vicinity of the Windscale complex shortly after the accident revealed that the deposition of particulate material extended 4 km south to south-east from the site. This is of particular relevance to the present study since both Lady Wood and the River Esk lie in this direction and it can be assumed that significant deposition occurred, certainly within the woodland.

1.3.3 Emissions of particulates from the Windscale Piles

Uranium oxide particles were emitted from the Windscale stacks during routine operations prior to the accident in the years 1954 to 1957 (Jones *et al.*, 1996; Stather *et al.*, 1986). These particles were comparatively large, being between 10 and 100 μm in size, and were deposited for the most part within a few kilometres of the release point. Fortunately, the large size of the particles and their deposition velocities under the conditions at the time of release, mean that little activity would have remained on the edible herbage and therefore limited food chain transfer is expected to have occurred (Chamberlain, 1987). Original estimates suggested 12 kg of uranium oxide were released but this has been revised to 20 kg (Chamberlain, 1987; Jones *et al.*, 1996; Stather *et al.*, 1986). This later estimate accounts for the measured activity of ^{90}Sr and ^{137}Cs in soil cores sampled between 1957-1965 (Chamberlain, 1987; Jones *et al.*, 1996). These emissions are important sources of ^{90}Sr and ^{137}Cs deposition to the Lady Wood site because of its proximity to the Pile chimneys.

1.3.4 Chernobyl Accident

Detailed information on the Chernobyl accident and its aftermath is presented elsewhere (Appleby and Luttrell, 1993; Cambray *et al.*, 1987; Fry, 1987; Rudge, 1989; Sandalls *et al.*, 1993; Savchenko, 1995). The key features for the present study are the actual radionuclides released and the deposition of these across the UK, in particular in the Cumbrian region. Much of the material released from the reactor consisted of irradiated fuel enriched with noble gases and various volatile radionuclides such as isotopes of caesium, iodine and tellurium. The subsequent dispersal of the radioactivity depended in part on the nature of the

radionuclides released and in part on the climatic conditions at the time of the accident. Initially it was estimated that about 3% of the transuranic elements present within the reactor core were released during the accident and these were emitted mainly as large, heavy particles which were readily deposited back to ground within a 30 km radius of the Chernobyl plant, primarily as a consequence of gravitational settling (Appleby and Luttrell, 1993; Hohenemser *et al.*, 1986; Sandalls *et al.*, 1993). Kulakov *et al.* (1990) reported a corrected estimate of 3.5 ± 0.5% and, from this, they calculated that approximately 23 kg of plutonium was released to the environment. The majority of the volatile ^{134}Cs, ^{137}Cs and ^{131}I were dispersed considerable distances by the ambient meteorological conditions. Measurements at the time of the accident showed that these radionuclides in particular were associated with particles of diameter <3 μm and that for ^{137}Cs particles <1 μm dominated (Persson *et al.*, 1987). An estimated 850,000 TBq of ^{137}Cs were released from the reactor core (ApSimon *et al.*, 1988). Local heating caused by the explosion is suggested to have given rise to the formation of convection currents which caused these particles and gases to rise. ApSimon and Wilson (1986) report that an inversion at 3,000 m would have confined the radioactivity in the troposphere. Consequently, the movement of the plume was determined by surface winds and prevented the Chernobyl debris from being spread globally. The air movements at the time of the accident have been described elsewhere (Albergel *et al.*, 1988; ApSimon and Wilson, 1986; Persson *et al.*, 1987; Wheeler, 1988).

The importance of the particle size data is firstly that the caesium isotopes and other radionuclides bound to small particles travelled considerable distances in the atmosphere before being deposited, primarily as a result of particulate scavenging (wet deposition). Also, the particle size was comparable to those studies investigating weapons testing fallout (section 1.3.4). Secondly, the particle size affects the foliar uptake and retention time of radionuclides deposited to the surface of vegetation. The patchy nature of the deposition across the northern hemisphere is also partly due to the particle size because strong correlations between local deposition and heavy rainfall were obtained, suggesting that particulate scavenging was the principal method of deposition (ApSimon *et al.*, 1988; Fry, 1987; Hohenemser and Renn, 1988; Wheeler, 1987). As shown in Chapter 3, this is important when assessing the spatial distribution across a small study site because an even deposition of the Chernobyl-derived radionuclides is expected as a result of wet deposition.

The radioactive plume from Chernobyl passed over the UK on 2nd May 1986 and coincided with a period of heavy rainfall. The results for total caesium deposition across the country clearly indicate a patchy distribution correlated with areas of high rainfall (Cambray *et al.*, 1987; Clark and Smith, 1988). The worst affected areas include: Cumbria, Gwynedd, areas of Yorkshire and areas of south-east Scotland (Fry, 1987). Despite the Chernobyl accident's obvious damage to human, environmental and economic concerns, the accident provided a

unique opportunity to study the consequences of a large-scale nuclear accident, and to examine the subsequent transfer of radionuclides through ecosystems. It should be noted that the total activity released from the accident was considerably less than the combined contributions from the weapons testing of the 1950s and 1960s (section 1.3.4); but there were important differences in the deposition with the fallout from Chernobyl being patchy in nature depending upon the local climatic conditions, whereas weapons testing fallout was dispersed globally. Subsequently, these differences have affected the mechanisms determining the transfer of the radionuclides into terrestrial and aquatic ecosystems.

Estimates of the caesium deposition in Cumbria, especially around the Sellafield complex, show that between 30 and 50% of the ^{137}Cs present in soil cores to a depth of 15 cm is attributable to the Chernobyl accident (Cambray *et al.*, 1987; Cawse and Colle, 1988; Rudge, 1989) but varies proportionally with the amount of rainfall received. This is reflected by reported deposition values for ^{137}Cs which range from 7.4 kBq m^{-2} around the Drigg headland (Fulker, 1987) to 48 kBq m^{-2} recorded by Jackson *et al.* (1987) for an upland area a few kilometres inland from Drigg. Typical deposition values in the locality of Sellafield were however closer to 10 kBq m^{-2} (Cambray *et al.*, 1987; Clark and Smith, 1988). Only very limited deposition of plutonium and americium occurred as a consequence of the Chernobyl accident especially when compared to the levels of actinide discharge from the Sellafield complex itself and weapons testing fallout (section 1.3.4).

1.3.5 Weapons Testing Fallout

Weapons testing began in the early 1950s and peaked in 1961/1962, resulting in the highest deposition of emitted radionuclides in 1963. Since the moratorium on weapons testing, the annual deposition across the UK has been declining steadily (Cambray *et al.*, 1989). The main feature of the deposition of weapons-testing debris is that it occurs on a global scale. This is a function of the material being injected into the stratosphere where global transport and mixing can occur, and where the associated residence time is estimated at one year. In contrast, for debris from the Chernobyl accident released to the troposphere, the residence time was estimated between 20 and 40 days (Aarkrog, 1988; Kownacka and Jaworowki, 1994). Total ^{137}Cs emission to the environment following the testing has been estimated as 910,000 TBq (Cambray *et al.*, 1989) and for $^{239+240}Pu$ 13,300 TBq (Perkins and Thomas, 1980). Hardy *et al.* (1973) reported that the total $^{239+240}Pu$ deposit by 1970 was in the order of 12,000 TBq, and for ^{238}Pu, 285 TBq, although the deposition pattern around the globe is irregular (Perkins and Thomas, 1980). Consequently, whilst the Chernobyl and Sellafield emissions are clearly important components in the levels of radionuclides around Sellafield, both are at least an order of magnitude lower than the radiation released through the early weapons testing.

In the immediate vicinity of the Sellafield complex the atmospheric discharges of plutonium dominate the inventory and are reported to be in excess of 200 times those expected for the area from weapons testing fallout alone (Howorth and Eggleton, 1988). The global fallout from weapons testing will be strongly influenced by rainfall as was the Chernobyl fallout, therefore areas of high rainfall like Cumbria will be subject to an enhanced deposition of this radioactivity. Estimates of between 3,000 and 4,000 Bq m^{-2} for ^{137}Cs deposition in the locality of Sellafield as a result of weapons testing have been reported (Cambray *et al.*, 1987; Howorth and Eggleton, 1988). The deposition of $^{239+240}Pu$ from weapons testing has been estimated as between 70 and 80 Bq m^{-2} at a number of locations along the Cumbrian coastline (Howorth and Eggleton, 1988).

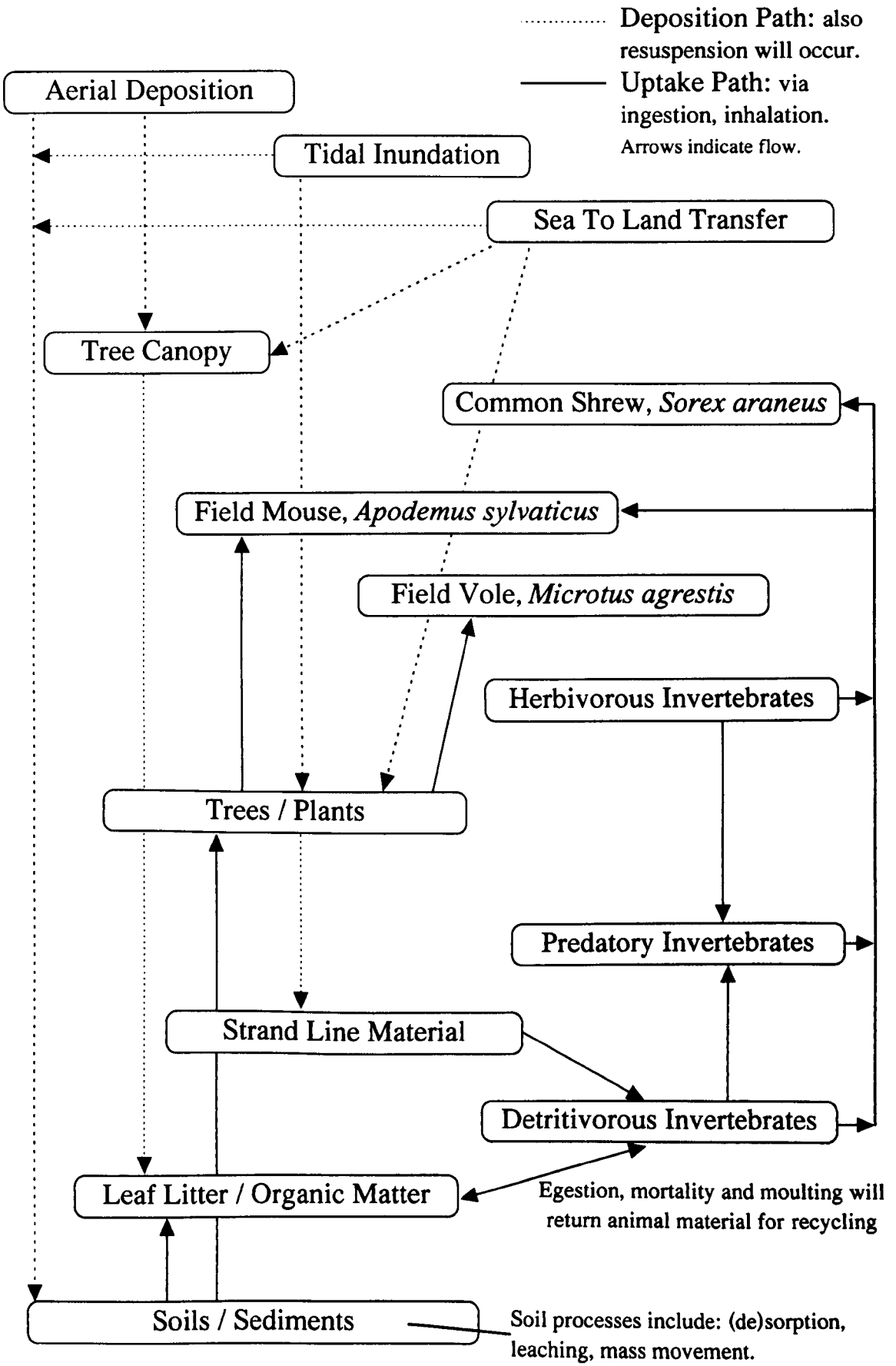
1.3.6 Miscellaneous

Several smaller accidents have contributed to the environmental radioactivity measured around Sellafield. Two incidents at Sellafield occurred in 1979 and 1984. In the former, about 11 GBq of plutonium-α were released from the effluent treatment plant (BNFL, 1980; Gray *et al.*, 1995). In 1984, a release of 0.4 GBq of ^{241}Am occurred from a sludge storage tank (BNFL, 1985; Gray *et al.*, 1995). Added to these are releases relating to the complex as Windscale. There have been three releases of note: the early plutonium-finishing operations in the 1950s, the fuel storage ponds associated with the Windscale piles and the plutonium contamination in one of the Calder reactor cooling tower basins. These are estimated to have released in the order of 80 GBq (Jones, 1996; Howorth and Eggleton, 1988; Stather *et al.*, 1986).

1.4 FOOD CHAIN COMPONENTS

A résumé of the components of a representative food chain pertinent to the study area is presented below to reduce duplication in later chapters. More specific information is provided as appropriate in the later sections. Figure 1.6 presents a model, indicating the links between the major components of the food chains found on the three field sites. Both aerial and sea to land transfer deposition can occur across each site, although the importance of the individual mechanisms is determined by the location and features of each site. Deposition via tidal inundation only occurs on the salt marsh. Sections 3.1, 4.1 and 5.1 describe the deposition mechanism for each study site and discuss some of these factors in greater detail.

Figure 1.6: **Flow diagram of the major food chain components and their relationships in this study.**



1.4.1 Soils/Sediments

Artificial radionuclides enter terrestrial ecosystems from the atmosphere, either as direct aerial deposition or as a consequence of sea to land transfer (Chapter 5). These radionuclides may be deposited on to the surface of vegetation (section 1.4.2) or directly on to soil or sediment. Intertidal sediment, such as that found in salt marshes, also receives a radionuclide input via tidal inundation (Chapter 4). In both cases, the majority of the deposited radionuclide inventory is ultimately transferred to soil or sediment. Several authors have indicated the importance of soils and sediment as major repositories for radionuclides (Adriano *et al.*, 1981; Rickard *et al.*, 1982). The behaviour of radionuclides therein, and the rate of deposition of radionuclides, are dependent upon a number of factors; for example, particle size, chemical form, climatic conditions and retention of the deposited material on the surfaces of vegetation.

Consequently, soils form a convenient medium for studying and monitoring the spatial distribution of radioactivity deposited in the environment. Over time, areas of undisturbed soil integrate the radioactive material deposited, producing a record of past deposition specific to a particular area or site (Cambray *et al.*, 1987). Depending upon the deposition mechanisms involved, large fluctuations in the radionuclide deposition can occur across an area. Thus, by assessing the soil inventory, the accumulation and spatial distribution of radionuclides can be determined. This forms a site specific reference level from which to examine the behaviour and transfer of radionuclides through natural food chains.

Most of the information on the behaviour of radionuclides within soils (including many of the models which have been developed) refers to the initial deposition of radionuclides to the soil surface. Originally, these models used the global fallout from weapons testing (for example, Evans and Dekker, 1966; Graham, 1958; Menzel, 1965), and subsequently the accidents which have occurred at nuclear power stations. Alternatively, they have been based on the tracking and observation of tracer solutions added to soil or crop in the field (Milbourn *et al.*, 1959; Squire and Middleton, 1966; Waller and Olson, 1967) and laboratory (Evans and Dekker, 1966; Romney *et al.*, 1957; Tukey *et al.*, 1961). In all cases, the retention and mobility of the radionuclides have been related to the physico-chemical properties of the radionuclide, soil type and characteristics (for example, particle size, organic matter content, pH, water content) and the vegetation species present. For example, ^{90}Sr and ^{137}Cs deposited from weapons testing fallout consisted largely of water soluble and exchangeable forms and was therefore potentially for uptake by plants, via direct foliar absorption or root uptake from the soil (Kirchmann *et al.*, 1993) although the eventual sorption of these radionuclides to soil minerals would reduce bioavailability. In contrast, the same nuclides which were deposited within the 30 km exclusion zone around the Chernobyl power plant, were associated with fuel

particles. These particles are insoluble in water and therefore the radionuclides were biologically unavailable (Konoplev and Bobovnikova, 1990). However, measurements of the Chernobyl-derived ^{137}Cs in the United Kingdom during May 1986 showed that over 70% was in a water soluble form. This compared to measurements taken in 1959 and 1987 on weapons testing fallout and aerial emissions from Sellafield respectively, where only 8% and 50% of the ^{137}Cs was present in a water soluble form (Hilton *et al.*, 1992). However, the low solubility of the ^{137}Cs from the weapons fallout may be attributable to the sampling technique employed in 1957, and the storage period between collection and analysis (Hilton *et al.*, 1992). The results do nevertheless indicate that there are differences in the availability of ^{137}Cs from Chernobyl and weapons testing fallout as noted by other authors (Santschi *et al.*, 1988). It is significant that water solubility is often only temporary, as across many areas the ^{137}Cs fallout is rapidly combined with clays within the soil matrix by cation exchange and thus becomes biologically less available (Allen, 1984; Bocock, 1981).

Transuranic elements such as plutonium and americium exhibit a range of chemical forms because of the number of oxidation states in which they can exist. This is important when considering the mobility of plutonium within soil. For example, in its higher oxidation states, plutonium is soluble and therefore present within the soil solution. As discussed below, various physico-chemical characteristics of the soil may alter the oxidation state of transuranic elements, thus altering their biological availability (Bondietti and Tamura, 1980).

Several studies have investigated soil type and characteristics and their influence upon the availability for plant uptake of the more radiologically important radionuclides such as ^{137}Cs, $^{239+240}Pu$, ^{241}Am. From these studies the major influential factors in the soil are: sorption, resuspension, mass transport and leaching (Kirchmann *et al.*, 1993). These are described below.

1.4.1.1 Sorption/Leaching

Deposited radionuclides may bind irreversibly on to ion exchange sites on soil particles, they may exist in the soil solution or colloidal phase, or bind to organic matter. This controls availability for plant uptake (Morgan, 1990). Several methods for determining the degree of sorption to soil particles have been devised, commonly utilising a sequence of chemical solutions with each solution progressively removing radionuclides which are more tightly bound to soil particles (McLaren and Crawford, 1973; Tessier *et al.*, 1979). Typically, these remove, in order, radionuclides present within: the soil solution, water soluble/exchangeable, specifically absorbed, organically bound, oxide or hydroxide bound fractions and finally a residue which is irreversibly bound to the soil particles (McLaren and Crawford, 1973; Schmitt and Sticher, 1991; Tessier *et al.*, 1979). The radionuclide components bound to the

first four of these extracts are thought to be biologically available for plant uptake (Gupta and Aten, 1993; Schmitt and Sticher, 1991). However, Nirel *et al.* (1986) point out that there are limitations to the sequential extraction methods used to determine the chemical speciation of a radionuclide, particularly because of an apparent under-estimation of the activity present in the early phases of the extraction sequence. Furthermore, sample collection methods and subsequent storage can alter the redox conditions, producing analytical problems (Forstner, 1993). For the radionuclides in the present study, ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am, typically exhibit low concentrations within the more readily exchangeable groups and therefore exhibit low mobility in soils (Cook *et al.*, 1984; Morgan, 1990; Yamamoto *et al.*, 1980).

Leaching of radionuclides through the soil column is related to sorption, soil structure and rainfall. It is difficult to quantify losses via leaching because the range of parameters that need to be assessed is very large; but an understanding of the more important soil and radionuclide characteristics can help to determine the leaching rate. For example, there have been experimental investigations of the infiltration of radionuclides in soils under different rainfall conditions. Schimmack *et al.* (1994) studied migration in forest soils under 'heavy rain shower' and 'slight to moderate rainfall' conditions by altering the rate of irrigation. The results showed that the organic soil layer (0 to 4 cm) was effective at retaining the radionuclides, ^{60}Co and ^{137}Cs, under light rainfall. Under high rainfall conditions, only 70% of the ^{137}Cs and 90% of ^{60}Co was retained by the organic layer with the remainder infiltrating down to 14 cm. This indicates that during heavy rain, any current radionuclide deposition will penetrate further into the soil profile and that leaching of the upper soil layers will also be more effective. The more important soil parameters which affect the sorption of radionuclides and therefore determine the biological availability of them are described below and have been reviewed by Allen (1984); Bocock (1981); Cawse and Horrill (1986); Morgan (1990); Ross (1994); and Sposito (1989).

The presence of clay leads to a strong retention of ^{137}Cs within the soil matrix compared to calcareous soils (Squire and Middleton, 1966) and specific clay minerals such as illite are another important influence (Cawse, 1983; Cook *et al.*, 1984). Similarly, Livens and Baxter (1988) have shown that the specific activity of ^{137}Cs associated with clay particles may be as much as 40 times that of the sand fraction. Cook *et al.* (1984) reported that the movement of ^{137}Cs in clay soils was minimal, being determined largely by the physical illuviation movement of the clay particles themselves. Of course, the overall influence of the clay particles will be determined by the particle size distribution of the soil. For example in a sand dune ecosystem (Chapter 5), any clay present may well have a high specific activity, but the total activity will be determined by the mass dominance of the less active sand particles. A greater penetration of ^{137}Cs has also been reported for sandy soils (McHenry and Ritchie,

1977; Morgan, 1990). As with ^{137}Cs, isotopes of plutonium are associated with clay particles (Muller, 1978; Yamamoto *et al.*, 1980). This is a significant relationship. Muller and Sprugel (1977) found that 60 to 75% of the plutonium was bound to particles <4 μm, but this fraction represented only 25 to 35% of the soil by mass.

The adsorption of radionuclides on to soil particles is not determined solely by the particle size and soil composition. As indicated earlier (section 1.4) the chemical form of the radionuclide when it is deposited is also important. Subsequently, this may be modified by the soil pH and the effects of pH vary between different metal ions, some becoming more soluble, others more tightly adsorbed (Berrow and Burridge, 1991; Hughes *et al.*, 1980; Bergeijk *et al.*, 1992). Notably, Cawse and Horrill (1986) and Davis (1963) report no correlation for ^{137}Cs retention and pH under normal field conditions.

Effects of pH and other physico-chemical properties of soil are related to the time allowed for adsorption and will be modified by competing cations present within the soil solution (Evans, 1989). It is well known that caesium is physiologically analogous to potassium through their physical and chemical similarities, and therefore under conditions of high potassium, caesium uptake by plants will decrease and *vice versa* (Davis, 1963). Other radionuclides behave in a similar manner; for example, ^{90}Sr is analogous to calcium (Russell and Newbould, 1966). In addition, there are active uptake routes for many stable metals in plants, consequently, analogous radionuclides will be transferred via these routes as well.

Other metal ions in the soil solution will compete with radionuclides for soil binding sites (Sposito, 1989) and therefore affect the availability of radionuclides for plant uptake (Shaw and Bell, 1991). This relates to the soil's cation exchange capacity; a measure of the available binding sites which may be modified further by the presence of chelating agents (Essington *et al.*, 1962) or soil colloids (Graham and Killion, 1962; Jacobson and Overstreet, 1948). Both caesium and plutonium are absorbed at cation exchange sites but for plutonium little is held in an exchangeable form (Cook *et al.*, 1984; Muller, 1978; Whicker, 1983). In addition to the above, radionuclide fixation in soil occurs as complexes are formed with ligands of inorganic (for example, OH^-) and organic matter (Davydov *et al.*, 1990; Gerritse and Driel, 1984). Plutonium, in particular, associates with organic matter (Livens *et al.*, 1986) and this maybe a stable combination that actually reduces plutonium bio-availability (Cleveland and Rees, 1976; Cook *et al.*, 1984; Livens *et al.*, 1987). For ^{137}Cs however, organic matter may provide an additional source of radionuclides available for plant uptake, particularly in semi-natural habitats where there is a standing crop of dead material (Cawse and Turner, 1982; Bergeijk *et al.*, 1992). This is because plants satisfy most of their nutrient requirements via root uptake from surface organic horizons (Burmann *et al.*, 1994). Moreover, the radionuclides may be

incorporated directly into the food chain via the decomposer community (Adriano *et al.*, 1981; Ritchie *et al.*, 1970).

1.4.1.2 Resuspension

Radionuclides investigated within this study generally exhibit low mobility. Typically, this means that they remain bound in the upper soil layers and thus subject to resuspension. Resuspension is defined as *'the entrainment into the atmosphere of surface contamination that was originally airborne but deposited to the ground surface'* (Kocher, 1980; Sehmel, 1980). Following deposition, radionuclides usually become attached to soil particles and it is these that undergo resuspension. Resuspension is a complex process influenced by many factors such as wind speed, moisture, vegetation cover, season, mechanical disturbance and particle characteristics. Healy (1980), Linsley (1978) and Morgan (1990) provide more detailed reviews of the resuspension mechanism and its effects.

The resuspension of radionuclides associated with soil particles is a mechanism for their removal or re-distribution through the action of wind and rain; but the passage of animals can also disturb and resuspend soil particles (Sehmel, 1980). Resuspension can provide an important source of contamination for small mammals and other biota. It is suggested that this occurs mainly through inhalation which is particularly important when considering the dose received from the actinides, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am (Bondietti and Tamura, 1980; section 1.4.4.3).

1.4.1.3 Mass Transport

Mass transport is the movement of radionuclides through soil by physical or biological processes. Physical movement involves the downward migration of soil particles through macropores, a process known as illuviation. Biological transport is dominated by soil organisms and the burrowing activity of larger animals facilitates migration of radionuclides through the soil profile. Bishop (1989) reviewed this topic and reported that biotic transport of radionuclides through the actions of burrowing animals represents a significant pathway for the migration of radionuclides. A wide variety of animals is involved, for example: badgers, rabbits, moles, mice, voles, earthworms, and ants (Bishop, 1989). It is the construction of burrows, underground tunnels and chambers which result in the transport of radionuclides within the soil profile. Firstly, during construction, soil may be redistributed vertically or horizontally. In the former, soil is moved from depth to the surface; in the latter, mixing of soil will occur. This results in the redistribution of radionuclides within an area. Secondly, the volume of soil moved can be considerable; for example, earthworms can move in the order of 10 kg/m^2/year in established pasture, equivalent to a depth of 14 mm (Bishop, 1989; Evans,

1948). Therefore, for those radionuclides irreversibly bound to soil particles, mass transport, represents a significant method for radionuclide redistribution dependent upon the population size, burrow form and structure, burrow depth, soil type and the species involved.

Where burrows open on the surface, radionuclides present in rain and as gases can migrate to much greater depths than by direct surface deposition, and they also allow the release of gases that may permit radionuclide escape (Kennedy *et al.*, 1985). In addition, burrows created by larger animals, allow the penetration of water to a greater depth; for example, burrows upwards of 50 cm deep have been recorded for kangaroo rat, *Dipodomys ordii,* (Reynolds and Arthur, 1983) and ground squirrels, *Spermophilus townsendii*, (Reynolds and Laundre, 1988). Further to the action of animals, plants can also increase the migration of radionuclides within the soil profile. There is even some evidence to suggest that the activity of burrowing animals actually encourages plant root growth (Bishop, 1989; Edwards and Lofty, 1978, 1980). It is feasible that radionuclide uptake into plants could thus be promoted. In addition, root penetration, followed by root death, will leave sub-surface channels down which soil particles can move. Moreover, root uptake, translocation and then death of above ground parts of plants will lead to a redistribution of radionuclides from the soil back to the surface (Kennedy *et al.*, 1985).

Transport of radionuclides adhered to soil particles may occur following the internal or external contamination of animals through ingestion, inhalation and surface contact (Landeen and Mitchell, 1982). Subsequent territorial movements or migration from the contaminated area will redistribute radionuclides. It is considered that this will be a minor factor in the overall spatial distribution of radionuclides in most contaminated areas, although it will depend upon the numbers of each species involved. However, the radionuclides present as either internal or external contamination have a potentially important role in the transfer of radionuclides to higher trophic levels (Arthur and Markham, 1983; section 1.4.4).

1.4.2 Vegetation

Radionuclides enter plants by foliar absorption or root uptake. Much of the data relating to plant uptake havebeen determined using crop species because of the direct relevance and transfer to man. Most of the data obtained relate to global weapons testing fallout (Romney *et al.*, 1963; Wilson *et al.*, 1967), the operation of nuclear establishments or nuclear accidents (Adriano *et al.*, 1981) and tracer studies on plant physiology (Middleton, 1958; Tukey *et al.*, 1961). Some of the features of radionuclide uptake are determined by the vegetation species and are therefore specific; however, general conclusions can be made about the uptake and translocation of radionuclides. These have been reviewed in more detail elsewhere (Cawse and Turner, 1982; Coughtrey and Thorne, 1982; Koranda and Robison, 1978; Russell, 1966).

1.4.2.1 Foliar Absorption

Foliar absorption occurs through the direct contamination of the above ground parts of a plant by either the direct deposition of airborne particulates/aerosols or through soil particles which may be splashed, or blown, on to leaves (Dreicer *et al.*, 1984; Hakonson *et al.*, 1981). Following the deposition of radionuclides to the leaf surface by processes such as sedimentation, impaction, interception and Brownian motion (Chamberlain and Little, 1981), they may be absorbed within the plant either by cuticular penetration or through the stomata. Material taken in through the stomata will be in either the aqueous or gaseous phase.

Cuticular penetration is dependent upon a series of factors; for example, the morphology of the leaf (will determine both deposition and retention rates), chemical composition of the cuticle and the time allowed for absorption to occur. Studies have shown that the longer material remains on the leaf surface, the greater the amount of absorption. Therefore, foliar uptake may be affected by the season during which deposition occurs. For example, Miller and Hoffman (1982) reported a range of environmental half-lives (geometric means of 7 to 20 days), determined experimentally for a range of plant species, radionuclides and climatic conditions. When determining radionuclide exposure and bioavailability, these half-lives are used to determine the loss of radionuclides from plant surfaces via radioactive decay and environmental processes, for example rain- and wind-mediated removal. This may result in the resuspension of airborne particulates, often of respirable size, leading to the exposure of internal organs and/or the redistribution of radioactivity across an area downwind. As for soils, this resuspension is important for actinides whose alpha decay events are more damaging to cells when they are irradiated internally.

The principal factor determining the deposition and retention time for particles deposited to the plant surface, is the leaf morphology. Leaf shape, orientation and density will determine the initial deposition of material (Nay, 1967). Witherspoon and Taylor (1970) showed that plants with a large surface area of foliage, consisting of small leaves, intercepted more of the small particles present in an applied radionuclide aerosol. Furthermore, larger particles were observed bouncing off leaves. These differences are due to changes in the rate of impaction. Impaction occurs when particles deviate from the air flow as a consequence of their inertia, as the air is forced around an object. As the air flows around a large number of small leaves, it is disrupted, leading to a greater rate of impaction. Furthermore, impaction of particles and their subsequent retention are affected by the degree of resinous or waxy coating on the leaf which can entrap the particles (Chamberlain and Little, 1981).

Witherspoon and Taylor (1970, 1971) also showed that leaf shape and form were important in determining the retention time of deposited material. In addition to leaf shape and density,

physical adaptations of different plant species to their environment have an effect on the deposition and retention of particulates. For example, physical features such as: sunken stomatal pits, hairs (stellate or simple), glands (resinous or salt), waxy deposits and spicules will influence deposition and retention rates (Allen, 1984; Fitter and Hay, 1987; Koranda and Robison, 1978). The presence of hairs will increase the rate of interception, as particles are 'filtered' from the air (Chamberlain and Little, 1981).

Following deposition of particulates to the leaf surface, transfer to the internal plant cells is determined by the cuticle. The cuticle surrounds all the external living structures of plants and typically consists of waxy plates which are embedded in a matrix of pectic materials. The cuticle itself is composed of long chain fatty acids, alcohols and esters and acts as a selectively permeable membrane (Koranda and Robison, 1978), often through the use of plasmodesma. These are specialised cells which extend into the epidermis and provide direct connections for the transport of material across the cuticular barrier.

Most of the above discussion has centred on the transfer of particulates deposited on the above ground parts of plants through dry deposition. However, during rainfall the particulates may also be dissolved. The amount retained on vegetation then, will depend on the quantity and droplet size of deposited water. Under heavy rainfall, 'run-off' will occur which may also wash previously deposited material from the leaves. Similarly, droplet size affects deposition and therefore foliar uptake. Under mist or fog conditions, the fine droplet size leads to more absorption, although entry is mainly via the stomata (Russell, 1966).

Gaseous uptake of radioactive materials is well known in plants that absorb carbon dioxide, oxygen and water vapour through stomata. Much of the present knowledge of gaseous uptake derives from the study of non-nuclear contaminants such as sulphur dioxide (Atkinson and Winner, 1990; Jarvis, 1981). For radioactive gases or small particles, foliar absorption may occur similarly through the stomata by simple diffusion processes (Russell, 1966).

1.4.2.2 Root Uptake

Following deposition of radionuclides to soil, they may be absorbed into vegetation through root uptake. Most radionuclides absorbed via root uptake are incorporated within plant tissues, although some re-excretion may also take place. Both soil and plant factors are involved in this process. The overall root uptake of radionuclides is controlled primarily by the degree of fixation and, therefore, their availability in the soil. This is important when current deposition rates are much less than those historically, because soil/root uptake may dominate. In the present study, it is believed that current deposition rates are still high because of the aerial

discharges from BNFL, Sellafield. Furthermore, resuspension and soil splash are expected to contribute significantly to the accumulation of contaminated soil on the plant foliage.

A number of studies have investigated root absorption, and their subsequent translocation, using dilute chemical solutions in the laboratory (D'Souza and Mistry, 1973; Handley and Babcock, 1972). However, these are not directly applicable to the field situation where soil parameters largely control their availability. Therefore, soil to plant transfer factors are used to determine the relative uptake of radionuclides from different soil types, and for different vegetation species. Soil to plant transfer is defined as *'the ratio of radioisotope concentration in the plant (Bq kg^{-1} dry or wet weight) to that in soil to a defined depth (Bq kg^{-1} dry or wet weight)'* (Bettencourt *et al.*, 1988; Kirchmann *et al.*, 1993).

Plant factors that affect root uptake include: rooting depth, root morphology, solute concentrations of a given radionuclide in both the soil solution and within the plant; the enhanced uptake of organic complexes (for example, humic acids which may have complexed with the radionuclides within the soil); whether an active uptake mechanism exists within the plant; and the role of micro-organisms associated with the roots. Several radionuclides are, for example, isotopes of essential elements required by the plant for survival (e.g. iron-55 and potassium-40), or are analogous to elements utilised by the plant (e.g. caesium and strontium). In these cases, an active uptake mechanism exists whereby the plant expends energy to absorb these metals (Russell, 1966). Models of the behaviour of radionuclides in soil and their transfer into plants have been produced (Thorne and Coughtrey, 1983; Crout *et al.*, 1990). Other studies have indicated that the role of mycorrhizal associations may be important in, firstly, liberating metals from the soil binding sites (Frissel, 1987), and secondly, in their transfer into plants (Berthelsen *et al.*, 1995; Byrne *et al.*, 1976; Natural Environment Research Council (NERC), 1993).

1.4.2.3 Translocation

Whether radionuclide absorption into plant tissues occurs through direct foliar deposition or indirectly root uptake, the incorporated radioactive material may then be translocated. This is somewhat dependent upon the radionuclides involved. Tukey *et al.* (1961) reported that radioisotopes of phosphorus and calcium are rapidly translocated following foliar absorption, with phosphorus in particular accumulating in the actively developing meristematic regions such as root tips, flowers, fruits and vegetative growing tips. In a similar way, it has been reported that ^{137}Cs absorbed through the foliage, is readily translocated to developing organs and down to the roots. In contrast, there is relatively low uptake from soil (Tukey *et al.*, 1961). Adriano *et al.* (1981) established that about only 3% of the measured plutonium in field crops near a nuclear establishment was accumulated by root uptake, while subsequent

soil pot experiments in the glasshouse on the same soil and vegetation species exhibited an order of magnitude increase in total plutonium (^{238}Pu and $^{239+240}$Pu) through root uptake under conditions of reduced atmospheric deposition.

One point to be noted, is that root uptake results in the accumulation of radionuclides within the plant tissues while foliar deposition includes radionuclides transported into the plant through foliar absorption but also material superficially bound to the leaf surface. This results in differential activities between the external and internal components. These differences could have an influence on the transfer of radionuclides across the gastro-intestinal tract of those species which consume the vegetation.

1.4.3 Leaf Litter and Flotsam/Strand Line Material

In the present study, plant detritus in the form of leaf litter or strand line material was found on two of the field sites; the coniferous woodland and the salt marsh. Here the detritus formed an integral part of the ecosystem and much of the invertebrate and small mammal activity was observed within. Furthermore, it has been reported that detritus derived from the decomposition of vascular plants has a high adsorptive capacity for radionuclides and other pollutants (Odum and Drifmeyer, 1978). Moreover, the strand line material along the salt marshes of the Esk estuary was water-borne, so any radionuclides present in the water body itself (Chapters 4 and 5) may already have adsorbed on to the detritus before it was deposited. Likewise, leaf litter in the woodland would originally have formed the canopy and therefore would have received primary radionuclide deposition (Chapter 3). Consequently, the detritus forms an important part of these ecosystems both in terms of radionuclide activity and as one of the components in the food chains under investigation. Radionuclide deposition to organic matter or plant detritus is a primary route for the direct transfer of radionuclides into food chains via detritivores and herbivorous small mammals (Adriano *et al.*, 1981; Rickard, 1967; Ritchie *et al.*, 1970).

1.4.4 Invertebrates and Small Mammals

Although there have been many studies on the uptake, distribution and retention of radionuclides in animals, many of them focus on laboratory animals that have been utilised to assess the dose to man following inhalation or ingestion of radionuclides (Kirchmann *et al.*, 1993). Recent concern over the chronic effects of environmental radioactivity and the requirement for a better understanding of the transfer of radionuclides in ecosystems has prompted the examination of food chains without a human focus. In the present study, the radionuclide burden of species of invertebrates and small mammals was assessed to facilitate the understanding of the transfer of radionuclides through semi-natural ecosystems.

1.4.4.1 Invertebrates

Data on the radionuclide burden of invertebrate species in the context of food chains is sparse, mainly because of difficulties in the determination of individual radionuclide activities of samples resulting from the collection of a small biomass in the field. As a consequence, many of the early studies used tracers to monitor the transfer of radionuclides through food chains (Crossley, 1963a, b; Marples, 1966; Reichle, 1967; Reichle and Hook, 1970) or reported radionuclide transfer and retention within a single species (Crossley and Pryor, 1960; Reichle *et al.*, 1971). However, the role of invertebrates in the uptake of environmental pollutants should not be ignored; for example work has shown that uptake and transfer of stable heavy metals through food chains does occur (Hunter, 1984; Hunter *et al.*, 1987a, 1987b; Rabitsch, 1995a, b, c), and significant effects of radionuclides on invertebrate populations following the Chernobyl accident have been described (Spirin *et al.*, 1990).

The parameters that affect invertebrate food chain transfer of radionuclides are similar to those experienced for small mammals. Therefore, the most important are described in the following sections using small mammals as a model. It should be noted that, as with earlier studies, the measurement of ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities in samples of invertebrates collected in the present study was restricted by the small biomass and the time available on analytical equipment (Chapter 2). For these reasons, it was necessary that only those invertebrate species or families which formed key components at each field site were examined (sections 3.7, 4.7 and 5.7).

1.4.4.2 Small Mammals

Several studies have investigated the radionuclide transfer through food chains using small mammal species as bio-monitors (Dunaway and Kaye, 1961, 1963; Johnson *et al.*, in press; Mascanzoni *et al.*, 1990; Rudge, 1993a, b). For the purposes of this study, the definition of the term 'bio-monitor' is taken from O'Brien *et al.* (1993) and is: '*Monitor: organism in which changes in known characteristics can be measured to assess the extent of environmental contamination so that conclusions on the health implications for other species or the environment as a whole can be drawn.*' In this study, changes in the levels of individual radionuclides were measured over and above those of control site animals which are indicative of background radiation levels (section 2.3.6).

Small mammals have been used to assess the impact of non-nuclear pollutants released to the environment, mainly because the analysis of samples of soil, vegetation and water provide information about the contamination in the local area but none regarding the availability and transfer into higher trophic levels. For example, heavy metal concentrations and their transfer

in contaminated ecosystems have been determined using *A. sylvaticus*, *M. agrestis*, and *S. araneus* (Hunter, 1984; Johnson *et al.*, in press; Roberts and Johnson, 1978).

The small mammals found in most ecosystems display a range of dietary habits and, because they remain in a localised area, they are exposed continuously to source of contamination. Furthermore, they also provide a mammalian model to contamination that may, in some ways, be relevant to human exposure (Talmage and Walton, 1991). However, caution should be exercised when extrapolating data to human subjects (Smith, 1992). In practical terms, small mammals are easily trapped and their populations can sustain planned trapping over a period of time allowing temporal variation to be assessed. O'Brien *et al.* (1993) propose a number of criteria from which to determine the best species available to monitor exposure to pollutants. They suggest that the following parameters are important: size, sensitivity (to the pollutant), physiology, longevity, food chain position, migration and abundance. The small mammal species under investigation in this study: the field mouse, *Apodemus sylvaticus*; field vole, *Microtus agrestis*; and common shrew, *Sorex araneus,* fulfil most of these criteria.

In the literature, the most studied small mammals belong to three families: Soricidae (shrews), Cricetidae (mice, rats and voles) and Muridae (Old World mice and rats). These encompass a wide range of dietary habits including: granivores, herbivores, insectivores and omnivores. Other characteristics of these families are: small in weight; highly abundant; common life histories; small home range sizes; important role in the energy flow of an ecosystem; and a short life span but with high reproductive rates (Ryszkowski and French, 1982; Talmage and Walton, 1991). Therefore, any damage to individuals or populations of small mammals caused by exposure to pollutants can be monitored in the present and future generations and there are techniques which provide direct diagnostic assessment of such damage to the animals. For example, cytogenetic tests such as the micro-nucleus test have been carried out on *A. sylvaticus*, the bank vole *Clethrionomys glareolus*, *M. agrestis* and *S. araneus* following the Chernobyl accident and in areas around nuclear establishments (Cristaldi *et al.*, 1985, 1990; Krishna *et al.*, 1991; Manscanzoni *et al.*, 1990). Moreover, the whole body burdens of the same species have been assessed on a low level radioactive waste site (Rudge, 1993a, b).

Small mammals have been widely used to monitor both natural (Burns *et al.*, 1987; Maslov *et al.*, 1967) and artificial radionuclides (Comar, 1966; DiGregorio *et al.*, 1978; Dunaway and Kaye, 1963; French, 1967; Manscanzoni *et al.*, 1990; Rowley *et al.*, 1983) in the environment. Most of these studies have examined environmental effects indirectly using the uptake, distribution and retention of radionuclides in laboratory animals to assess dose and then develop models for predicting the effects of radiation on man. Extrapolation of laboratory data to field situations is difficult. Burns *et al.* (1987) found transfer coefficients for ^{226}Ra from food to voles in the laboratory differed by a factor of two from those recorded in the field.

However, from these studies the important parameters involved in the transfer to-, and metabolism in-, animals have been determined (Kirchmann *et al.*, 1993), and can be summarised as:

1. the fraction of an orally ingested radionuclide that is absorbed by the gastro-intestinal tract and subsequently enters systemic circulation;
2. the activity level and bioavailability of radionuclides in foodstuffs;
3. fractions and half-lives of a radionuclide associated with specific pools in the animal, or with whole body retention;
4. fractions of absorbed activity excreted in urine, faeces, milk and sweat; and
5. the resuspension of radionuclides within the environment and the longevity of radionuclides in the lungs taking into account the chemical and physical form of input.

Clearly, the study of radionuclide transfer through food chains on three field sites precludes detailed examination of all of the above parameters. However, the results reported here can be used as pilot data for a further, more detailed study.

The major parameters affecting the radionuclide uptake and transfer through food chains have been described as: the sources of radionuclides and their chemical and physical form, identification of specific transport pathways and the assimilation at each link, and the turnover or retention rates for successive receptor species in the appropriate trophic chain (Reichle *et al.*, 1970). These factors have been discussed elsewhere (DiGregorio *et al.*, 1978; Kitchings *et al.*, 1976). A description of the transfer mechanisms for the uptake of radionuclides into small mammals is given below.

1.4.4.3 Inhalation

For animals living in an contaminated area, inhalation is a potentially significant route of exposure, both in terms of radionuclide uptake and dose received. Radionuclides may be inhaled in gaseous or particulate form, with subsequent deposition in the lungs for particulates. These particulates may then be solubilised and absorbed into the body. Gaseous radionuclides, such as ^{129}I, may be absorbed directly across the pulmonary lining. Inhaled radionuclides may: cause irradiation of the lung tissue, be absorbed into the systemic circulation and then translocated around the body, be moved up from the lungs by the action of the ciliated epithelium and then ingested subsequently with possible gastro-intestinal absorption and systemic translocation (Bair, 1960; Comar, 1966). The latter may also be important for particles larger than 5 to 10 μm which are generally considered too large to penetrate alveolar tissue (Kirchmann *et al.*, 1993).

Deposition, retention and absorption within the lungs are determined by a number of factors including: chemical form of the radionuclides adhered to the inhaled particle, particle size, and solubility. The damage caused is related to: the duration of exposure and the radioactivity associated with the particle (Comar, 1966; ICRP, 1989). Although it has been shown that absorption of radionuclides by animals through inhalation is less important than ingestion, Hvinden *et al.* (1964) indicated an expected difference of approximately three orders of magnitude in the uptake of ^{131}I in the milk of grazing cattle via the two different routes if the cattle were feeding on pasture at the time of deposition during the passage of a radioactive cloud. Summerling *et al.* (1984) showed that grazing animals were also subject to inhalation as well as ingestion but local measurements showed that atmospheric processes accounted for less than 1% of the daily intake. However, it is suggested that resuspension of radionuclides associated with soil particles will lead to the internal absorption of radionuclides through inhalation, particularly for small mammals which inhabit enclosed underground tunnels or chambers. As mentioned earlier, this can be more significant for the actinides which have very low transfer rates across the gastro-intestinal lining but which are associated with resuspended particles (section 1.4.4.5).

1.4.4.4 Direct Contamination of Fur and Skin

Animal skin, like the plant cuticle, forms a formidable barrier to the direct transfer of radionuclides into the body. However, damaged skin may lose some of its protective ability and a very limited transfer of radionuclides may occur. This is unlikely to be a significant component of the whole body burden. Nevertheless, radionuclides will be transferred on to the skin or fur of small mammals, usually through the adherence of soil particles, during their movement both around and within their burrows. Depending upon the activity on the soil particles, this will firstly produce radiation exposure localised to adjacent areas of skin and underlying tissue and, perhaps more significantly, material may be ingested during grooming with the possible transfer across the gastro-intestinal tract (Lang *et al.*, 1993). The close proximity of radionuclides to the animals' skin and underlying tissues will again be more important for the actinides since any alpha particles emitted are rapidly halted even by a few centimetres of air.

1.4.4.5 Ingestion

Ingestion of radionuclides either directly through the contamination of food items or indirectly through inhalation and/or grooming, forms the most significant pathway for the uptake of radionuclides. The rate of uptake is proportional to the feeding rate and the concentration of individual radionuclides within food items. During subsequent passage through the gastro-intestinal tract, the fraction of radionuclides assimilated into the body, is

related to its physical and chemical properties (Comar, 1966) and the physiology of the animal species (Kitchings *et al.*, 1976; Whicker, 1983). In addition, most animals also ingest small quantities of soil either accidentally or on purpose during grazing and any soil particles which are adhered to the exoskeleton, skin, gut, fur or hair of prey species will be accidentally ingested also (Comar, 1966; Howard, 1987; Rudge, 1989). Subsequent release and transfer across the gastro-intestinal tract will depend upon the properties of the radionuclide and the strength of its association with the soil particle (Kirchmann *et al.*, 1993). The acidity of the gut may also influence the disassociation of the radionuclide from the soil.

The absorption of radionuclides across the gastro-intestinal tract is related to a number of factors, not least the time spent in the gut. This is related to the function, length, number of compartments and size of the digestive tract; for example, the digestive tract in herbivores is much longer than in carnivores. There also tend to be more compartments in herbivorous digestive systems. In ruminants for example, the first compartment forms a large stomach, the rumen, for the digestion of fibrous cellulose plant material (Kirchmann *et al.*, 1993). These differences prolong the residence time for radionuclide absorption from the gut of herbivores. The bulk of the digested food is absorbed through the small intestine and there are different absorption rates for different radionuclides. This is because of the epithelial lining of the gut, which is impermeable to particulate matter, hydrocarbon emulsions, and water-insoluble, and lipid-insoluble solutes of molecular weights greater than 100 (Comar, 1966). The transfer of digested food across this lining can be an active or a passive process. This influences the expenditure of energy to actively pick up and then transfer material across the gut wall by processes such as: active transport, facilitated diffusion and pinocytosis; or by simple diffusion. This in itself leads to differences in the uptake of specific radionuclides. Calcium is absorbed by both active transport and simple diffusion while strontium is absorbed only by simple diffusion (Kirchmann *et al.*, 1993). This is reflected in an increased uptake of calcium compared to strontium. The actinides, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am, are barely absorbed at all. It should also be noted that radionuclides may also be excreted from the blood into the lumen of the gut (Kirchmann *et al.*, 1993) and that a dynamic equilibrium will be produced. Table 1.2 presents derived gastro-intestinal tract uptake factors for those radionuclides of interest in this study.

The behaviour of radionuclides absorbed across the gastro-intestinal tract is determined by the physiological characteristics of the animal and the physico-chemical properties of the radionuclides. Initially, radionuclides are present within the systemic circulatory system and are transported around the body as a whole.

Particular organs or tissues may then accumulate specific radionuclides. It is well known that iodine concentrates within the thyroid; caesium in the soft tissues such as muscle; and

strontium in bone (ICRP, 1986). Furthermore, radionuclides are subject to removal via the excretory system and through processes such as lactation, a potentially important mechanism for the transfer of radionuclides to young.

Table 1.2: Gastro-intestinal tract transfer factors for various radionuclides in different animal species as an example of differences between, americium, caesium and plutonium.

	Rat	*Human*
Caesium	0.25-0.85 [a]	0.56-0.90 [b]
Plutonium	0.006 to 0.41 [c]	0.001 [d]
Americium	0.03 [c]	0.001 [d]

Ranges given relate to the initial ingestion of the radionuclide in different chemical forms, for example plutonium as plutonium nitrate, citrate or as the insoluble oxide.
References: [a]=Kitchings *et al.* (1976); [b] = ICRP (1989); [c] = ICRP (1972); [d] = ICRP (1986).

The accumulation and fate of radionuclides within the body is determined by their physiological characteristics. This is affected by the rate of metabolism of the species involved. ICRP (1972) provide information on the accumulation and distribution of radionuclides following ingestion. For example, caesium is taken up and metabolised readily by mammals although the absorption across the gastro-intestinal tract varies and is influenced by a range of factors (Beresford *et al.*, 1992), including age and physiological status (Harrison and Fritsch, 1992; Mayes *et al.*, 1992). Absorption is greatest, for example, when caesium is ingested in soluble form while the accidental presence of clay particles in the gut has been shown to reduce uptake (Kirchmann *et al.*, 1993). ICRP (1989) reported that, in humans, absorption of ^{137}Cs varied from 56 to 90% with a mean of 78% following the consumption of contaminated food. Once absorbed, caesium is distributed uniformly through the soft tissues, such as muscle and some organs such as the kidney and liver (ICRP, 1989; Muller and Scheffer, 1982; Richmond, 1980), and in other respects behaves like potassium (ICRP, 1989).

Actinides, on the other hand, have a very low transfer rate across the gastro-intestinal tract which depends upon the chemical state of the element. They are nevertheless considered to be of great radiological significance. After absorption, over 80% of the plutonium present in the human blood stream accumulates in bone and liver, although the accumulation in bone is usually greater (ICRP, 1986; Spiers and Vaughan, 1976). Similar observations were made for americium. ICRP data used for these comments are from the study of volunteers and also from the extrapolation of data collected from animal exposures. Therefore, it can be assumed that the radionuclides in question will behave in a similar manner within the body of small mammals under investigation here.

Most of the data available on the retention of radionuclides within the body come from animal exposures. DiGregorio *et al.* (1978) provides a review of the retention times of different radionuclides for a number of terrestrial vertebrates. The usual methods of removal involve the transfer of radionuclides from the blood into the gut lumen followed by egestion or by excretion from the kidneys in urine. It has been reported that caesium is removed mainly in the urine (Stather, 1970). The actual rate of radionuclide removal is related to two components. The first is a function of radioactive decay and is therefore connected with the half-life of the radionuclide. The second is related to the physiology of the animal and physico-chemical properties of the radionuclide, much as for absorption and accumulation. In particular, species characteristics such as body weight, sex, age, metabolism and biochemistry and external environmental factors such as temperature, food intake and the continued presence of the radionuclide within the animals' dietary components, will control the excretion rate (Coughtrey, 1990; Hoek van den, 1989). DiGregorio *et al.* (1978), Mailhot and Peters (1989), Richmond (1980) and the ICRP (1989) describe these factors in greater detail and provide mathematical models to determine the rates of absorption and excretion, discussing related factors such as retention and biological half-lives.

The relationship between the absorption and excretion of the radionuclides will, for the chronic exposure conditions of the present study, produce an equilibrium for the intake and removal of radionuclides in small mammals and invertebrates. For ^{137}Cs this equilibrium is usually reached reasonably quickly, as empirical evidence shows. Following the pulse of Chernobyl-derived radioactivity deposited across the United Kingdom in 1986, ^{137}Cs levels in milk increased rapidly and then decreased during May and June of 1986, corresponding to the changes monitored on the vegetation on which the cattle fed. Similarly, Lowe and Horrill (1988) recorded a fall in ^{134}Cs concentrations in roe deer muscle from 108 mBq/g to 17 mBq/g between May 1986 and July 1986, two months later. This suggests that the animals rapidly equilibrate with the ^{137}Cs activities of their foodstuff. In contrast, the actinides are likely to require a considerable time before an equilibrium is reached because once in the body, the actinides accumulate particularly in the skeleton and it is known that the retention half life in humans is in the order of 50 years (Jones, pers. comm.).

In the present study, it was difficult to determine either a biological or ecological half-life since a source for the continued input of radionuclides to the local environment exists and there is a substantial inventory at all three sites of radionuclides accumulated in soil over the period of many years. Certainly this is true for the ecological half-life which is defined as: *'the time required for the nuclide concentration within the body to decrease by half while the animal remains in its natural habitat following an accidental release or the cessation of a controlled release of radionuclides'* (Lowe and Horrill, 1988). For most animal species, a short and a long component have been found for the biological half-life but this is related to

the method of introducing the radionuclide into the organism (DiGregorio *et al.*, 1978). Typically, these components last a few hours to a few days, although the specific times are related to the body weight and metabolism of the animal species. DiGregorio *et al.* (1978) quote laboratory studies which obtained biological half-lives for ^{137}Cs in the order of 6.3 to 8.6 days for a 130 g cotton rat, *Sigmodon hispidus*, compared to 3.5 to 3.9 days for the smaller harvest mouse, *Reithrodontomys humulis*, which weighs less than 10 g. The latter, is similar in size and weight to *S. araneus*, whilst *A. sylvaticus* and *M. agrestis* vary in weight up to about 30 g. Consequently, it is expected that ^{137}Cs in these species would have biological half-lives in the order of a few days, with *S. araneus* being slightly shorter due to the increased metabolic rate and smaller size. Since it is generally assumed that the time required to reach equilibrium is related to the biological half-life and metabolism, these values provide further background evidence that concentrations in the small mammal species of the present study, will rapidly equilibrate with the ^{137}Cs levels within the dietary constituents.

One of the reasons for studying three small mammal species, *A. sylvaticus*, *M. agrestis*, and *S. araneus*, is their dietary differences. This permits an examination of firstly, the transfer of radionuclides through different food chains within each field site and, secondly, the concentration of the radionuclides within the body and the subsequent doses received. Within this study, two species are almost entirely insectivorous or herbivorous, *S. araneus* and *M. agrestis* respectively, whilst the third, *A. sylvaticus*, is primarily granivorous but will also take invertebrates and vegetation at specific times of the year. To avoid repetition later, the dietary habits, seasonal changes, metabolism and foraging methods are described for each species below.

1.4.4.6 Diet of the Field Mouse, *Apodemus sylvaticus*

The field mouse, or wood mouse as its also known, *A. sylvaticus*, is found throughout most of Britain. It is an abundant species of many different habitats and in the present study was the only small mammal species to be caught at each field site. It forms a complex network of underground burrows and chambers used for both food storage and nesting. Corbet and Southern (1977) reported that the species is commonly found wherever cover is provided by trees or shrubs but it is particularly common in woodlands. The behaviour has been well studied and in general the species is nocturnal, with dawn and dusk activity peaks during the winter and a single peak of activity in summer around dusk.

Males are significantly more active than females, with home range sizes which vary according to the season and habitat from 0.1 to 0.17 ha for females and 0.18 to 0.31 ha for males. Generally, male home range sizes are twice that of females (Corbet and Southern, 1977; Kikkawa, 1964; Korn, 1986). The individual life expectancy in the wild is somewhere

between 18 and 20 months, although only a small proportion of the population can be expected to achieve such longevity. The population numbers peak between September and October, declining to the lowest point in March or April. There is no evidence of cyclic fluctuations in the population size (Flowerdew, 1985; Gurnell, 1978). The population dynamics and behaviour of *A. sylvaticus* have been reviewed in detail by Flowerdew (1985) and Montgomery and Gurnell (1985).

The omnivorous diet of *A. sylvaticus* is more complex than for the other two species caught in this study. However, it is predominantly granivorous (Churchfield and Brown, 1987; Hansson, 1985; Miller, 1954; Montgomery and Montgomery, 1990; Watts, 1968; Zubaid and Gorman, 1991). Watts (1968) provided a detailed breakdown of the dietary components during the year and clearly indicates the importance of seed, which ranges from 20% by volume of ingested food during the summer to 99% during the winter/early spring. In late spring, seedlings and new vegetation growth become important as the availability of seed declines (forming up to 50% by volume). Into the summer months, the presence of invertebrates within the diet becomes more important, constituting up to 80% of the ingested food by volume, although Watts (1968) suggests that this may be higher than normal because of an outbreak in leaf eating caterpillars during the sampling period. Churchfield and Brown (1987) found that the invertebrates commonly present within faecal pellets were mainly from the insects: adult Coleoptera, Lepidoptera, Diptera and Hemiptera. The latter three were mainly larvae but a small percentage of adults were also recorded. In addition, Araneidae and Lumbricidae also formed 7 and 12.5% of the samples respectively. One further dietary change noted by Watts (1968) is the increase in the ingestion of fruits and the fruiting bodies of fungi during the autumn (up to 40% by volume). This is important because it has been recognised that certain fungal species are efficient accumulators of radionuclides, particularly radiocaesium (Clint *et al.*, 1991; Dighton *et al.*, 1991).

1.4.4.7 Diet of the Field Vole, *Microtus agrestis*

The field vole is ubiquitous throughout Britain and is slightly larger than *A. sylvaticus* with a maximum weight of 35 to 40 g. It favours rough ungrazed grassland where well-formed runways through the base of the vegetation provide evidence of their presence. Low density populations are also found in woodlands, particularly deciduous which have a good understorey layer, hedgerows, dunes, and moorland (Hansson, 1971). Voles are active throughout the day, although there is some evidence of a peak around dusk. Home ranges vary depending upon the habitat and presence of other voles, ranging from 0.17 to 0.07 ha. As with *A. sylvaticus*, males tend to have larger territories than females, although these tend to alter slightly in size and distribution over time (Myllymaki, 1977). *M. agrestis* populations typically exhibit cyclic fluctuations with a periodicity of three to five years between

maximum densities. A number of hypotheses have been suggested to explain these cycles, for example, a time-lag density dependence mechanism (Hornfeldt, 1994). This, and the other hypotheses, are extensively reviewed by Krebs and Myers (1974). Individual lifespans of approximately 15 months have been reported, although Corbet and Southern (1977) indicated that only 2.3% of the population can be expected to reach this age. Corbet and Southern (1977) also provided a more detailed review of the behaviour and population dynamics of *M. agrestis*.

M. agrestis is almost entirely herbivorous (Churchfield and Brown, 1987; Evans, 1973; Faber and Ma, 1986; Ferns, 1976; Hansson, 1971). Churchfield and Brown (1987) reported that 80 to 100% of the diet throughout the year comprised the fresh growth of leaves and stems of the grasses (Gramineae: *Holcus lanatus*, *Agrostis capillaris* and *A. stolonifera*). The remainder consisted of fresh growth of herbs and a small (up to 9% by volume) proportion of invertebrates, mainly insect larvae of Diptera and Lepidoptera. These may have been ingested more by accident than intention, a mechanism described by Zamora and Gomez (1993). Other invertebrate species include Isopods, Araneids and Lumbricids, but there was no reported seasonal pattern in their consumption. At other localities, mosses (up to 20% by volume), rushes, grass seeds, liverworts and herbs have been recorded (Faber and Ma, 1986; Ferns, 1976). These showed seasonal patterns in consumption, especially for mosses which increased in importance during the spring and summer (Faber and Ma, 1986; Ferns, 1976). The food utilisation of bark, root and dead stem tissue has also been reported, although this comprises a minor part of the diet, as do fungi during the fruiting season in the autumn (Corbet and Southern, 1977; Faber and Ma, 1986; Kalela, 1962).

It should be noted that *M. agrestis* is selective in terms of which species are consumed. In mixed grassland swards the more palatable species such as *Agrostis* and *Festuca* are favoured over the coarse grass, *Dactylis* (Ferns, 1976; Summerhayes, 1941). This appears to relate to digestibility of the different species and has been assessed by Ferns (1976). However, selective feeding is important because different species of vegetation take up radionuclides in different proportions.

1.4.4.8 Diet of the Common Shrew, *Sorex araneus*

S. araneus is much smaller than *A. sylvaticus* and *M. agrestis*, weighing up to 7 or 8 g. Partly due to their small size, and partly due to the diet, *S. araneus* has a much higher metabolic rate than other species, to the extent that it is active for most of the diurnal cycle (Buchner, 1964). Corbet and Southern (1977) reported that there are about ten periods of activity during 24 hours, punctuated by short periods of rest. These activity periods peak around dawn and dusk. The energy requirements of *S. araneus* mean that a daily food intake equivalent to about three

quarters of their body weight is necessary (Crowcroft, 1957; Gebcynski, 1965; Hawkins *et al.*, 1960). However, like mice and voles, *S. araneus* have a maximum life expectancy of 18 to 20 months, and may be much shorter due to an apparent replacement of older animals by the juvenile generation during the summer months. The annual summer mortality of old adults may be related to their inability to maintain a territory in the presence of juveniles (Corbet and Southern, 1977). As with *M. agrestis*, there are population fluctuations which are cyclic, with a period of three to four years between the maximum densities (Heikura, 1981).

Like *A. sylvaticus* and *M. agrestis*, *S. araneus* is distributed throughout Britain in almost all habitats, provided that there is some low vegetation cover for protection. It is most abundant in rank ungrazed grasslands, bushy scrub and bracken. Like most small mammals, it forms runways in the litter/vegetation and tunnels through the top 30 mm of the soil. This tunnelling is related to their foraging behaviour following sub-surface prey detected by auditory and tactile stimuli (Churchfield, 1980; Holling 1958).

The diet of the common shrew is almost entirely insectivorous (Meharg *et al.*, 1990; Pernetta, 1976; Rudge, 1968). Because the shrew is a opportunistic predator which relies on tactile, olfactory and auditory methods of prey detection, the diet consists mainly of invertebrates within the surface soil and organic litter horizons. The major dietary components are: Lumbricidae, Mollusca, Opiliones, Araneidae, adult and larvae forms of Coleoptera and Diptera, and larvae of Lepidoptera. A small amount of vegetation, mainly grass leaves and stems, has also been recorded in samples collected during the spring and summer months (Butterfield *et al.*, 1981; Churchfield and Brown, 1987). *S. araneus* also feeds whilst burrowing so the diet contains a higher proportion of Lumbricidae and other soil-dwelling organisms. The abundance of prey may be responsible for seasonal trends in the dietary components recorded by Churchfield (1984), with larvae of Coleoptera and Diptera being more prevalent during the winter and spring. Lumbricids, gastropods and adult Coleoptera form the backbone of prey utilised throughout the year.

One additional feature of *S. araneus*, noted by Crowcroft (1957) and studied by Loxton *et al.* (1975), is coprophagy. Coprophagy has been observed in *S. araneus* and occurs when the animal everts its rectum and licks a white liquid from the everted tube. Coprophagy occurs predominantly during the daytime, when activity and food intake are lower. It is suggested that coprophagy is a mechanism employed during periods of inactivity to supplement the reduced food intake, possibly by allowing extraction of more nutrients and energy from previously ingested food (Alexander, 1993; Loxton *et al.*, 1975).

1.4.4.9 Dose Assessment

The invertebrates and small mammals inhabiting the field environments around BNFL, Sellafield, are continually exposed to low levels of radiation. Living in such close and continuous proximity to the dispersed radionuclides, there may be genotypic or phenotypic stresses placed upon the animals. Recent work on non-nuclear contaminants has shown that *C. glareolus* and *M. agrestis* inhabiting grasslands contaminated by industrial sources of fluoride, exhibit severe dental lesions (Boulton *et al.*, 1994; 1995). Similar stresses have been shown for invertebrates and small mammals exposed to heavy metals (Dodds-Smith *et al.*, 1992; Pascoe *et al.*, 1994) and pesticides (Cathey, 1982; Ebere and Akintonwa, 1995).

The influences of ionising radiation on plants and animals depend on the level of radiation within the environment. For example, damage to wildlife and vegetation around the Chernobyl nuclear power plant in 1986 was in response to the acute radiation exposure resulting from the high levels of radionuclides released, and their concentrated deposition in the local area. In comparison, the levels of radionuclides around BNFL, Sellafield are at a much lower, but chronic, level. The effects of the different levels of radiation exposure have been studied for a wide range of plant and animal species, and at different scales of biological organisation from cells, organs to the whole body. This has been reviewed recently by Woodhead (1993) and Prasad (1995) but also for plants by Whicker and Fraley (1974) and animals by Turner (1975).

In general, ionising radiation acts by depositing energy and thereby inducing change at the molecular level which may then be reflected at higher levels of biological organisation. The effect of the ionising radiation is radionuclide specific and also to the plant and animal tissues which are affected. Furthermore, the effects of radiation may not be apparent unless there is a clear response of individuals by way of increased mortality (at the most extreme), reduced fertility and fecundity, and damage to the individual's genes (Woodhead, 1993). By studying individual populations inhabiting contaminated areas, these characteristics may not be apparent, and only by comparing them with control populations may differences in the 'fitness' of individuals be observed.

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Chapter Two
ANALYTICAL METHODS

2.1 INTRODUCTION

Details concerning the sampling programme for each of the three study sites are given in the relevant chapter and include a discussion about the sample collection and preparation for counting (sections 3.3, 4.3 and 5.3). Each sample collected during the project was analysed for a number of gamma-emitting radionuclides, particularly ^{134}Cs, ^{137}Cs, and ^{241}Am, using gamma spectroscopy. In addition ^{238}Pu and $^{239+240}Pu$ activities were determined by alpha spectroscopy. Where possible ^{241}Am was analysed by gamma spectroscopy because of the longer time required for the radiochemical separation of ^{241}Am. Similar methods of data analysis were employed for the two counting systems but are discussed separately (sections 2.2.5 (alpha spectroscopy) and 2.3.7 (gamma spectroscopy).

2.2 ALPHA SPECTROSCOPY

2.2.1 Principles of Alpha Spectroscopy

An alpha decay event results in the emission of an alpha particle which consists of a helium atom ($^{4}He^{2+}$) released from a parent nucleus. Therefore the progeny has a mass number four less, and an atomic number two less, than the parent nucleus. The emission energies for alpha particles are typically high and cover a range of energies, for example 2.0 MeV (^{148}Sm) to 11.1 MeV (^{217}Ac) (Browne and Firestone, 1986). Despite these high emission energies, the physical size and electrical charge of the helium nucleus limit the range or distance that an alpha particle can travel. For example, for alpha particles of energy between 4.0 and 10.0 MeV, the penetration range in silicon has been measured as between 15 and 65 μm (Deme, 1971). The alpha-emitting radionuclides of interest in this study were ^{238}Pu (5.50 MeV), $^{239+240}Pu$ (5.19 MeV) and ^{241}Am (5.49 MeV) (Browne and Firestone, 1986) so that the penetration range in silicon could be closer to 15 than 65 μm.

The inability of alpha particles to penetrate any distance in most materials and in air, and the self absorption within samples of significant mass, provide unique problems for measuring the activities of alpha-emitting radionuclides. As a consequence, it was necessary to prepare samples for alpha spectrometry analysis, firstly by chemical separation to obtain a high purity extract of the element in question and, secondly, to produce thin almost weightless sources which were in a geometry suitable for counting. Obtaining thin sources of low mass was critical to avoid the partial absorption of the emission energy within the sample. If self-absorption occurs, the energy spectrum of the emitted alpha particles is degraded and consequently more difficult to analyse. This is particularly important when determining the

activity of radionuclides with similar emission energies such as isotopes of americium and plutonium, because a good peak shape is required to resolve them clearly.

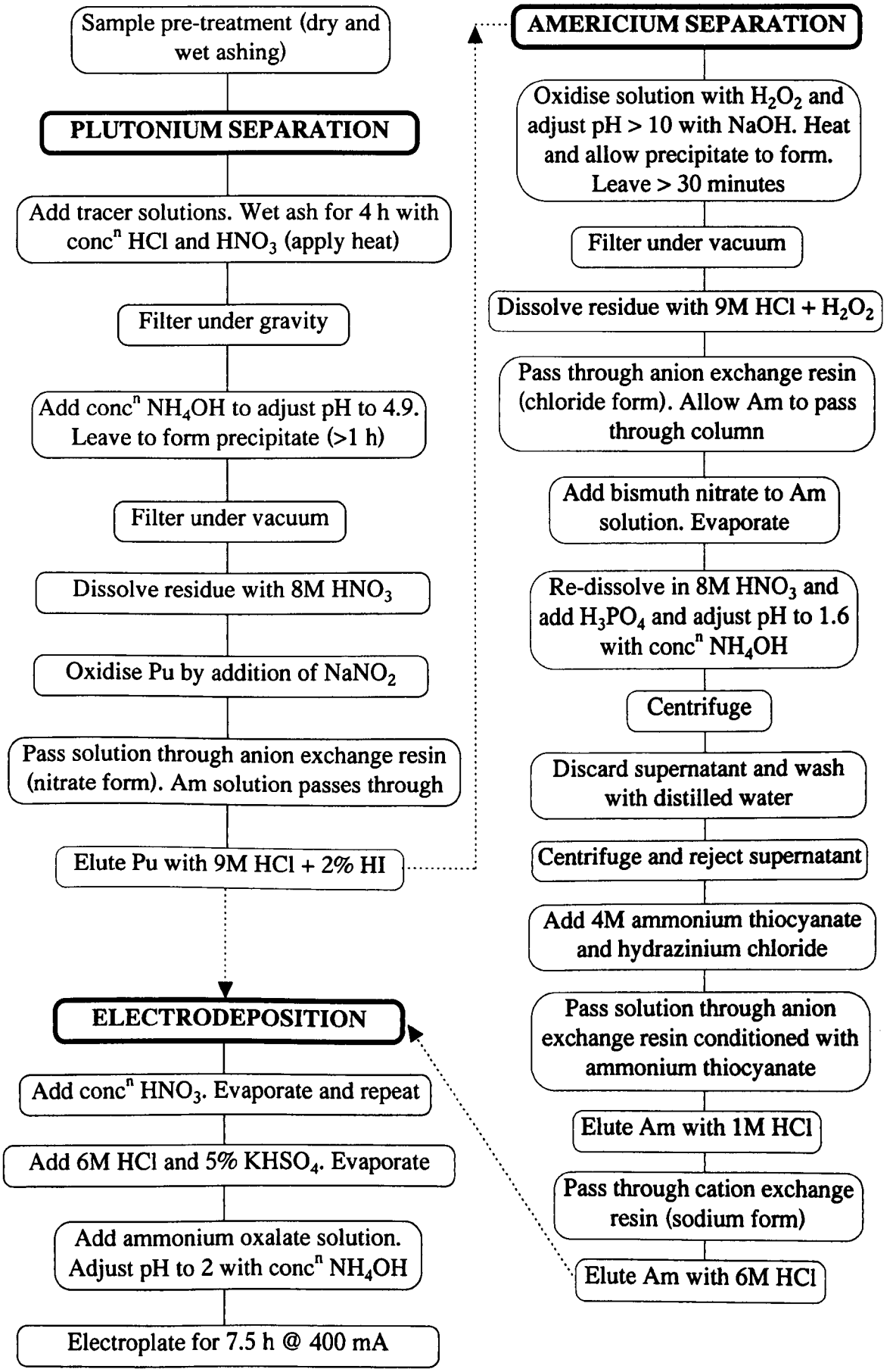
There are many techniques that have been developed to separate isotopes of alpha-emitting radionuclides, in particular americium and plutonium, from environmental samples (Bunzl and Kracke, 1994; Cross and Hooper, 1987; Department of the Environment (DoE), 1989; LaRosa *et al.*, 1992; Singh and Wrenn, 1988; Talvitie, 1971). BNFL have also developed methods for determining levels of americium and plutonium in environmental media. The method used for the chemical separation of americium and plutonium in this study is presented in detail in Appendix A. This method was developed from the BNFL and MAFF methods (Lovett *et al.*, 1990) to allow separation and subsequent electrodeposition of americium and plutonium from a range of environmental materials with widely differing radionuclide concentrations. The full method is reported here in detail (Murdock, pers. comm.).

Figure 2.1 presents a flow diagram of the radiochemical separation method described in Appendix A. A typical radiochemical batch consisted of eight samples. The performance of the procedure was monitored through the addition of tracer solutions at the start of the separation. Each sample in the batch was spiked with ^{236}Pu and, where necessary, ^{243}Am. By spiking individual samples, an accurate estimate of the recovery of the americium or plutonium from each individual sample could be made. Also, because all eight samples were spiked, it was possible to calculate a mean recovery for the batch. This allowed the consistency of the radiochemistry to be monitored both within, and between, sample batches. Typical mean recovery of the ^{236}Pu standard within a batch was 85.0% ± 10.8% (one standard deviation). Over the course of the study period the yield recovery showed acceptable consistency with an average of 87.7 ± 10.4% (one standard deviation). For ^{243}Am, these values were 60.9 ± 6.7% and 59.4 ± 9.8% respectively. It should be noted that the calculated uncertainties for alpha-emitting radionuclides reported here do not include any component originating from uncertainty in estimation of the yield recovery.

It has been reported (Cross and Hooper, 1987) that ^{242}Pu is a better yield tracer when determining both ^{238}Pu and $^{239+240}Pu$ concentrations within environmental matrices, principally because during the alpha spectroscopy, the tail of the ^{236}Pu may degrade and extend into the ^{238}Pu peak. It should be noted that during this study no such problems were encountered and for all samples analysed the peaks were clearly resolved. There are no such problems with the use of ^{243}Am.

After the americium and plutonium had been chemically separated, the nuclides were electroplated on to scribed stainless steel discs. This ensured that the samples were in a

Figure 2.1: Flow chart of radiochemical separation method (Appendix A).



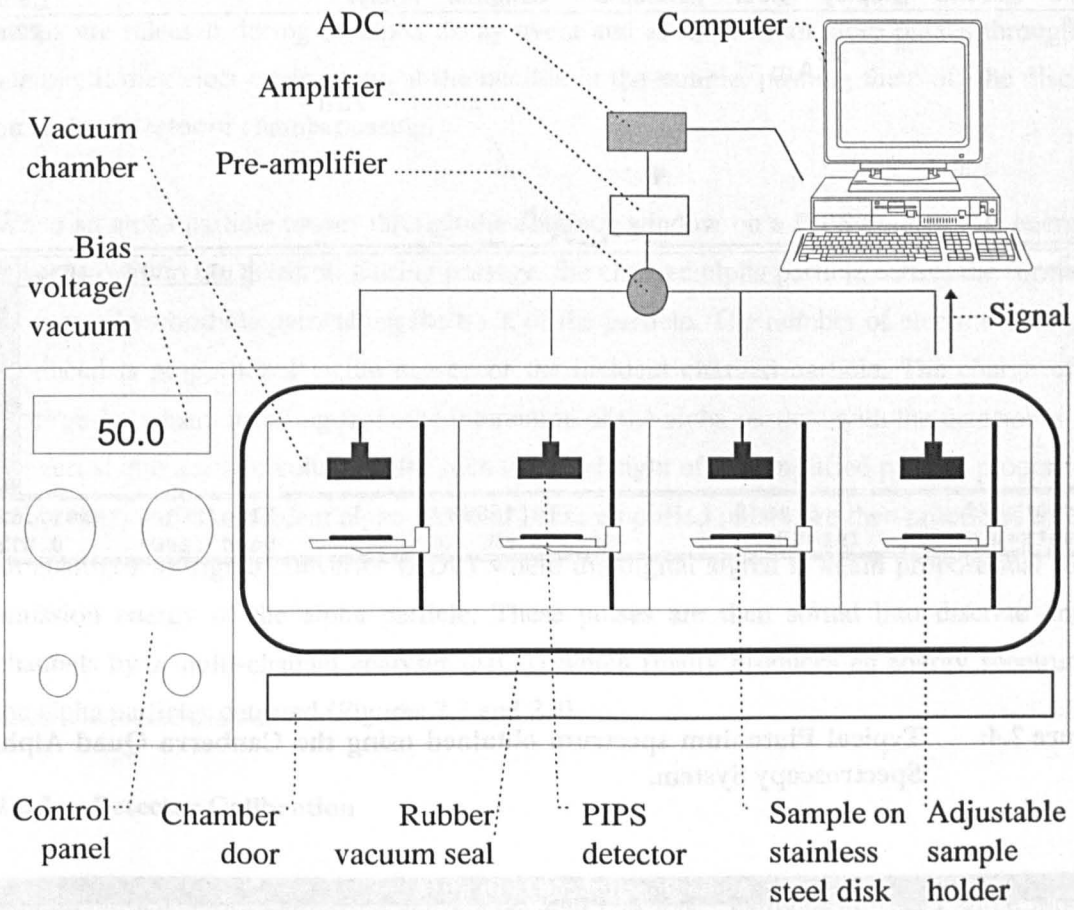
standard geometry for counting. In addition, the method of electrodeposition forms a very thin deposit of the nuclide, thus reducing the problem of self-absorption. The electrodeposition method was modified from the MAFF procedure that describes the use of ammonium oxalate at pH 2.0 (Lovett *et al.*, 1990), mainly so that disposable electrodeposition cells could be used. A number of the samples analysed, particularly those of the invertebrate and small mammals, were of low activity and it had been noted in the past that contamination of the glassware and particularly the electrodeposition cells could cause problems. In addition, samples were separated into either high or low activity categories based on previous experience and separate glassware was used as appropriate. Occasionally, blank samples were run and any build-up of contamination was assessed. In all of these cases, when using the modified electrodeposition method, it was not possible to distinguish the blank samples from the backgrounds measured in the detector chambers.

2.2.2 Determination of Alpha Activity

During this study, two Canberra QUAD-ALPHA spectrometers were used to count separated alpha samples of americium and plutonium. Each spectrometer contained four Passivated Implanted Planar Silicon (PIPS) detectors with a surface area of 450 mm^2 and was made from low alpha background materials. The samples were placed into moveable holders which, for consistency, were fixed at 10 mm from the PIPS entrance window. Previous work showed that a distance of 10 mm provided optimum counting efficiency with good peak resolution (Murdock, 1992). Once the samples were placed into the detector chambers, the doors were closed and the system evacuated. The detectors have to be operated in a vacuum of $\leq$ 20 millibars because the presence of any water or gaseous particles within the detector chamber affects the penetration range of alpha particles. This in turn, affects the energy resolution of the peaks in the spectrum (Knoll, 1989). Moreover, at higher atmospheric pressures, there would be a significant current leakage from the surface of the detectors which would decrease the response rate and energy resolution of the detector (Deme, 1971). When the pressure within the detector chambers reached a pre-determined level, the detector bias supply was applied in stages until it reached 50 V. The sample counts were then acquired. Each detector was connected in sequence to a preamplifier, amplifier, analogue to digital converter (ADC) and finally the count information was routed to an IBM PS/2 personal computer running the Canberra Series 100 acquisition software. Figure 2.2 presents an outline of the alpha spectrometer arrangement and Figures 2.3 and 2.4 show typical spectra obtained from counting americium and plutonium sources.

The use of PIPS detectors has a number of advantages over the previously used silicon surface barrier detectors, primarily the ease with which they can be decontaminated. This is important because contamination leads to a higher background that will interfere when counting samples

Figure 2.2: Schematic outline of the Quad Alpha Spectroscopy System.



of low activity. Silicon surface barrier detectors utilise fragile evaporated metal contacts which cannot be touched or cleaned easily. PIPS detectors, on the other hand, employ photolithographic procedures which allow accurate detector junctions to be made; also, these junctions are buried within a silicon wafer. Both of these features help to reduce current leakage (Knoll, 1989). Canberra Nuclear, the suppliers of the PIPS detectors, have also developed proprietary techniques for generating thin (<0.05 μm) ion implanted entrance windows which are rugged enough to withstand cleaning. These features improve the resolution and counting efficiency of PIPS detectors compared to silicon surface barrier detectors.

To decontaminate the detectors and minimise the background, they were removed from their housing and carefully swabbed with methanol. This was carried out at regular intervals. The detector surfaces were wiped several times with fresh swabs to ensure thorough

Figure 2.3: Typical Americium spectrum obtained using the Canberra Quad Alpha Spectroscopy System.

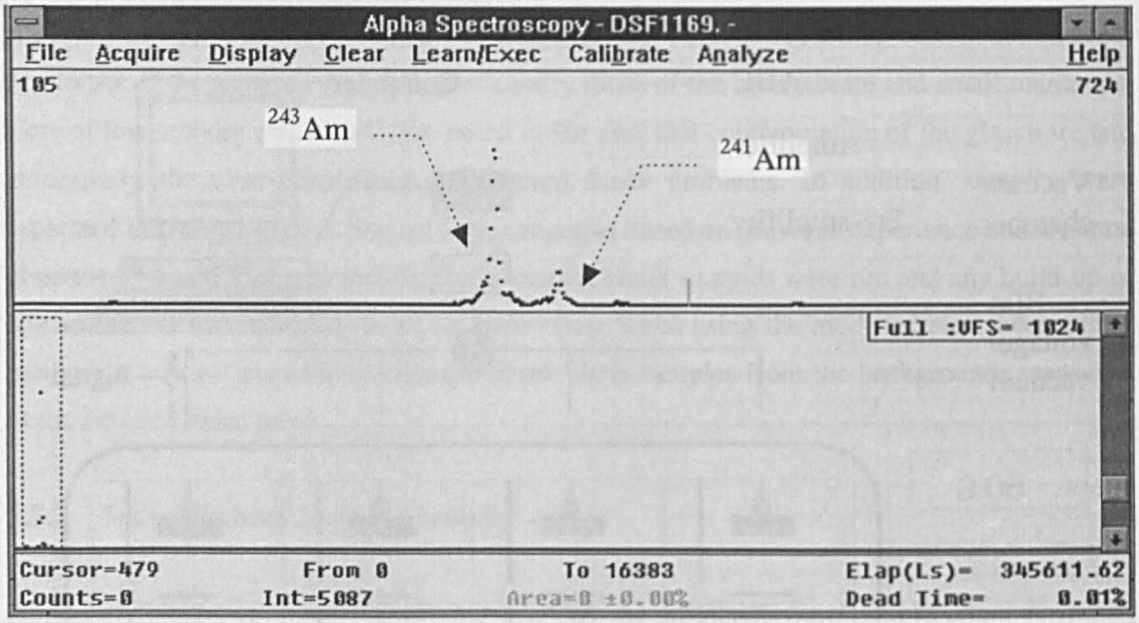
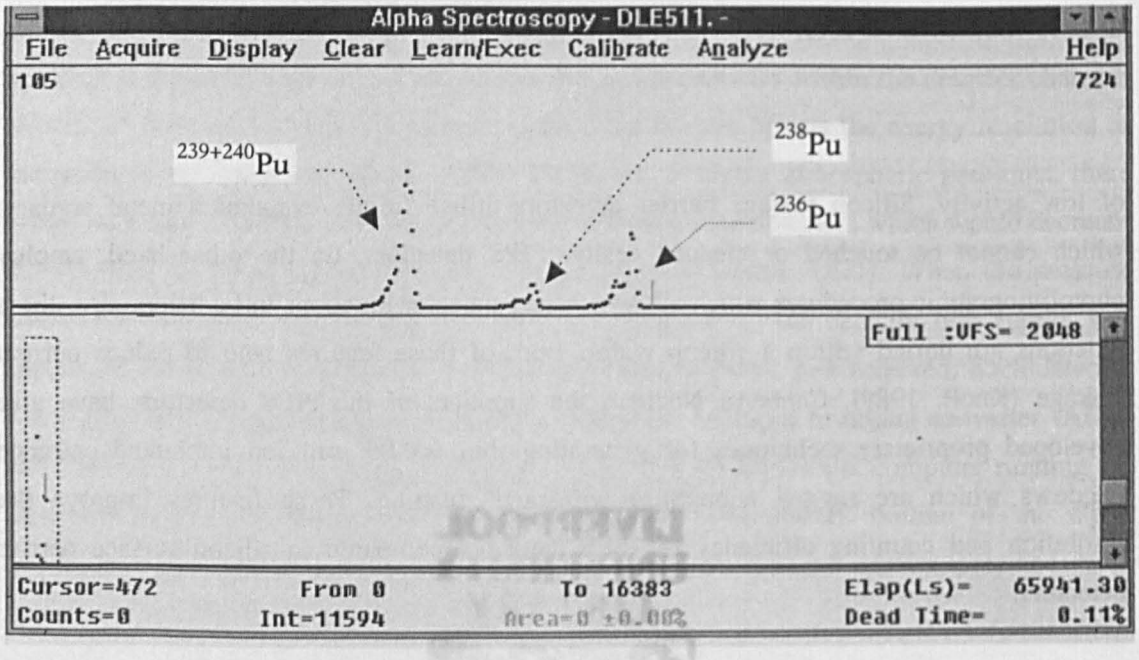


Figure 2.4: Typical Plutonium spectrum obtained using the Canberra Quad Alpha Spectroscopy System.



decontamination. In addition, routine measurement of the background in each detector chamber was carried out every three months (section 2.2.4). The detector chambers were also wiped to remove any contamination. Most of the contamination within the chambers arises from recoil of some of the sample from the stainless steel disc. Recoil may occur when helium atoms are released during an alpha decay event and as the helium atom passes through the sample, it may eject other atoms of the nuclide in the sample, pushing them off the disc and on to the detector or chamber casing.

When an alpha particle passes through the entrance window on a PIPS detector, its energy is absorbed within the detector. During passage, the charged alpha particle causes the formation of many electron-hole pairs along the track of the particle. The number of electron-hole pairs produced is proportional to the energy of the incident charged particle. The charge of the electron-hole pairs resulting from the interaction of the alpha particle with the detector is then converted into a single voltage pulse such that the height of the amplified pulse is proportional to the energy of the incident alpha particle. These amplified pulses are then processed through an analogue to digital converter (ADC) where the digital signal is again proportional to the emission energy of the alpha particle. These pulses are then sorted into discrete energy channels by a multi-channel analyser (MCA) which finally produces an energy spectrum of the alpha particles detected (Figures 2.3 and 2.4).

2.2.3 Detector Calibration

The alpha detectors required regular energy and efficiency calibration. Every three months, energy calibrations were carried out using an electroplated disc of mixed radionuclide content (^{233}U, ^{239}Pu, ^{238}Pu, ^{244}Cm and ^{252}Cf) supplied by AEA Harwell Fuel Services. These radionuclides covered a range of energies from 4.82 MeV (^{233}U) to 6.12 MeV (^{252}Cf) and therefore included the section of the energy spectrum for americium and plutonium, the nuclides of interest in this study. Energy calibration related the MCA channel number to a specific energy of the incident alpha particle. Calibrations were counted for 1,000 seconds.

Efficiency calibrations were performed at the same time as the energy calibrations, using source discs of plutonium of known activity. These discs were prepared by electrodeposition on to a stainless steel disc of equivalent surface area to the discs used for the samples (12 mm in diameter). The plutonium source discs were calibrated against certified standards as reported by Murdock (1992). The PIPS detector specification has a claimed counting efficiency that is constant with energy for the region 4.0 MeV to 6.5 MeV. This was verified using the multi-nuclide alpha source from AEA Harwell (Murdock, 1992). Overall there was negligible variation between the eight alpha detectors, with the mean efficiency being 20.57 ± 1.66% (2σ error). The average efficiency measurements of the individual detectors over time

Table 2.1: **Mean efficiency (± two standard deviations) for the QUAD-ALPHA spectrometers over the course of this study, n=9.**

Detector	*Mean efficiency (%)*
1	21.27 ± 0.48
2	19.32 ± 0.36
3	20.50 ± 0.86
4	19.98 ± 0.44
5	20.37 ± 0.60
6	21.16 ± 0.68
7	20.89 ± 2.72
8	21.06 ± 2.06

are presented in Table 2.1. For both energy and efficiency calibrations counting for 1,000 seconds reduced the peak area uncertainty to less than 2% (2 σ).

2.2.4 Background Levels

Background signals within an alpha spectrometer are almost exclusively related to contamination of the detector chambers with active source material since alpha particles can only travel short distances. As with all environmental measurements, an assessment of the background level of radiation needs to be made in order to facilitate the analysis of samples of low activity. Results of the background counts during the course of the study for each detector are presented in Table 2.2.

Background counts were acquired, using blank stainless steel discs counted for 500,000 seconds (i.e. >5 days). For the majority of samples the background signal for ^{236}Pu, $^{239+240}Pu$ and ^{243}Am was <1% of the net count rate of the sample. For ^{238}Pu and ^{241}Am it was closer to 5 and 15% respectively. These higher values resulted partly from contamination and partly because those samples with the highest background to net count signal resulted from samples of very low activity which were counted for considerable time periods (in excess of 500,000

Table 2.2: **Mean background rate (± two standard deviations) for the QUAD-ALPHA spectrometers over the course of this study, n=9.**

Detector	*Mean Background Count Rate (counts per 100,000 seconds)*				
	^{236}Pu	^{238}Pu	$^{239+240}Pu$	^{241}Am	^{243}Am
1	7.70 ± 2.72	15.3 ± 3.37	7.54 ± 7.43	10.6 ± 4.10	13.6 ± 4.09
2	2.29 ± 0.932	6.14 ± 3.42	3.24 ± 2.78	4.07 ± 1.90	6.54 ± 3.10
3	2.63 ± 2.26	5.93 ± 3.08	3.33 ± 3.67	3.77 ± 2.58	6.97 ± 3.86
4	2.47 ± 1.37	9.32 ± 3.97	3.28 ± 4.82	6.15 ± 4.48	9.46 ± 4.04
5	1.88 ± 1.27	5.82 ± 2.83	2.50 ± 3.25	3.07 ± 1.79	6.37 ± 3.56
6	1.58 ± 1.07	7.44 ± 3.425	2.52 ± 2.46	3.65 ± 2.59	7.80 ± 3.71
7	1.58 ± 1.42	7.16 ± 4.08	3.39 ± 4.13	3.61 ± 1.80	7.42 ± 4.26
8	2.43 ± 2.06	7.11 ± 3.41	3.46 ± 3.92	3.76 ± 2.80	7.82 ± 4.03

seconds). If the high background signal was attributable to contamination the detectors were decontaminated as described in section 2.2.2.

2.2.5 Data Analysis and Activity Calculations

Using the Canberra S100 acquisition software, samples were counted for fixed periods of time. Once spectrum acquisition ceased, each sample's spectrum was saved and then analysed. To analyse the spectra, predetermined regions of interest were defined around the peaks. The gross area of each peak was calculated and reported (Table 2.3). With the calibration and background information also recorded, the activity of each of the isotopes of interest within a sample was calculated using the following equation:

$$\text{Activity (A) in Bq kg}^{-1}\text{ or Bq g}^{-1} = \frac{G - B}{DE * R * Mass * LT}$$

where G = gross counts in either peak
B = background counts in either peak
DE = fractional detector counting efficiency
R = fractional recovery of tracer standard following chemical separation
Mass = mass of the sample in kg or g
LT = live time in seconds

The background count in the peak (B) was determined by multiplying the live time of the sample count by the relevant background count rate (cps) as given in Table 2.2. The recovery (R) of the tracer solution added at the start of the chemical separation was determined from the following equation for the peaks of ^{236}Pu and ^{243}Am:

$$\text{Recovery (R)} = \frac{G - B}{DE * LT * At}$$

where At = activity of the tracer spike added

These equations were used to calculate the activity of the isotopes of the nuclide of interest in either Bq kg^{-1} or Bq g^{-1}. The counting uncertainty for each isotope was estimated from $\sqrt{N}$ where N represents the number of counts in the net area of the peak (i.e. after the background counts had been subtracted). As the value of N increases, the corresponding uncertainty decreases. This method of calculating sample activities and uncertainties has been described elsewhere (Cross and Hooper, 1987; Murdock, 1992).

Table 2.3: **Example of a Canberra Series 100 software report for a Plutonium spectrum. In the region of interest summary, 1 = $^{239+240}$Pu, 2 = ^{238}Pu and 3 = ^{236}Pu.**

MCA #1 - Canberra S100 - DVA42SF2 -

Tag Number	671	Readout:	Thu 26 Oct 1995 @ 11:38:39
Report Group:	2/16	Acquire Started:	Mon 23 Oct 1995 @ 17:20:53
Input: 2 Group: A			
Group Size:	1024		

Elapsed Live Time:	237853.62 sec.
Elapsed True Time:	237932.76 sec.
Dead Time:	0.03%

ADC:	
Detector:	Canberra alpha spec 7404
Geometry:	10 mm detector-sample dist
Sample Description:	12 mm diam deposit ED disc

MCA Mode:	PHA+ M/R, Inputs: 16, Type: 1520, Mode: Route
Preset Conditions:	Live Time = 1000000 sec.

Area Statistics: Background Channels = 4
% Error Sigma = 1.65

REGION OF INTEREST REPORT

ROI	#	From(Ch) To (Ch)	Integral Area CPS	% Error	Peak(Ch) FWHM(Ch)
	1	310	13321	13205	354.50
		371	0.06	1.48	9.86
	2	377	3050	2836	410.68
		425	0.01	3.65	11.07
	3	430	26423	25753	455.83
		476	0.11	1.09	10.56

The majority of soil and vegetation samples within the study were analysed for plutonium only by alpha spectroscopy, typically with count times in the region of 20,000 seconds for soils (i.e. 5 hours) and 85,000 seconds for vegetation (i.e. 24 hours) to yield an uncertainty of <5% (2 sigma error). A number of invertebrate and mammal samples required the determination of ^{241}Am by alpha spectroscopy. Typically the count times for these samples ranged from 85,000 to 500,000 seconds (24 to 120 hours) and frequently the uncertainties determined were in the order of 20% (2 sigma error).

A number of these invertebrate and small mammal samples contained very little americium or plutonium and there was concern over the validity of these results. Consequently, the results were compared to calculated limit of detection (LOD) values. In this case, the LOD is defined as '*the smallest amount of sample activity that will yield a net count sufficiently large so as to imply its presence.....The size of this* [limit] *is governed by a pre-selected statistical risk, the probability of concluding falsely that sample activity is present*' (Currie, 1968; Pasternack and Harley, 1971). The equation employed is derived from Pasternack and Harley (1971) and is as follows:

$$LOD = \frac{2\sqrt{2}\,k * S_b}{DE * R}$$

where $2\sqrt{2}\,k$ = constant related to degree of confidence based on the normal distribution for concluding whether the measured activity is true or false

S_b = standard deviation of the background

DE = fractional detector counting efficiency

R = fractional recovery of tracer standard following chemical separation

Using 95% confidence levels, the value of k is listed as being 1.645 and therefore the value of $2\sqrt{2}$ k equates to 4.66 (Chieco *et al.*, 1992). This is the figure adopted in the present study such that the LOD, as defined here, corresponds to the quantity of detected activity at which there is a 5% probability of that apparent quantity arising from fluctuations in the background count rate.

As an example, Table 2.4 lists calculated LOD values in mBq for each of ^{238}Pu, $^{239+240}$Pu and ^{241}Am at three different live times. These live times were chosen to be representative of the actual sample count times required by invertebrate and small mammal samples in this project. The S_b of the background was determined by counting stainless steel discs as described in section 2.2.4. The number of counts which were attributable to the background levels within the detector chamber were then resolved for each radionuclides' region of interest. The

standard deviation of the background was estimated from $\sqrt{N}$, where N was the number of counts measured. It should be noted that for all samples the methods used attempted to achieve the best analytical accuracy but there was a conflict between the analytical equipment/counting time available and the attainable analytical precision (described in more detail in section 2.3.8). Moreover, the variation inherent in biological systems necessitated replication and therefore it was unfortunate but unavoidable that higher analytical uncertainties had to be accepted on occasions. Where samples were less than the LOD value, this is indicated in the text and the calculated LOD value is reported.

No results reported in this study for $^{239+240}$Pu and ^{241}Am fell below the LOD and only a few of the ^{238}Pu results did so. Principally, this is because of the location of the ^{238}Pu peak within the spectrum. The energy of alpha particle emitted by ^{238}Pu is 5,500 keV and is close to the energy of the ^{243}Am peak (5,280 keV). Consequently, there is some overlap between these two peaks with a corresponding contribution of the ^{243}Am tracer to the background of the ^{238}Pu peak.

Table 2.4: Examples of the Limit of Detection calculated for ^{238}Pu, $^{239+240}$Pu and ^{241}Am at three different live times.

	Detector Number							
LOD	*1*	*2*	*3*	*4*	*5*	*6*	*7*	*8*
^{238}Pu								
100 ks	1.06	0.95	0.82	1.01	0.81	0.88	0.95	0.95
300 ks	0.61	0.55	0.47	0.58	0.47	0.51	0.55	0.55
500 ks	0.47	0.42	0.37	0.45	0.36	0.40	0.42	0.42
$^{239+240}$Pu								
100 ks	0.53	0.33	0.39	0.46	0.43	0.39	0.38	0.35
300 ks	0.31	0.19	0.22	0.27	0.25	0.22	0.22	0.20
500 ks	0.24	0.15	0.17	0.21	0.19	0.17	0.17	0.15
^{241}Am								
100 ks	0.71	0.52	0.52	0.66	0.46	0.39	0.58	0.44
300 ks	0.41	0.30	0.30	0.38	0.26	0.23	0.33	0.25
500 ks	0.32	0.23	0.23	0.30	0.21	0.18	0.26	0.20

Units are quoted in mBq at 95% confidence levels. For these examples, average recoveries of 85% and 62% were used for plutonium and americium respectively. For the actual sample LOD values, the individual recovery as determined from the internal tracer spike was used. The latest background and detector efficiencies were used.

2.3 GAMMA SPECTROSCOPY

Gamma spectrometry was used to determine the levels of ^{134}Cs, ^{137}Cs and ^{241}Am. Low activity levels of ^{134}Cs in the majority of samples resulted in the reporting of values below the limit of detection of this gamma spectroscopy system. ^{241}Am was analysed by gamma spectroscopy and, when necessary, by radiochemical separation and alpha spectroscopy (section 2.2). Sections 2.3.2 to 2.3.4 describe the decay scheme for each of the radionuclides determined by gamma spectroscopy. All the nuclide data reported in sections 2.3.2. to 2.3.4 are taken from Browne and Firestone (1986).

2.3.1 Principles of Gamma Spectroscopy

When a nuclide undergoes radioactive decay, whether it is alpha or beta decay or by other nuclear reactions, the product nucleus may be left in either its ground state or in an excited state (Friedlander *et al.*, 1981). Excited states are unstable, so as the nucleus returns to a ground state energy must be lost from the system. This loss occurs through the emission of electromagnetic radiation where the wavelength of the radiation is determined by the difference in the energies between the excited and ground states of the nucleus. This type of radiation is called gamma radiation and is characterised by the loss of energy without a change in either the mass or atomic number of the parent nucleus.

Gamma spectroscopy is based on the principle that a proportion of the gamma ray photons incident on a detector interact with atoms within the detector crystal in a process known as photoelectric absorption (Knoll, 1989; Krane, 1988). If the entire energy of the incident gamma ray photon is absorbed within the detector crystal a photoelectron of a discrete energy equal to that of the incident gamma ray minus the binding energy of the electron in its original shell is produced (Mann *et al.*, 1980). Any photoelectrons produced are collected within the crystal and the kinetic energy of the photoelectron produces a peak in the energy spectrum. This peak is proportional to the energy of the incident gamma ray because of the relationship between the photoelectron and the gamma ray. The energy from incident gamma rays on a detector may be retained in the crystal in a number of other ways.

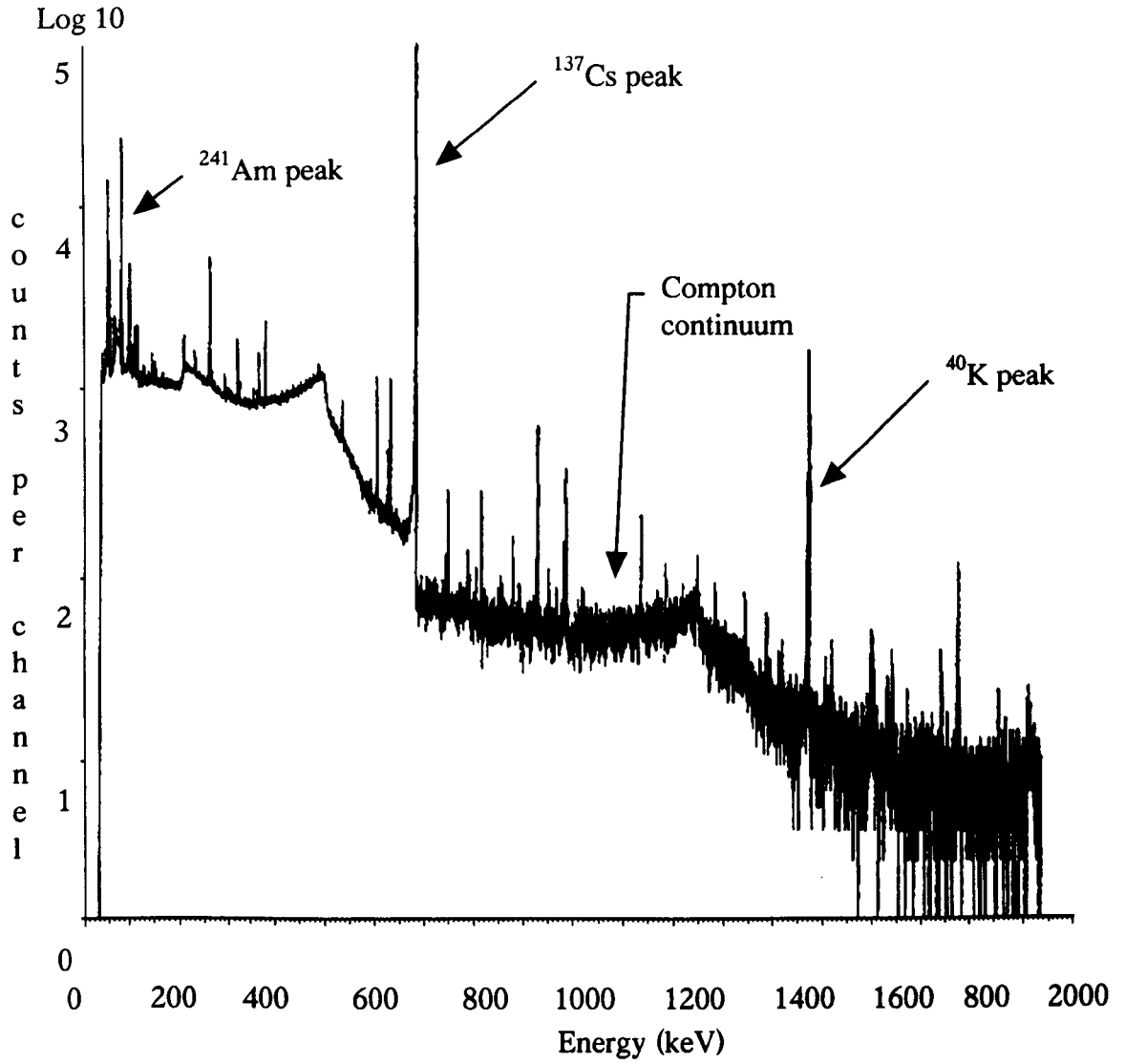
The first of these is by Compton scattering. This occurs as a result of incomplete charge collection within the detector crystal, that is to say only part of the energy of the incident gamma ray is transferred to form a photoelectron within the detector crystal. This photoelectron will have a lower energy than the incident gamma ray because of the incomplete gamma ray absorption. Incomplete charge collection can occur for a number of reasons. For example, a gamma ray may pass through the detector crystal, thus imparting only a small proportion of its energy to the detector crystal, alternatively, as the gamma ray passes

through the detector, it may be deflected and thus pass out of the crystal again resulting in incomplete charge collection (Knoll, 1989). Because this can result in a range of energy peaks in the spectrum, a continuous energy spectrum is produced. The magnitude and importance of Compton scattering is dependent to some extent upon the count rate of the sample. As an example of the large number of gamma ray/detector interactions that result in the formation of a Compton background, Figure 2.5 shows the true peaks sitting on the Compton background in a typical soil spectrum obtained during this study. A third significant gamma-ray interaction is pair production. Pair production is the process by which a positron-electron pair is created when a gamma ray photon interacts in the field of a nucleus of the absorbing material. A minimum energy of 1.02 MeV is required to make this interaction energetically viable. Any excess energy over and above 1.02 MeV manifests itself in the form of kinetic energy which dissipates over a very short distance. As the kinetic energy decreases, the positron becomes unstable and eventually annihilates with the electron. Two annihilation photons of energy 511 keV are then produced, resulting in a 511 keV peak in the energy spectrum (Knoll, 1989).

Other processes which occur during sample acquisition involve count summation. There are two forms of summation, random and true coincidence summing (TCS). Random summing is the chance of any two gamma ray photons from the same or different radionuclides being incident upon the detectors at or within the resolving time of the detectors. As a result, random summing is affected by the sample activity. As the sample activity increases, so the probability of two gamma rays being incident at the same time also increases. True coincidence summing, on the other hand, occurs when gamma ray photons emitted from the different nucleus energy levels of the same radionuclide follow one another very rapidly (usually within a few milliseconds). When this occurs there is a chance that the two gamma ray photons will incident upon the germanium crystal and be measured together. This is independent of the count rate and therefore can be a significant source of error at all sample activities. In both cases, summing results in a detected event with the combined energy of the two gamma rays (Gilmore and Hemingway, 1995). This reduces the number of counts which should be present in the true nuclide peak and will result in an overestimation of the true sample activity unless steps are taken to correct the problem.

There is little that can be done to reduce the effect of random summing with the exception of increasing the distance of the sample from the detector for samples of very high activity. Since gamma ray emission occurs in all directions, increasing the distance of the sample from the detector effectively reduces the detector area for interactions. The result is a decrease in the number of gamma ray photons incident upon the detector and hence a reduction in the chance of two incident photons. Fortunately, random summing is only really important at very high sample activities, much greater than the environmental levels found within the study.

Figure 2.5: **Typical spectrum obtained after counting a soil sample. Figure shows ^{40}K, ^{137}Cs and ^{241}Am peaks sitting on the Compton continuum.**



True coincidence summing can be corrected by using single isotope calibration sources and counting them at different heights above the detector. This would allow correction factors to be calculated for individual radionuclides. During this study, no TCS corrections were applied (section 2.3.2).

There are a variety of detector types that may be in use to determine the presence and activity of gamma-emitting radionuclides in environmental samples, all using either scintillation crystal or semiconductor technology. The main types are sodium iodide doped with thallium (NaI (Tl)) scintillators and lithium drifted germanium (Ge(Li)) or high purity germanium (HPGe) detectors. Sodium iodide detectors convert the incident gamma rays into light which is transmitted through the NaI crystal to a photocathode. These produce high counting efficiencies compared to the germanium detectors, for example a typical HPGe detector has a

Table 2.5: Gamma Detector Specifications.

	Detector A	*Detector B*
Detector Type	High purity N type germanium coaxial	High purity N type germanium coaxial
Manufacturer	EG&G Ortec	EG&G Ortec
FWHM at 1.33 MeV	<1.90 keV	<2.50 keV
Relative Efficiency	30%	20%
Arrangement	Vertical	Horizontal

2 to 3% efficiency for ^{137}Cs compared to reported values of 20% in NaI. However NaI detectors produce spectra with broad peaks due to their poor resolution. This makes the determination of multi-nuclide samples more difficult. Resolution is measured by the spread of the peak at half the maximum peak height (FWHM). The resolution of the ^{137}Cs peak at 661.6 keV for a typical NaI detector is in the order of 200 keV compared to <2.0 keV for a high purity germanium detector. For this reason, the two gamma spectroscopy detectors used here were HPGe detectors. Details of the two detectors are presented in Table 2.5.

Each detector was surrounded by 100 mm thick cylinder of lead with two inner rings of cadmium and then copper, both between 2 and 3 mm thick. These reduced the background radiation incident on the detectors (section 2.3.6). As an example, Figure 2.6 outlines the arrangement of detector A, a poptop type coaxial high purity germanium detector. The detectors were cooled with liquid nitrogen to reduce thermal induced leakage current and improve the energy resolution of the detector. This cooling to a temperature of approximately 77 °K is achieved through the use of an insulated Dewar containing a reservoir of liquid nitrogen which is kept in thermal contact via a cryostat with the detector crystal.

2.3.2 Decay Scheme of ^{134}Cs

Both ^{134}Cs and ^{137}Cs decay by beta emission (β-decay). β-decay involves the release of high energy electrons in a continuous spectrum of radiation. ^{134}Cs has a half-life of 2.062 years and decays through several energy levels (Figure 2.7) to stable ^{134}Ba. During the process, a cascade of gamma rays is emitted from each energy level. The main gamma rays are at 569.3, 604.7 and 795.9 keV (relative abundance 15.4, 97.6 and 85.4% respectively). These gamma rays were measured through the use of the germanium detectors and the analysis software calculated the activity of the ^{134}Cs in each sample, usually using the 604.7 or 795.9 keV peak. A small percentage (0.0003%) of radioactive decay events occur as a result of electron capture and result not in stable ^{134}Ba but in stable ^{134}Xe.

Figure 2.6: Schematic of vertical coaxial poptop gamma spectroscopy system.

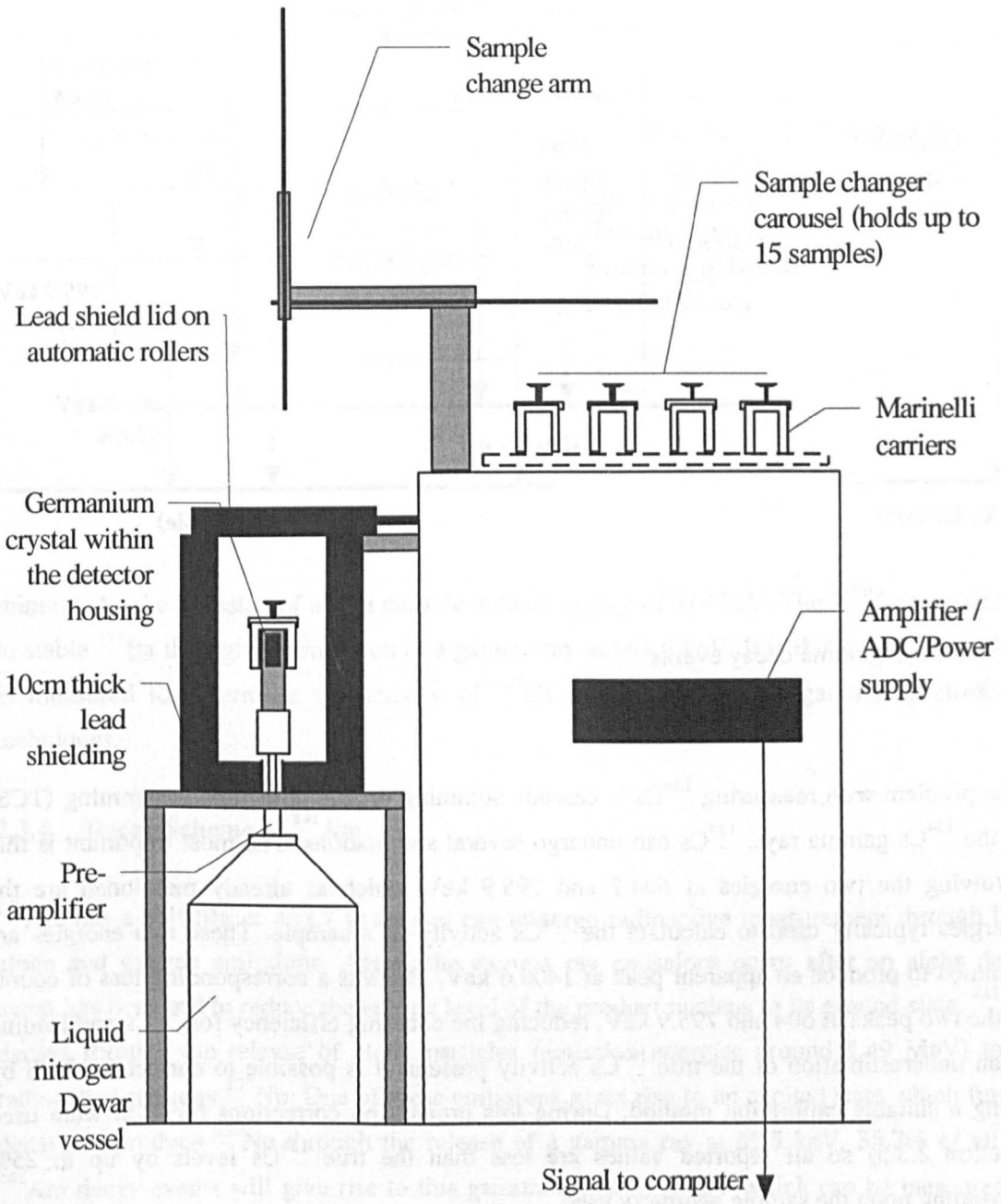
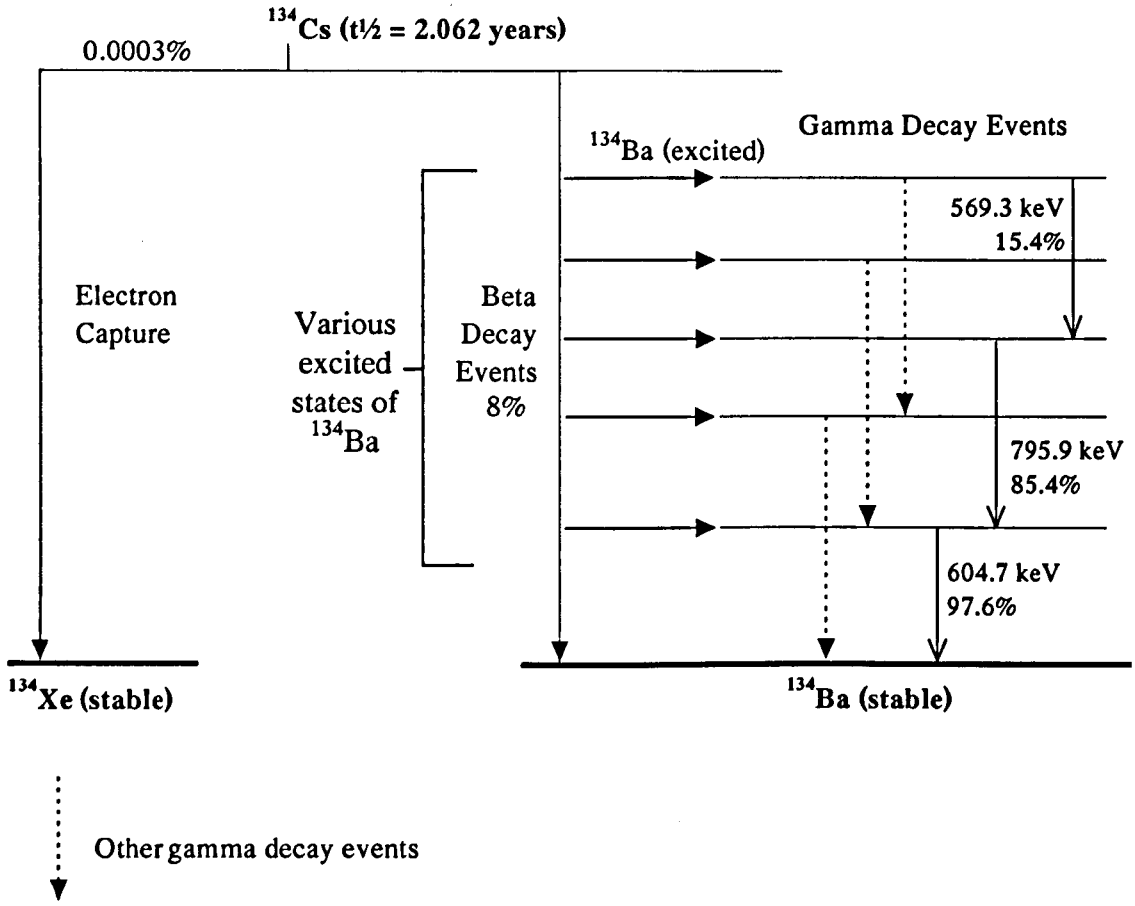


Figure 2.7: **Simplified ^{134}Cs decay scheme showing the common gamma ray decay events.**

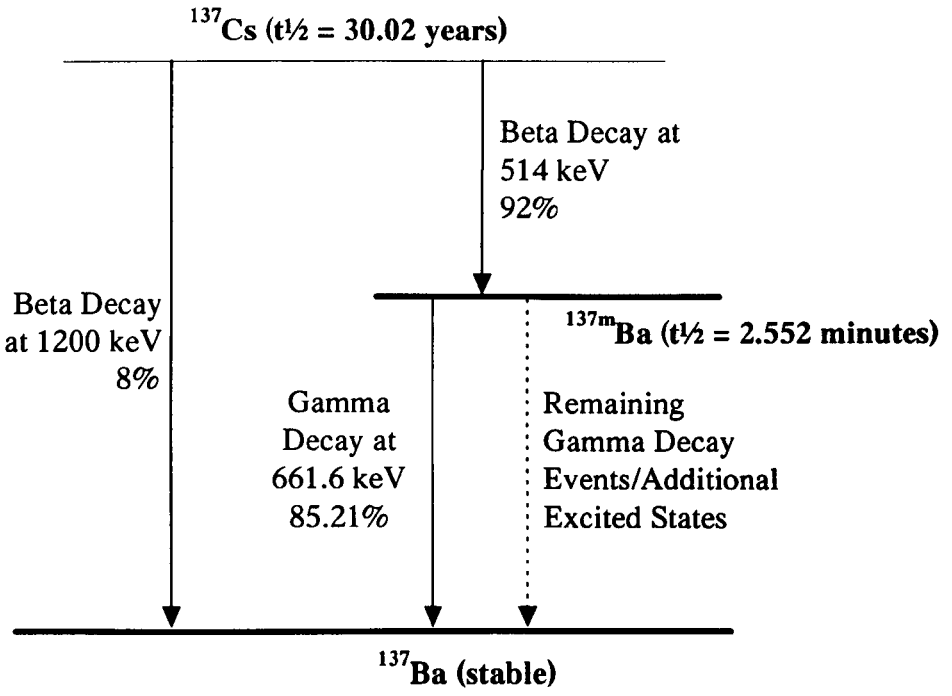


One problem with measuring ^{134}Cs is cascade summing or true coincidence summing (TCS) of the ^{134}Cs gamma rays. ^{134}Cs can undergo several summations. The most important is that involving the two energies at 604.7 and 795.9 keV which as already mentioned are the energies typically used to calculate the ^{134}Cs activity of a sample. These two energies are summed to produce an apparent peak at 1400.6 keV. There is a corresponding loss of counts in the two peaks at 604 and 795.9 keV, reducing the counting efficiency for ^{134}Cs and leading to an underestimation of the true ^{134}Cs activity present. It is possible to correct for TCS by using a suitable calibration method. During this project, no corrections for TCS were used (section 2.3.5) so all reported values are less than the true ^{134}Cs levels by up to 25% depending upon the sample geometry used.

2.3.3 Decay Scheme of ^{137}Cs

^{137}Cs has a 30 year half-life and, compared to ^{134}Cs, a simple decay scheme illustrated in Figure 2.8. ^{137}Cs decays by beta emission to ^{137}Ba either directly (8% of the decay events) or indirectly through the intermediate stage of the metastable state of ^{137}Ba (half-life 2.55

Figure 2.8: Decay scheme of ^{137}Cs.

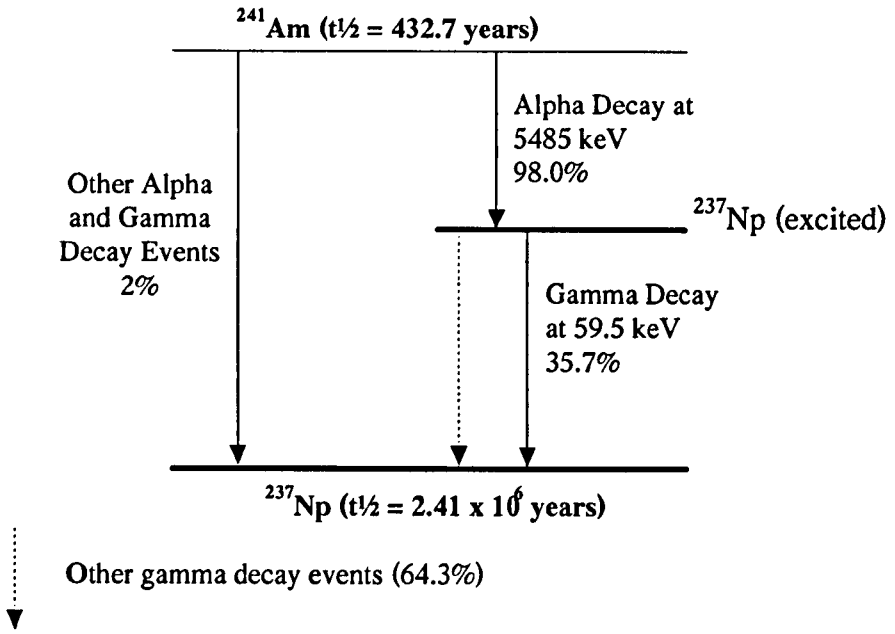


minutes) by the emission of a beta particle with an energy of 514 keV. The ^{137m}Ba then decays to stable ^{137}Ba through the emission of a gamma ray at 661.6 keV. It is this gamma ray which is measured to determine the activity of ^{137}Cs in a sample using gamma spectroscopy techniques.

2.3.4 Decay Scheme of ^{241}Am

^{241}Am has a half-life of 432.7 years and can undergo radioactive measurement through both alpha and gamma emissions. Again, the gamma ray emissions occur after an alpha decay event has occurred to reduce the energy level of the product nucleus to its ground state. ^{241}Am decays through the release of alpha particles (emission energies around 5.49 MeV) to its radioactive progeny ^{237}Np. One of these emissions gives rise to an excited state which further decays to produce ^{237}Np through the release of a gamma ray at 59.5 keV. 35.7% of all the ^{241}Am decay events will give rise to this gamma ray at 59.5 keV which can be measured by gamma spectroscopy. If ^{241}Am was not present in sufficient quantity within a sample during this study, radiochemical separation and alpha spectroscopy techniques were employed as already described (section 2.2).

Figure 2.9: Simplified decay scheme for ^{241}Am.



2.3.5 Detector Calibration and Counting Geometries

Prior to counting samples, it is necessary to calibrate the detectors for both energy and efficiency and also to assess the background levels of radiation (section 2.3.6).

2.3.5.1 Energy Calibration

Energy calibration relates the multi-channel analyser (MCA) channel number to the specific energy of an incident gamma ray photon. This is required to allow correct identification of the peaks present within a spectrum. An energy calibration was performed using a certified multi-nuclide source which was counted for 1,000 seconds (15 minutes). Table 2.6 presents the details of the radionuclides included within a typical calibration standard. There are two types of energy calibration, firstly a two point calibration for the MCA Maestro software. For this, the ^{241}Am (59.5 keV) and ^{60}Co (1332.6 keV) peaks were routinely used. This ensured that within the MCA software the channel number related to the correct gamma ray energy. Secondly, a full energy calibration was performed using the calibration program supplied with the Omnigam software. This applied a polynomial fit to the calibration data and used multiple radionuclides to ensure a more accurate energy calibration was obtained.

The energy calibration on detector B was found to be stable over time but for detector A there were significant energy shifts which fluctuated with changes in room temperature. To counter

Table 2.6: Details of the radionuclides contained within the multi-nuclide clay standard.

Nuclide	*Half-Life (days)*	*Activity (Bq)*	*Energy (keV)*	*Branching Ratio*	*Overall Uncertainty (%)*
^{241}Am	158043.7 ± 182.6	4695.3	59.5	35.7	3.9
^{109}Cd	462.6 ± 0.4	32079.0	88.0	3.6	3.8
^{57}Co	271.8 ± 0.1	1198.8	122.0, 136.0	85.5, 10.7	3.7
^{123m}Te	119.7 ± 0.1	1605.8	159.0	84.0	3.8
^{113}Sn	115.1 ± 0.1	5809.0	392.0	64.0	3.3
^{85}Sr	64.8 ± 0.02	6959.7	514.0	99.3	3.2
^{137}Cs	10957.5 ± 73.1	4817.4	661.6	85.2	3.4
^{54}Mn	321.2 ± 0.1	4769.3	835.0	99.9	3.8
^{88}Y	106.6 ± 0.02	11581.0	898.0, 1836.0	92.7, 99.4	3.3
^{60}Co	1925.2 ± 0.4	6008.8	1173.0, 1332.0	99.9, 99.9	3.3

Data taken from Certificate of Calibration for a Clay standard. Uncertainty on the data <2%.
Overall uncertainty is reported as 2 σ.

this, a range of energy calibrations was maintained and the appropriate energy calibration selected based upon the channel number for the ^{137}Cs peak for individual samples at the time of analysis.

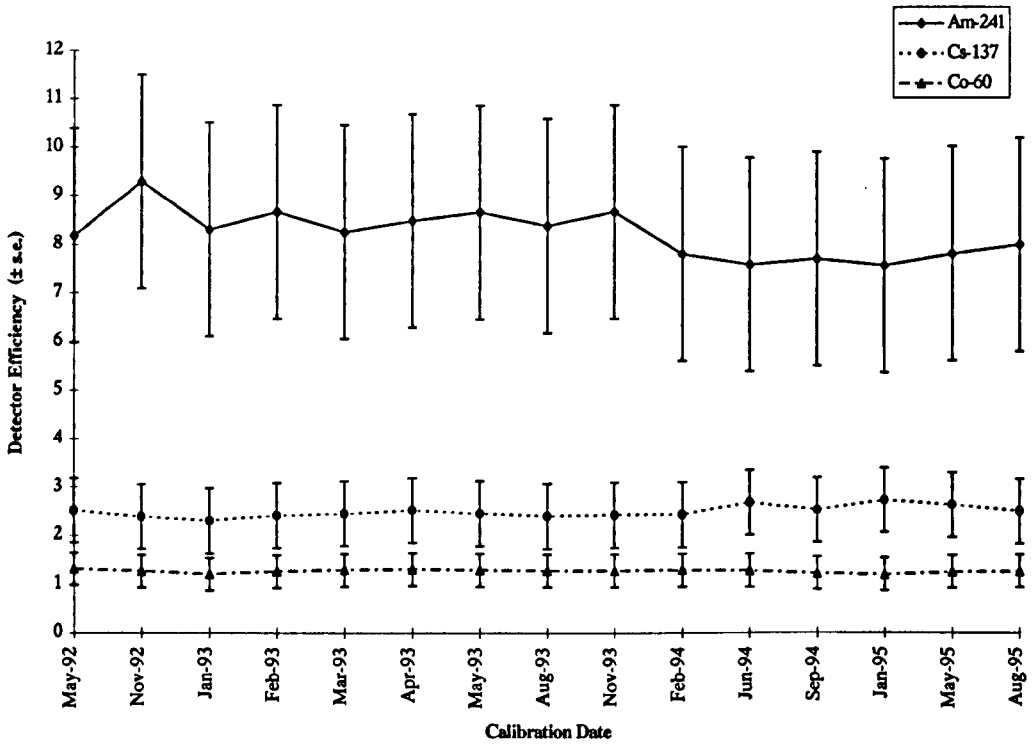
2.3.5.2 Efficiency Calibrations

Efficiency calibrations consist of two components. Firstly, that related to the proportion of counts registered per gamma ray incident on the detector. This is related to the emission energy of the gamma ray. Secondly, a component related to the geometry of the sample which measures the proportion of gamma rays incident on the detector from each radioactive decay event within the sample matrix.

Efficiency calibrations were performed by counting samples of known radionuclide content and activity and with a fixed and standardised geometry on each detector. Count times for the standards were originally 60,000 seconds (i.e. 17 hours). However, as some of the radionuclides decayed, the count time was increased to 85,000 seconds. Efficiency calibrations were carried out at three month intervals as it was shown that there was little variation in the detection efficiency over time (Figure 2.10).

A typical calibration curve is presented in Figure 2.11. At low energies (<30 keV), the percentage of gamma rays incident on the detector is low because of self-absorption within the sample matrix or detector casing. As the gamma energy increases so the percentage of gamma rays incident on the detector increases until the knee energy is reached (around 100 keV). Above this point, it can be seen that the efficiency declines with increasing energy

Figure 2.10: Detector efficiency (%) variation over time.



because the probability of complete photoelectric absorption within the detector crystal decreases and therefore fewer photons contribute to the observed photopeak. Consequently, for each sample geometry it was necessary to obtain a separate efficiency calibration curve because differences in the sample matrix affect the amount of self-absorption of the gamma rays and also the probability of gamma rays being incident upon the germanium crystal. The variation in detector efficiency with sample matrices has been reported by Rudge (1989). Since the detector efficiency varied with energy, and because it was used in the calculation of sample activities, it was critical that accurate calibrations be performed.

2.3.5.3 Counting Geometries and In-House Standards

For all gamma spectroscopy work steps were taken to optimise the counting efficiency and background levels in order to minimise the counting times required. Two counting geometries were chosen based upon typical sample masses collected in the field. Marinelli beakers were used to hold samples of soil, sediment and vegetation because the larger volume and surface area of the beakers allowed more sample to be exposed to the detector. For samples such as the invertebrates and small mammals, where small mass (typically <5 to 10 g) was common, a planar geometry was used.

Figure 2.11: Typical efficiency against energy calibration curve (taken from a soil standard).

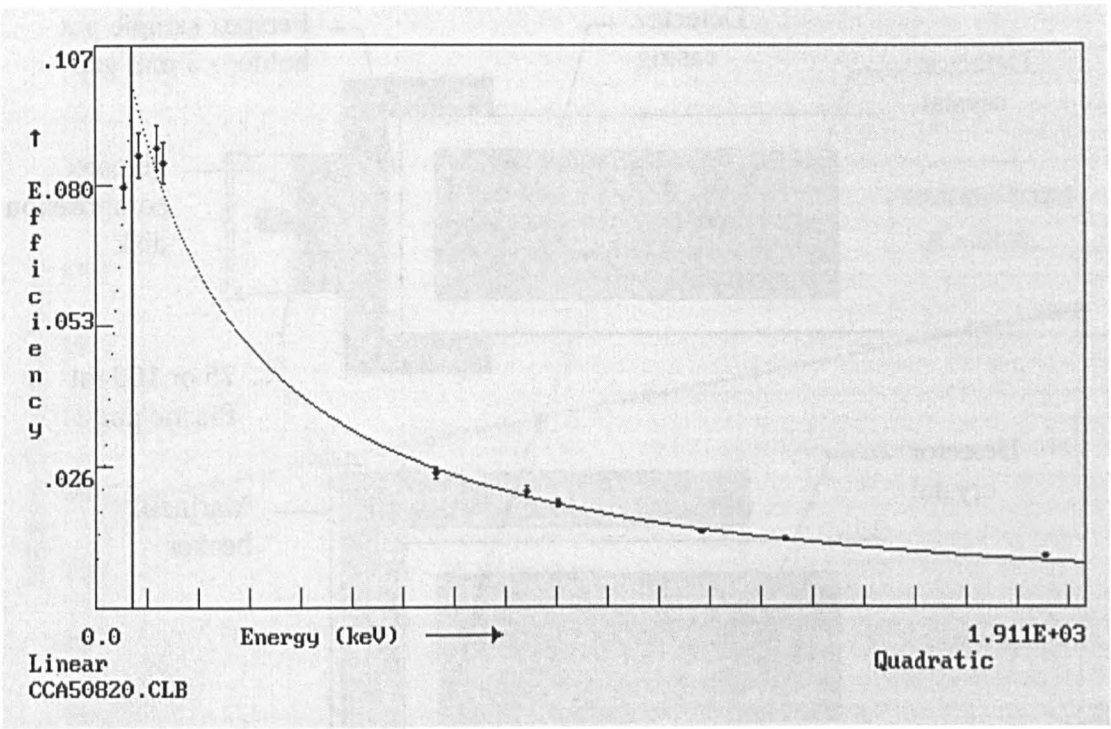
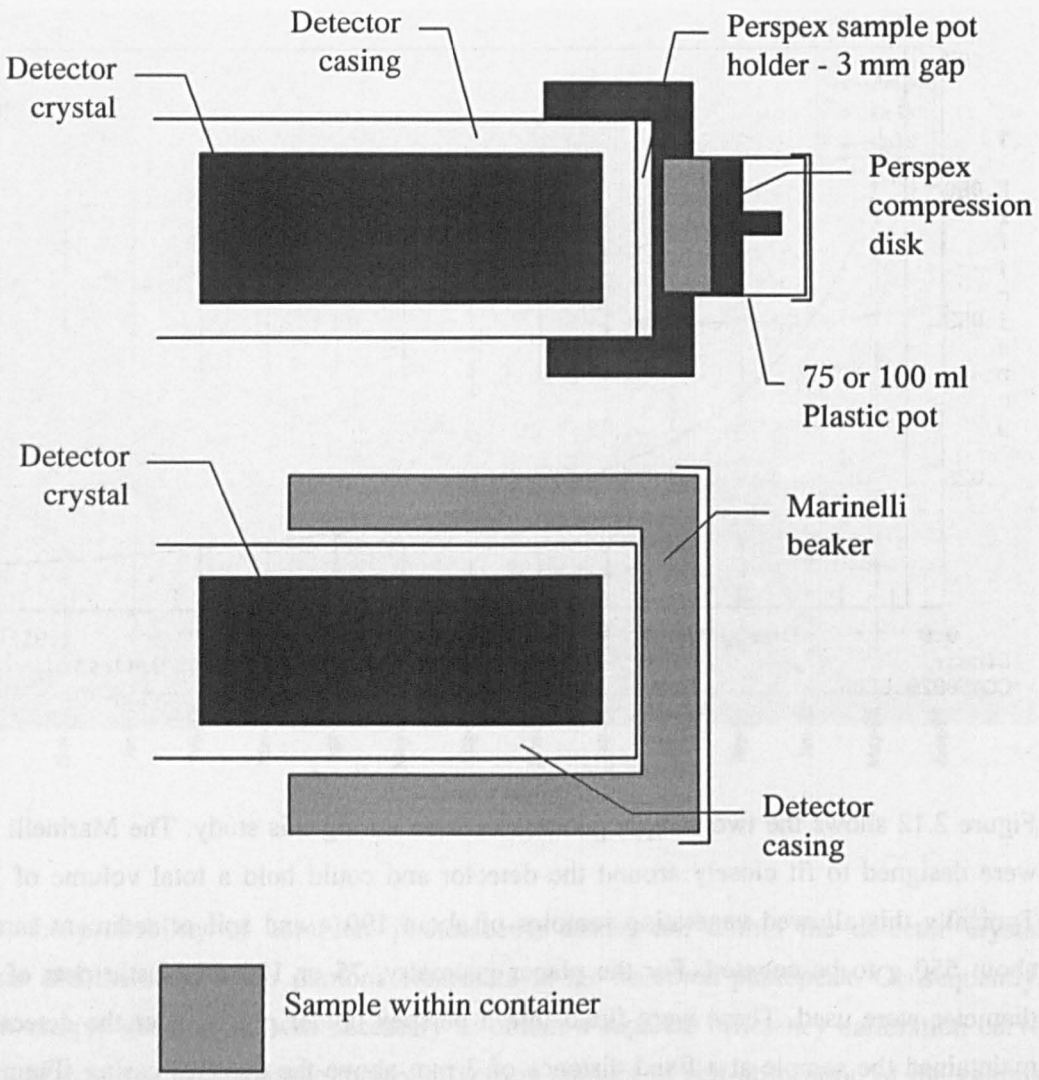


Figure 2.12 shows the two sample geometries used during this study. The Marinelli beakers were designed to fit closely around the detector and could hold a total volume of 330 ml. Typically this allowed vegetation samples of about 100 g and soil or sediment samples of about 550 g to be counted. For the planar geometry, 75 or 100 ml plastic pots of 49 mm diameter were used. These were fitted into a perspex holder placed over the detector. This maintained the sample at a fixed distance of 3 mm above the detector casing (Figure 2.12). The use of the holder and the compression of the sample within the pot using perspex discs ensured that standardised conditions were maintained and allowed accurate sample depth measurements to be made.

With the planar geometry samples, even after compression, there were differences in the depth of the sample. Several authors have indicated the need to correct for even small differences in the geometry of samples on different types of gamma detectors (Appleby *et al.*, 1992; Gilmore and Hemingway, 1995). Correction factors were therefore determined to relate sample depth to a standard of fixed depth (5 mm) so as to calculate the efficiency of detection for samples of different masses. Figure 2.13 shows the correction factors determined using a multi-nuclide liquid source. The source was added to standard 75 ml pots of constant diameter (49 mm) which were filled with a range of depths from 2 to 10 mm and then counted on the detectors as appropriate. The detection efficiency for each sample was then determined using the Omnigam software routines. The detector efficiency at each sample depth is expressed as a correction factor relative to the 5 mm standard used for regular detector calibration as

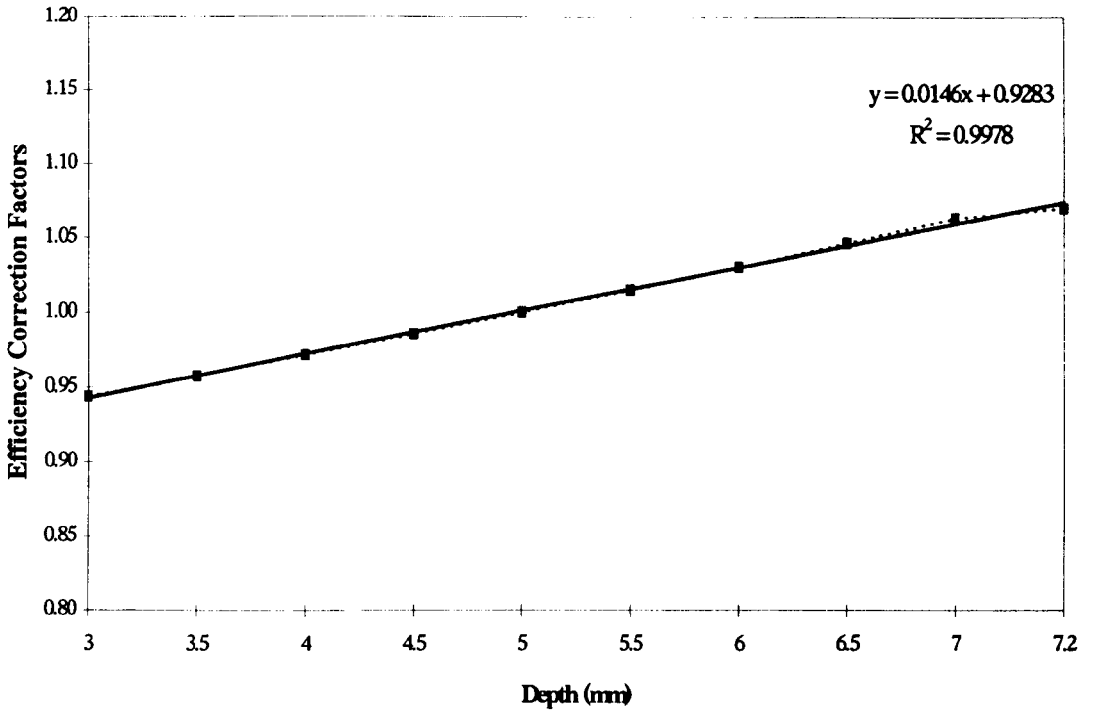
Figure 2.12: Sample geometries used for gamma spectroscopy.

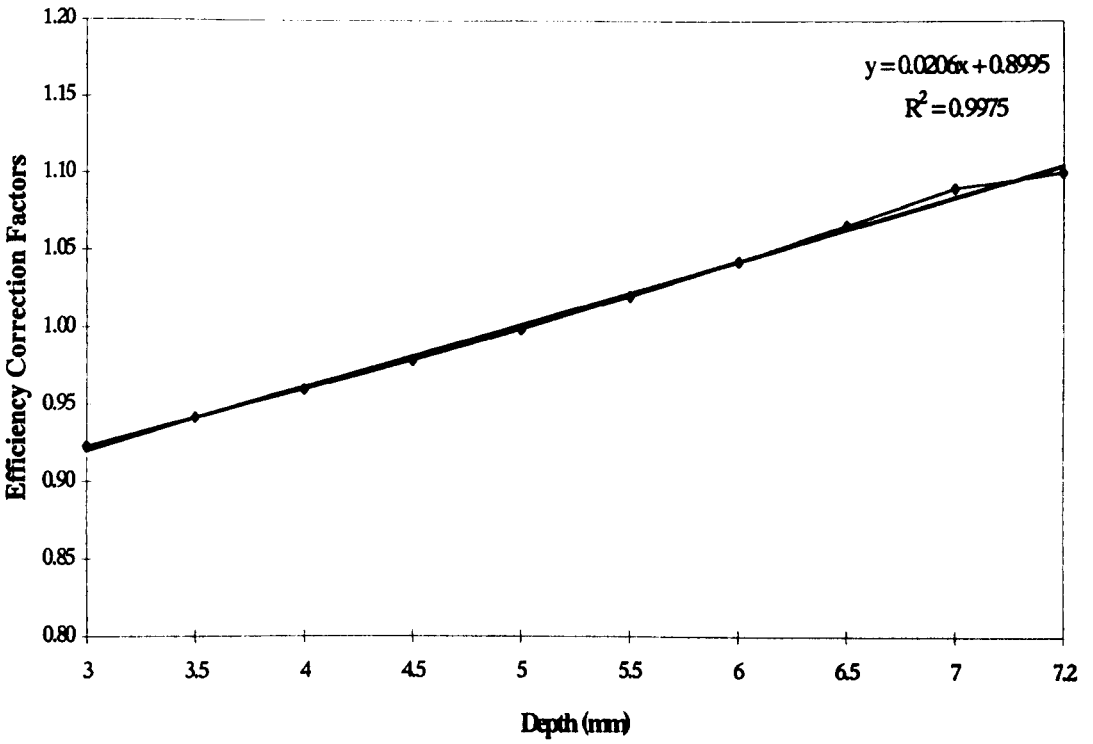


outlined previously. Calculating correction factors reduced the need to calibrate a wide range of samples at different depths which is both expensive and time consuming. The calculated correction factors were small for ^{241}Am and had a negligible effect on ^{137}Cs values. Consequently, depth corrections were applied only to ^{241}Am values where it was measured by gamma spectroscopy. During these calculations, it was assumed that the sample density remained constant. Those radionuclides with low emission energies, such as ^{241}Am, are affected most by increasing the sample depth.

Energy and efficiency calibration standards were produced in a number of matrices (clay, vegetation, invertebrate and mammal) and comprised a mixed gamma radionuclide source. This ensured that similar matrices to the real samples were used for calibrating the detectors. Internal standards were produced by counting 'clean' samples prepared as described in sections 3.3, 4.3 and 5.3. Once the radionuclide content was known, an excess of each

Figure 2.13: **Correction factors and regression equations for ^{137}Cs and ^{241}Am respectively, as measured via gamma spectroscopy.**





radionuclide was added to each sample matrix. Each matrix was then wetted with distilled water and the sample and standard solution mixed thoroughly. The standards were then dried. The standards were re-wetted, mixed and dried to ensure homogeneous dispersal of the radionuclides through the sample matrix. Several sub-samples were then carefully removed and counted to ensure that the standards were homogenised. If necessary, further mixing was performed. Once finished, the standards were packed to a specified depth and sealed prior to routine use.

2.3.6 Background Levels

Unlike the background radiation found in the alpha spectrometers (section 2.2.4), gamma spectrometers detect radiation from a wide range of sources. These include both external sources and contamination from the samples used within the detector chamber. Therefore, different methods are required to reduce the background levels of gamma radiation to facilitate the counting of environmental samples.

2.3.6.1 External Gamma Radiation

Cosmic radiation and natural terrestrial radiation present in the environment form a continuous background signal to all gamma radiation detectors. The magnitude of the signal depends upon the location of the radiometric laboratory, detector types and size and the degree of shielding placed around the detector crystal. Some of the natural radionuclides contributing to this type of background radiation are ubiquitous within the environment and are often incorporated, albeit at very low levels, within the materials used to manufacture both the detector casing and the shielding which surrounds it. Surrounding the detector with lead shielding as shown in Figure 2.6 can effectively reduce the background signal from natural radiation. Lead is a common shielding material, being of high atomic number it is very effective at stopping the passage of gamma rays. The lead shielding is often supplemented, as in this study, through the use of a cadmium and then copper layer. Both these layers are 2 to 3 mm thick and form a graded shield. Graded shields are employed because x-rays are produced by the irradiation of the lead by both the sample and external background radiation. The cadmium and copper layers absorb these lead x-rays, emitting only low energy or weakly penetrating x-rays themselves. In addition to sources of natural radiation, anthropogenic radionuclides are present in the environment, largely as a result of the weapons testing in the 1950s and 1960s. These contribute to the background signal in the same way as natural radiation. The problem is that these 'man-made' radionuclides are often the same as those being measured in the samples. This emphasises the importance of accurate background measurements and the use of suitable detector shielding.

Background measurements were taken at three month intervals with an unused Marinelli beaker counted for 220,000 seconds (i.e. 60 hours). When the planar geometry samples were introduced, background measurements were also made using the plastic pots. Comparing the results from the two sample containers, it was clear that the different material in the containers had an insignificant effect ($p>0.05$) upon the background measurements. Table 2.7 shows a typical background reading on detector B.

Table 2.7: Typical background table used by the Omnigam software for peak background correction on Detector B.

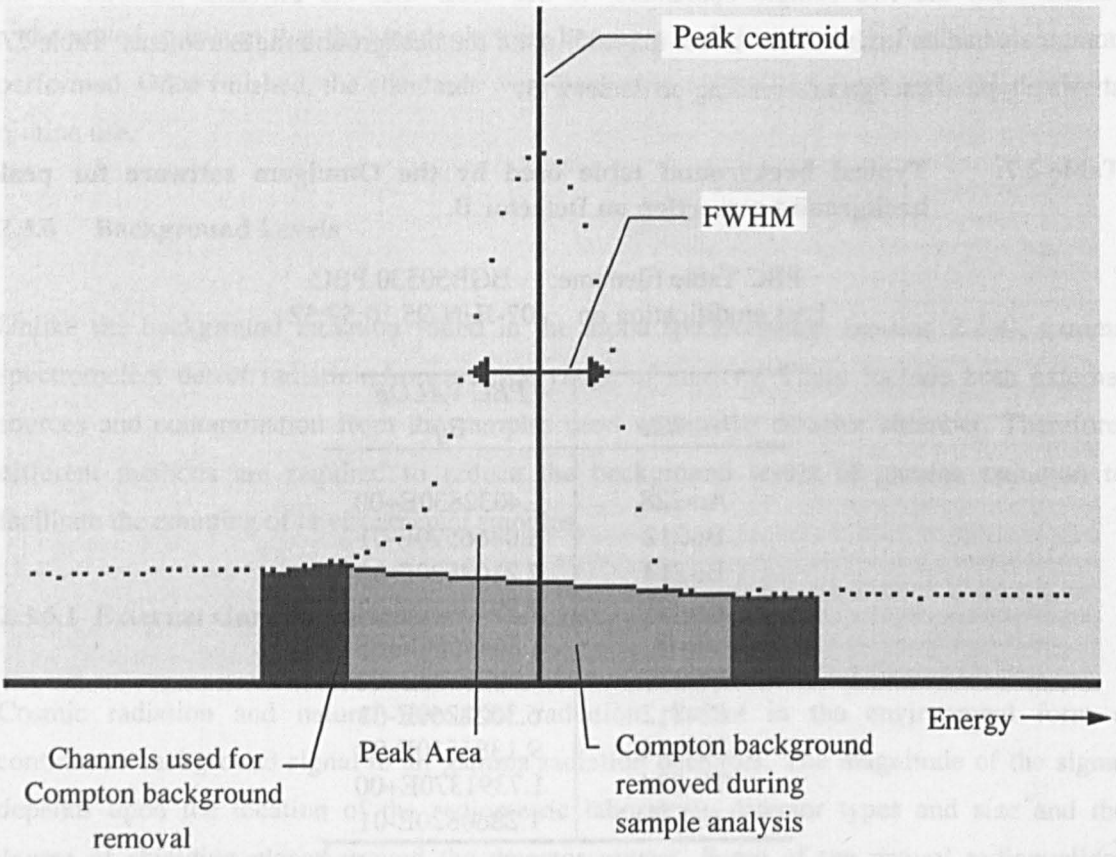
PBC Table filename: BGB50530.PBC
Last modification on 07-JUN-95 10:57:52

Nuclide	*PBC VALUE Bq*
Ac-228	1.4032830E+00
Bi-212	3.0466570E-01
Bi-214	7.2103520E-02
Cs-137	7.9913420E-02
K-40	1.8644660E+00
Pb-210	4.9076130E+00
Pb-212	6.3022260E-01
Pb-214	9.1386510E-02
Th-234	1.7391370E+00
U-235	1.2880820E-01

2.3.7 Data Analysis and Activity Calculations

Samples were counted on either of the two coaxial germanium detectors and saved to disk using the EG&G Ortec Maestro software. The spectra were then interrogated and analysed using EG&G Ortec Omnigam software. Typically this involved locating peaks and recording information concerning the peak area, channel centroid and FWHM (Figure 2.14) in a logbook. This was carried out by locating suitable peaks. Typically a sensitivity level which accepted half of the peaks was used. This selected the best shaped or largest peaks within the spectrum. The peaks chosen fitted a Gaussian shape or a Gaussian shape with an exponential component to account for any low energy tail caused by incomplete charge collection within the detector. The chosen peaks were then fitted with a region of interest as determined by the software using a best fit method. Typically the software selected a set number of channels on either side of the peak centroid based upon the FWHM values. However, additional calculations were required when two peaks were in close proximity. In these cases the peaks were deconvoluted, that is the software separated them using in-built mathematical routines.

Figure 2.14: Stylised Spectrum Peak.



Having set regions of interest, automatic routines within the Omnigam software were used to calculate and subtract the Compton background from the peaks. The software selected an optimum number of channels on either side of the region of interest and then assessed the Compton background component before its subtraction. Essentially the software uses the channels on either side of the region of interest to draw a line under the peak. Those counts below this line are then subtracted from the peak area, leaving the true sample counts and the contribution of peak background counts (caused by contamination within the detector chamber). Figure 2.14 shows a stylised peak and its component background parts.

The specific activity and uncertainty of each radionuclide were then calculated using the following equation:

$$\text{Activity (A)} = \frac{G - B}{DE * RA * Mass * LT}$$

where

	(A)	=	activity of the sample in Bq/kg or Bq/g
	G	=	gross counts in peak
	B	=	background counts in peak (true peak background counts)
	DE	=	fractional detector counting efficiency
	RA	=	relative abundance of a gamma ray at a specific energy (keV) as a fraction
	Mass	=	mass of the sample in kg or g
	LT	=	live time in seconds

The background count in the peak (B) was determined by multiplying the live time of the sample count by the relevant background count rate (cps) as determined from a background count. Peak background counts were determined by counting empty Marinelli beakers. Data were analysed using the normal software programmes and then by running an additional routine which determined the count rate of individual radionuclides before storing the information in a background file for use during sample analysis (Table 2.7).

Radioactive decay is essentially a random process so the uncertainty of the decay rate as one standard deviation can be estimated by the function of $\sqrt{N}$ where N represents the net number of counts in a peak. As count time increases, so the number of net counts increases and the counting uncertainty decreases. However, as count times increase so the background count rate from both the Compton scattering and the peak background count will increase. Subtracting the Compton and true peak backgrounds from a peak will inevitably lead to an increase in the uncertainty in the sample activity. This uncertainty is relative to the activity of the sample and consequently is greater for samples of low activity. Therefore this requires an accurate background assessment. Since a large proportion of the samples analysed during this project were of low activity, particularly the invertebrate and small mammal samples, background counts were averaged over the course of a year and applied manually to samples of low activity. This provided a more accurate background count but, more importantly, improved the counting uncertainty associated with the background count, thus reducing the overall sample uncertainty. Table 2.8 provides information regarding count times and typical counting uncertainties for different sample matrices. Where necessary, limit of detection values were determined using the Omnigam software routines provided.

Table 2.8: Typical count times and uncertainties for different sample matrices.

Sample Matrix	*Count Time (seconds)*	*Uncertainty (2σ)*
Soil	<15,000	<2%
Vegetation	85,000 - 250,000	<20%
Leaf Litter	<50,000	<10%
Invertebrates	85,000 - 400,000	<30%
Small Mammals	>250,000	<30%

Counting uncertainties reported as 2 sigma (95%) levels.

2.4 REPLICATION AND STATISTICAL METHODS

Table 2.8 indicates the counting statistics which were achieved for the most part during this study. However, studies of the environment also have to contend with additional problems regarding the complexity and heterogeneity of biological systems. Therefore to facilitate study of the transfer of radionuclides through natural food chains two components must be considered, firstly the counting uncertainties and secondly the biological variation inherent in ecosystems. The former is determined by the counting time allowed and the activity of the sample. The latter ideally requires measurement of large numbers of replicates from each component of the food chain in order to assess the biological variability within an ecosystem. Inevitably, in a study of this type and size, these were competing aspects and consequently compromise was necessary.

The present project was designed and implemented with biological variation, limited analytical time and project objectives in mind. It was essential that the sampling programme still fulfilled the requirements to observe the spatial, temporal, and biological variation within different components of the food chain under investigation across three field sites. Therefore, for each site, individual sampling regimes were devised which utilised sub-areas of each site as replicates, in order to reduce the overall number of samples. Each sub-area was selected randomly and then marked out with a systematic sampling grid which was used to produce sub-samples of the population for each food chain component. Subsequent sampling visits utilised the same area of the field site as suggested by Eberhardt (1976) for studying temporal change at a site. Furthermore, at the outset of the project, additional samples of soils and vegetation were collected and analysed to determine the spatial distribution of the radioactivity across each site. Subsequently, replicates were collected from each site on several occasions to observe temporal variation. Where necessary and required by the limitations of sample analysis, sample replicates were pooled prior to analysis and this is described as appropriate in later chapters.

In addition to the problems of sample replication and available analytical equipment, sampling small mammal and invertebrate species using the trapping methods described later, was subject to fluctuations. This was because each of the three sites were small and therefore the numbers of small mammals and invertebrates varied, for example, via natural population cycling and migration to and from adjacent areas. Consequently, the pre-determined minimum number of small mammals or mass of invertebrates required for statistical analysis was not always achieved. This has led to the need to use caution when conducting and interpreting statistical analysis of those data where replication is small.

Where sufficient replication exists, results are reported as arithmetic means with an associated measure of the dispersion of the data around the average, usually either standard deviation or standard error. On those occasions where only one sample exists, the error reported is the 95% counting uncertainty and this is indicated within the text.

One further complication when using statistical tests is the use of data sets containing 'less than' values. These are actually reported as the limit of detection (LOD) for the sample, radionuclide and detection equipment used. Inevitably, with samples of low mass or low activity, the number of LOD values within a data set increases. There are several options available for handling these LODs (after Gilbert and Kinnison, 1981):

1. Ignore LOD values and calculate all the statistics using the remaining positive detected data.

2. Replace the LOD values by zero and then complete the statistical analysis.

3. Replace the LOD values by a value between zero and the reported LOD figure and then complete the statistical analysis.

4. Use the LOD value for all statistical calculations.

Options 1 and 2 produce mean and variance values which would be overestimated and underestimated respectively, resulting in unrepresentative values for the data set. The third option assumes that all the values between zero and the LOD value are equally likely to occur and provides no easy mechanism for selecting a value to use and simply expands the final reported uncertainty. The final option, results in an overestimation of the mean and reduces the actual variation within the data set. However, this method, although not ideal, places an upper limit on the mean values and therefore is conservative at all stages. This is the method applied within the present study. Where LOD values have been applied, this is indicated in the

text and where appropriate the calculated mean values are reported as 'less than' values themselves.

Three statistical methods for comparing sample populations were employed within the present study. The first is the Student's *t* test, second the Analysis of Variance test for comparing sample populations which are normally distributed and the second is the nonparametric Mann-Whitney U test, the analogue to the two sample *t* test for distributions which are skewed (Siegel and Castellan, 1988; Sprent 1993). The Analysis of Variance (ANOVA) and Mann-Whitney U tests were performed using the Minitab statistical computer package and the results are reported where appropriate in the text. Following ANOVA calculations, a 5% Least Significant Difference (LSD) was calculated to allow comparisons of the mean of the data sets under investigation. Since samples were collected with the express intention of comparing certain data sets, the 5% LSD calculation was performed using the Bonferroni protected LSD for planned comparisons (Maxwell and Delaney, 1990). The Mann-Whitney U test was used after calculations of the skewness and kurtosis confirmed that the sample population distribution lay outside the bounds of a normal population thus requiring nonparametric methods of analysis. The skewness and kurtosis values are measurements around the population mean and the Mann-Whitney was used when the limits, β_1 and β_2, were at $p=0.05$ (Croxton, 1953; Siegel and Castellan, 1988).

Reference note: All botantical nomenclature as given in *Flora of the British Isles*. 2nd Ed. Clapham A.R., Tutin T.G. and Warburg E.F. (1962), Cambridge University Press.

Chapter Three
A CONIFEROUS WOODLAND ECOSYSTEM - LADY WOOD

3.1 INTRODUCTION

3.1.1 Background - previous studies

As mentioned previously, much of the research on terrestrial environmental radioactivity has focused on food chain pathways which culminate in man, primarily through agricultural ecosystems, and the countermeasures that may be applied to reduce any transfer (e.g. Aarkrog, 1983; Hove *et al.*, 1993; Lembrechts, 1993; Nair *et al.*, 1983; Prister *et al.*, 1993; Segal, 1993; Shaw, 1993; Summerling, 1983). However, as the Chernobyl accident in 1986 showed, the transfer of radioactivity through woodland ecosystems is also an important pathway to man primarily because woods are used to provide additional foodstuffs to supplement the diet, timber for building and fuel, and for recreation (Berg *et al.*, 1990; Desmet and Myttenaere, 1988; Kirchmann *et al.*, 1993; Melin *et al.*, 1994; Prister *et al.*, 1994; Tikhomirov *et al.*, 1993). Until recently, few studies have examined the effects and transfer of radionuclides within woodland ecosystems, particularly the relationship between plants and wild animals. It is important to assess the effects of radionuclides on the native flora and fauna not only because of their possible use by man (Grabowski *et al.*, 1994; Johanson and Bergstrom, 1994; Lindner *et al.*, 1994) but also because of the recent awareness of the possible adverse effects on ecosystems of long term behaviour and cycling of radionuclides within the forest environment (Myttenaere *et al.*, 1993). This can be considered to parallel the research undertaken for heavy metals such as cadmium, copper and zinc. Much of this literature considered the impact of the heavy metals on both soil processes (i.e. the actual rate of decomposition (Coughtrey *et al.*, 1979; Kohler *et al.*, 1995; Zwolinski, 1994) and the effects on the flora and fauna, in particular the species composition and diversity, (Curry and Good, 1992; Hughes *et al.*, 1980).

Before the Chernobyl accident, a few studies had investigated and modelled the transfer of radionuclides, particularly ^{137}Cs, from the soil through the woodland ecosystem by the addition of tracers to the soil or on already contaminated sites (Adriano *et al.*, 1981; Auerbach *et al.*, 1964; Rickard *et al.*, 1982). The results of these studies indicated the importance of the soil compartment to the accumulation of radionuclides (Auerbach, 1987). However, the Chernobyl accident focused attention on the role of forest ecosystems in the atmospheric interception, transfer and cycling of radionuclides, partly as 50 to 60% of the contaminated land around the Chernobyl nuclear power plant is under forest, and partly because the accident provided the opportunity to study the effects of a pulse release of radioactivity to the environment (Bunzl *et al.*, 1989). Since the accident several studies have investigated the interception of radionuclides by leaf canopies which have been shown to be effective

accumulators of atmospheric radioactivity (Coenen *et al.*, 1987; Sombre *et al.*, 1990a, 1990b; Tikhomirov 1990). However, even now, very little work has investigated the behaviour and movement of radionuclides through woodland food chains involving native flora and fauna.

3.1.2. Study Objective

The primary objective of this study was to describe the behaviour and transfer of radionuclides through food chains in a coniferous woodland with a continuing input of radionuclides. This was achieved through the analysis of soil, leaf litter, vegetation, invertebrate and small mammal samples collected over a period of two years. The following discussion includes a description of the spatial distribution of the radionuclides (^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am) through the woodland, and evidence for temporal variation. Transfer factors for each of the radionuclides to the invertebrate and small mammal species are presented.

3.1.3 Deposition Processes

Following the release of radionuclides to the atmospheric environment, either as a result of an accident such as Chernobyl or through the routine operations of a nuclear establishment such as BNFL, Sellafield, radioactive material can be deposited to the ground. The dispersal and deposition of these radionuclides are determined by a number of factors, including: wind direction and speed; particle size; climatic conditions (solar radiation, rainfall *etc.*); topographical features and the type of ground cover. The effects of these factors have been described more fully elsewhere (Harrison *et al.*, 1993; McKay and Pattenden, 1990; Nicholson, 1988a; Wheeler, 1987). Atmospheric radioactivity can be deposited by two principal mechanisms: precipitation scavenging (wet deposition) and dry deposition. Precipitation scavenging involves the removal of particulate matter and gases from the air by different forms of precipitation leading to the incorporation of radioactivity into rain water. Precipitation also has the ability to leach radioactive particles, deposited under dry conditions, from the surface of vegetation and other materials. Dry deposition occurs continuously within the surface boundary layer with radioactive particles being deposited through diffusion, impaction, interception and sedimentation under gravity. This deposition can be enhanced by a number of factors; for example, increased wind turbulence; electrostatic charges on the particles and surface material. Again these processes have been described in more detail elsewhere (Harrison *et al.*, 1993; Nicholson 1988a). Due to the complex nature and many interactions involved in depositional processes, a single generalised term is often used to describe radionuclide deposition. The term, deposition velocity (Vg), is defined as the deposition flux (to a unit area of land) divided by the air concentration (Harrison *et al.*, 1993).

As indicated above, the structure and nature of plant surfaces has a very strong influence upon the deposition velocity (Nicholson, 1988a). The interception of particulate material is determined by the particle size; wind speed; surface area available for deposition and other factors. In particular, coniferous forest leaf canopies provide excellent sites for effective interception of airborne particulate material to the extent that, for adjoining areas of woodland and grassland, the deposition of particulate material is greatly enhanced in the woodland (Belot *et al.*, 1994; Kirchmann *et al.*, 1993; Shaw *et al.*, 1994). As an example, deposition velocity measurements for trees have been shown to be one order of magnitude greater than those on to smooth surfaces (Tveten, 1990). This is mainly due to the increased surface area available for interception (measured as leaf area index) and to differences in the leaf surface structure. Differences in the available surface area are therefore dependent upon the tree species. For example, it has been reported that coniferous species are more effective at intercepting particulate material than deciduous species (Adriano *et al.*, 1981; Ritchie *et al.*, 1970). Primarily this is due to the greater surface area of pine needles compared to the equivalent photosynthetic surface for deciduous species thus increasing the surface area available for interception; also, being evergreen, coniferous stands are capable of intercepting material throughout the year.

3.1.4 Post Depositional Processes

Following deposition on to the leaves within a canopy, radionuclides may then be (a) absorbed and translocated, (b) washed or leached from the leaves by the passage of rain, (c) returned to the ground as the leaves and twigs are broken off or shed by the tree, and (d) resuspended from the leaves by wind (Kirchmann *et al.*, 1993). The term '*resuspension*' is commonly used to describe the transfer of material previously deposited on surfaces back into the atmosphere. The effects of resuspension can allow for the movement of radioactive material to adjacent areas and, more importantly for food chain studies, can permit the transfer of radionuclides by inhalation (Nicholson, 1988b). Once radionuclides reach the ground layer they may become available for transfer and uptake into plants and animals or may become bound to humic material or the soil itself. Subsequently, radionuclides may be leached down the soil profile (Kliashtorin *et al.*, 1994; Bachhuber, 1982) or transferred through the food chain (Fawaris and Johanson, 1995a, 1995b; Guillitte *et al.*, 1994; Strandberg, 1994).

3.2 SITE DESCRIPTION

Lady Wood (OSGR: NY 037045) is a coniferous woodland approximately 500 m north east of the perimeter fence around BNFL, Sellafield, Cumbria and as such receives a radionuclide input dominated by those aerial discharges arising from the site. Given the proximity of Lady

Wood to the sea (about 2 km), it is also likely that deposition due to the sea to land transfer of radionuclides will occur across the site. Several authors have shown that such transfer effects can be seen up to 15 km inland (Cambray and Eakins, 1980; Pattenden *et al.*, 1987). Figure 1.1 shows Lady Wood in relation to Sellafield. It is a privately owned woodland which has been undisturbed by major land use changes for the last forty years and is surrounded by agricultural land. The dominant tree species is sitka spruce (*Picea sitchensis*) which is densely planted, of similar age and covers an approximate area of 4.7 ha with a uniform canopy structure. Plate 3.1, taken from Sellafield towards Lady Wood, clearly shows the uniformity of the woodland. A small number of European larch (*Larix decidua*) are also present around the fringes of the woodland. Few grass and herbaceous species are able to grow under the dense canopy provided by *P. sitchensis*. A list of the species frequently found growing within the woodland is given in Table 3.1. These species grow in a narrow corridor, up to 3 m wide, along the front and back edges of the woodland and in small clearings where the canopy is not completely closed over. Some of the species listed are escapes from the neighbouring agricultural land. The remaining forest floor is covered by a newly deposited pine needle litter layer varying in depth over the partially decomposed litter by up to 5 cm. Plate 3.2 shows the dense canopy cover, leaf litter layer and the scarcity of vegetation within the wood.

Across the site, the typical soil pH is low at around 3.5 (Table 3.7) and the soil can be classified as a modified juvenile iron pan podzol (after White, 1987). The relatively young age of the plantation and the influence of coastal inputs of salts means that the iron pan has not yet formed completely. Visual inspection of 50 cm cores taken from the site showed a 3 cm organic horizon below which was the characteristic bleached appearance of the soil (the Ea

Table 3.1: Common species of vegetation found within Lady Wood throughout the study period.

Bracken	*Pteridium aquilinum*
Primrose	*Primula vulgaris*
White Deadnettle	*Lamium album*
Foxglove	*Digitalis purpurea*
Bluebell	*Endymion non scriptus*
Red Campion	*Silene dioica*
Lesser Celandine	*Ranunculus ficaria*
White Clover	*Trifolium repens*
Dandelion	*Taraxacum officinale*
Water Avens	*Geum rivale*
Northern Violet	*Viola selkirkii*
Wood Sorrel	*Oxalis acetosella*
Plaintain	*Plantago lanceolata*
Red Fescue	*Festuca rubra*
Tufted Hair Grass	*Deschampsia cespitosa*
Common Bent	*Agrostis capillaris*
Cocksfoot	*Dactylis glomerata*

horizon) where iron and aluminium oxides have been leached out, through to the orange red colour in the B horizon where the iron has precipitated out as ferrihydrite. Common features of this soil type are inhibited surface activity of invertebrates and the slow rate of organic matter decomposition. The slow decomposition rate is a function of the limited range of bacteria, fungi and invertebrate species capable of surviving at the low pH.

The selected study area was approximately 30 m from the southern entrance to the woodland at a point where the wood is 90 to 100 m wide. A 50 m strip through the woodland was marked out and four transects were set running parallel to the front edge of the woodland (see Figure 3.1 for a sketch map of the study site). Soil, leaf litter, vegetation and invertebrate samples were collected at intervals along each transect (section 3.3).

The wood faces Sellafield with no major land features or other woodlands to intercept any airborne particulate material before it reaches the front edge of the wood. The front edge of the woodland is also on a steep slope which ranges from 33 to 150%. The result of this is that for interception purposes, a large surface area of a uniform and dense canopy is presented along the front of the woodland.

For comparison, a small stand of sitka spruce trees was selected at Helsby, Cheshire (OSGR: SJ 484737). The site, along the edges of a golf course, was selected on the basis of having similar species of vegetation present and has been relatively undisturbed. The nearest nuclear establishment is BNFL, Capenhurst approximately 12 km away. BNFL, Capenhurst operates uranium oxide enrichment plants. Prior to 1982 the enrichment was carried out using a gaseous diffusion technique and the site now uses a gas centrifuge technique. The radioactive discharges arising from these processes consist mainly of isotopes of uranium and its daughter products, neptunium-237 and technetium-99. The discharges from the site are summarised in Table 3.2. The radionuclides listed in Table 3.2 and in the BNFL monitoring report (1994) are not of interest in this study. Consequently the Helsby site should reflect 'background' levels of the radionuclides of interest (^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am) as a result of historic weapons testing in the late 1950s and 1960s and from the Chernobyl accident in 1986. Sample collection methods employed were the same as the main study site (section 3.3).

Table 3.2: Annual radioactive discharges from BNFL, Capenhurst during routine operations in 1992 (BNFL, 1992).

Nuclide	*Liquid Discharge to Riveracre Brook*		*Solid Discharge to Clifton Marsh*	*Aerial Discharge*
	TBq	*Consented Discharge*	*TBq*	*TBq*
Total Alpha	<5	100		
Uranium	0.0016	0.02	0.00003	0.00002
Uranium Daughters	<0.007	0.02		
Non Uranic Alpha Emitters	0.00007	0.003		

Figure 3.1: Outline sketch of Lady Wood with sampling transects marked.

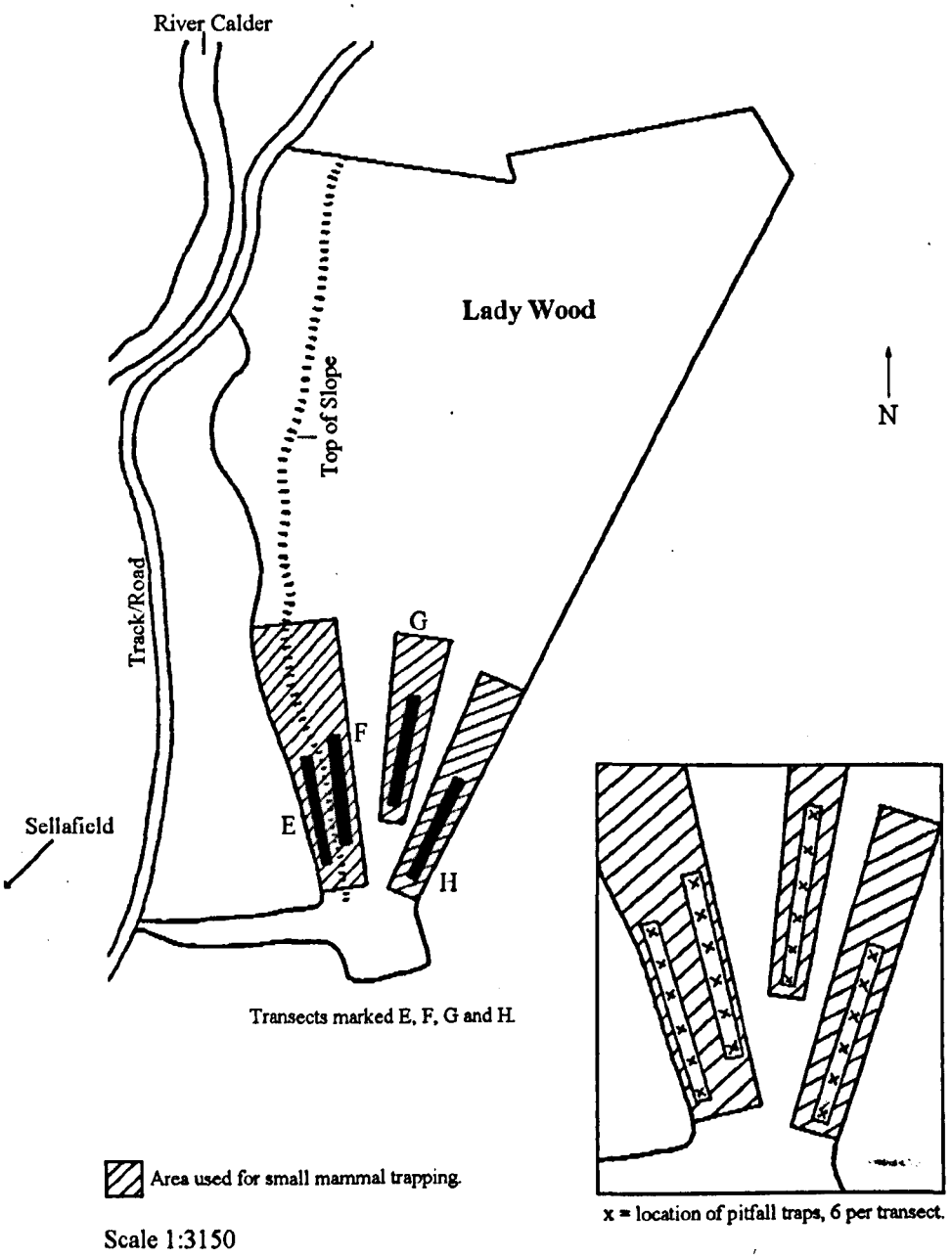


Plate 3.1: **Photograph of Lady Wood, taken from the perimeter of BNFL, Sellafield showing the uniformity of the woodland.**

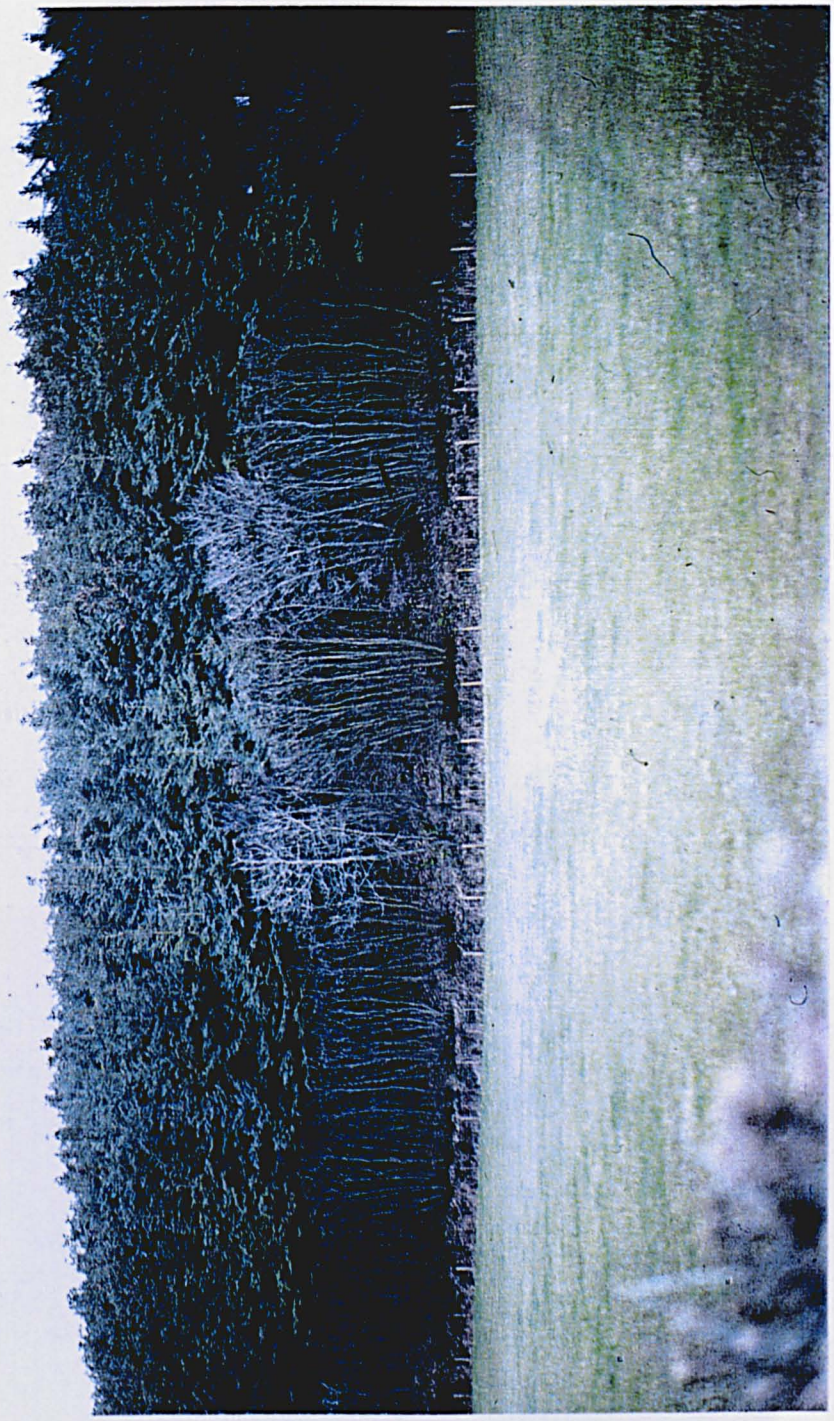


Plate 3.2: **Photograph taken inside Lady Wood showing the scarcity of vegetation.**

3.3 MATERIALS AND METHODS

3.3.1 Soil Core Sampling

Soil cores were obtained on three occasions between April 1993 and September 1994. A small area of leaf litter and/or vegetation was carefully cleared to the soil surface before a 10 cm diameter soil corer was used to extract soil cores to a depth of 9 to 12 cm. Visual inspection of the cores compared to the remaining hole, suggested that some vertical compaction (2 to 3 cm) had occurred. Depending upon the end use of the samples either two cores were collected at three points along each transect or five cores were collected along the whole of the transect. Each pair of cores was combined and large pieces of vegetation or stones were removed. Wet weights were recorded. The five core samples were sectioned in the field into 0 to 3 cm, 3 to 6 cm and 6 to 9 cm slices. The resultant sections were bulked together to provide sufficient material for counting. All of the samples were oven dried at 105°C for either 48 hours or until constant dry weights were obtained. The samples were passed through a 2 mm sieve to remove any fine organic material and stones before being homogenised in a rotary mill. The samples were then presented to the germanium detectors in Marinelli beakers. A Marinelli beaker held about 500 g of soil and these samples required in the order of 10,000 seconds to count. Sub-samples of 5 g were collected for radiochemical separation and analysis for plutonium isotopes. The radiochemical separation method is presented in Appendix A.

In addition to the short 9 cm core samples, two half metre cores of 10 cm diameter were taken from transects F and H during November 1993. These were extracted using a petrol driven vibro-corer before being sectioned into 5 cm slices. A cutting ring was then use to remove the outer edge of the core slice which was discarded to avoid any potential problems caused by the core 'smearing' during its collection. Each slice was prepared and presented to the detector as described above.

To assist interpretation of the results, a number of chemical and physical characteristics of the soil were determined. The samples used were collected from the same locations as the main soil cores described above. Weighing of samples ashed at 550°C for seven hours in a muffle furnace was used to estimate organic matter content by loss on ignition (LOI) following the procedure given by Allen (1989). Soil pH was measured with a calibrated glass electrode on fresh soil samples using a suspension of the soil in distilled water with a ratio of 1:5 (soil to water) (Allen, 1989). Calibration was carried out using buffered solutions of pH 4 and 7. A small number of samples were used to determine the particle size composition. Hydrogen peroxide (H_2O_2) was used to destroy the organic matter in 5 g of fresh soil in a solution of distilled water (Langeveld *et al.*, 1978) before the samples were passed through a laser granulometer manufactured by Malvern Instruments Ltd.

Comparison soil cores (12 cm) were collected from Cheshire during August 1993 and August 1994. Organic matter and pH measurements were also made.

3.3.2 Leaf Litter and Vegetation Sampling

Leaf litter samples consisting of pine needles from *P. sitchensis*, were collected at intervals from April 1993 to February 1995. Initially, samples were collected monthly until May 1994, then bimonthly. Samples were collected in plastic bags by carefully scraping the leaf litter down to the soil surface. Care was needed to avoid collection of the underlying mineral soil. Approximately 0.5 kg of recent litter was collected for each sample. Two samples were collected per transect on each sampling occasion. Samples were stored in a cold room (temperature below 5°C) until sorted (removal of large twigs and stones) before oven drying at 80°C for either 48 hours or until constant dry weight was achieved. Because the surface of some needles was contaminated with soil, the dried litter was placed into 2 mm and 212 μm sieves and gently brushed to remove the obvious adhered soil particles. The samples were homogenised in a Waring commercial blender and presented to the germanium detectors in Marinelli beakers. A Marinelli beaker held between 80 and 100 g of leaf litter and, depending upon the sampling location within the wood, samples required between 10,000 and 40,000 seconds to obtain sufficient data on ^{134}Cs, ^{137}Cs and ^{241}Am. Sub-samples of 25 g were collected for radiochemical separation and analysis. Comparison leaf litter samples were collected during August 1993 and August 1994.

As indicated in section 3.2, vegetation was sparse on the site. As a result, samples were only collected from the front and back edges of the woodland on three occasions during the course of the study. The samples contained a mixture of the species listed in Table 3.1 dominated by the grasses. Samples were collected by cutting the plants to ground level; wet weight was recorded and the samples were stored in a cold room at 5°C until they were oven dried at 85°C for either 48 hours or until constant dry weight was achieved. The samples were homogenised in a Waring commercial blender and presented to the germanium detectors in Marinelli beakers. Typically, a Marinelli held 40 to 60 g of vegetation but to obtain sufficient information on ^{241}Am activities, the samples required in excess of 200,000 seconds. Sub-samples of 25 g were collected for radiochemical separation and analysis. During subsequent interpretation, all of the activity results were decay corrected to the date of sample collection.

3.3.3 Sampling of Invertebrates

Invertebrate samples were collected using pitfall traps. Pitfall traps are recognised as being one of the standard methods for invertebrate sampling. They are commonly used in studies investigating invertebrate species and their ecology (Southwood, 1966). This method of

capturing invertebrate species for measuring radionuclide concentrations has been used elsewhere (Rudge *et al.*, 1993a) and for measuring heavy metal transfer through food chains (Hunter, 1984). Although the size and composition of the samples is affected by a range of factors, it is generally regarded that there is a strong correlation between the pitfall catch and the diet of small mammals such as the insectivorous shrews (*Sorex*) which find food predominantly through tactile patterns of behaviour (Pernetta, 1976) and the omnivorous mice (*Apodemus*) which are generally encounter feeders (Miller, 1954; Watts, 1968). In this study the pitfalls comprised 250 ml plastic containers with a 10 cm diameter neck which were sunk into the ground so that the lid of the container was level with the surface of the soil. Each pitfall was then charged with 100 ml of 2% formalin solution and covered with a hardboard lid to exclude rainfall, leaves and other plant debris and, to some extent, to discourage small mammals.

Four sets of six pitfalls were set within Lady Wood, one set of six in a line approximately 8 m apart along each of the transects used for sampling the soils and leaf litter (Figure 3.1). In the sampling plan, it was intended that the pitfalls placed along transects E and F would be combined to form one replicate and those set along G and H would form a second replicate. It was also expected based on work by Rudge (1989) that it would be necessary to combine the pitfall catches into quarterly samples to achieve sufficient sample mass for radionuclide determination. The pitfalls were changed at regularly intervals, usually monthly but during hot weather additional visits were made to top up the formalin solution or to change the pitfalls more frequently to prevent them drying out. Once collected, the samples were stored in a new solution of 2% formalin and placed into a cold room (<5°C). Prior to separating the invertebrate species into appropriate taxonomic groupings (section 3.7), the samples were rinsed in clean water to remove any surface contamination and to clean off the formalin solution. After grouping the species, the samples were weighed and then dried in an oven at 105°C for 48 hours. The dried invertebrates were re-weighed and ground with a pestle and mortar and placed into either 50 or 75 ml plastic pots before being presented to the gamma spectrometers. A sub-sample whose mass was dependent upon that of the original sample was also taken for radiochemistry. Typically samples were counted for 200,000 seconds and frequently ^{241}Am was determined by radiochemistry and alpha spectroscopy.

3.3.4 Sampling of Small Mammals

The small mammals were caught live using Longworth live traps (Chitty and Kempson, 1949; Phillips and East, 1961; Twigg, 1975) and the less efficient plastic Trip-Traps. Using live traps allowed some control over the type of animal (sex, age *etc.*) and the number of animals culled. This had several benefits, not least in maintaining a viable small mammal population for future work. Of the small mammal species found locally (wood mouse, *Apodemus*

sylvaticus, field vole *Microtus agrestis*, and common shrew, *Sorex araneus*) only the wood mouse was caught within Lady Wood. Each trap was baited with rolled oats, bird seed, cat food and fresh liver. This provided dietary items for each of the small mammal species. The bait was replenished every two days or after an animal was caught in a trap, whichever was sooner. In addition, straw bedding was provided within the traps to help maintain the animals once trapped as indicated by Flowerdew (1976) and Gurnell and Flowerdew (1990).

Within Lady Wood, traps were placed along the edges of the woodland and throughout the centre of the wood. Normally, traps would be positioned near to animal 'runs'; however, there was little evidence of any 'runs' in the leaf litter. Consequently traps were placed near the base of trees or by fallen logs, and near holes in the ground likely to be small mammal burrows. Trapping was carried out on four occasions during the course of this study (section 3.8). Suitable animals were culled in the field, weighed and placed into a cool box packed with ice before being frozen as soon as possible to prevent any tissue degeneration occurring. The animals were then washed before being sectioned prior to freeze-drying for 3 to 5 days in an Edwards High Vacuum system, model Supermodulyo. Dry weight was recorded. Dried samples were passed through a microhammer mill with a mesh 1.5 mm in size to produce a uniform sample matrix for radionuclide determination using a calibration standard made up in an equivalent matrix (section 2.3.5.3). Samples from the final trapping session were dissected into six parts: fur; gut; lungs; muscle; organs; and skeleton to assess the distribution of radionuclides within the body.

Trapping success varied from session to session (section 3.8). In addition, the very nature of small mammal work in confined study areas such as those in this project prohibited the removal of large numbers of each species of interest, in this case the wood mouse, *Apodemus sylvaticus*. However, for ^{137}Cs measurements, each individual mammal formed one replicate. It was only possible to measure ^{137}Cs via gamma spectroscopy, therefore ^{241}Am and the isotopes of plutonium were measured via radiochemical separation and alpha spectroscopy. To increase the efficiency of the radiochemical separation and reduce the count times required on the alpha detectors, the small mammals were bulked together as appropriate (section 3.8). Where necessary, random number tables were used to combine the individual mammal samples. Typical count times for the measurement of ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am were in the order of 250,000 to 400,000 seconds. It was not possible to detect ^{134}Cs.

3.4 SOIL RESULTS

Soils accumulate the radioactivity deposited across an area over a considerable time period. For undisturbed soils, such as those at Lady Wood, this provides a record of the local cumulative deposition for long-lived radionuclides (Cambray *et al.*, 1987). As large

fluctuations in the activity of these radionuclides can occur across a site, soils provide a useful baseline for measuring the spatial variation in deposition. The pattern of historic radionuclide deposition to a site can also be determined from soil depth profiles which are created by the leaching of radionuclides through the soil column. The data also provide information about the quantity of radionuclides present within the rooting zones of the plant species, and information about the radioactivity that might be found in the region of the burrows of small mammals. Organic matter content and pH also influence the availability of the radionuclides for plant uptake and are described later (section 3.6). Consequently, analysis of soil samples provides: (a) a baseline activity for each of the radionuclides of interest, (b) information relating to the local deposition events which have occurred in the past, and (c) information regarding the spatial distribution of radionuclides across the site.

3.4.1 Spatial and Temporal Variation

Transects E to H were sited based upon the hypothesis that if Sellafield is the principal source of radioactivity, the radioactive particles would be deposited across the site from the front to the back edge of the wood. Moreover, a greater proportion of the activity would be deposited near the front of the woodland for the reasons that are discussed below. It is important to assess the spatial variation within the study area to aid the understanding of the results obtained in the sections 3.7 and 3.8 on invertebrates and small mammals, since the mobility of these animals means that they may incorporate and integrate the radionuclide contamination from different areas within the woodland.

As an example of the spatial variability across the site, Figures 3.2 and 3.3 show the mean activity (n=3) and standard error for ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am along each transect for samples collected in April and late June 1993 and September 1994. The graphs show that the front of the woodland has received a greater amount of radioactivity and that the activity level of the radionuclides declines significantly ($p<0.001$) towards the back of the woodland. However, there is little difference between transects G and H (see below). This is in agreement with the original hypothesis.

As Table 3.3 and Figures 3.2 and 3.3 show, the radionuclide burden of the woodland is dominated by ^{137}Cs. The levels of ^{137}Cs are an order of magnitude greater than those of ^{134}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am at each sampling position. ^{137}Cs values range from an average of 1,500 ± 430 Bq kg^{-1} (± standard error, 2 significant figures) along transect E closest to Sellafield to around 460 ± 150 Bq kg^{-1} along transect H at the back of the woodland. These levels may be compared with 160 ± 33 and 86 ± 25 along transect E and 61 ± 15 and 21 ± 6 along transect H for $^{239+240}Pu$ and ^{241}Am levels respectively. The ^{137}Cs levels also exhibit a decline in activity over the 18 month sampling period along transect E, with the mean activity

Figure 3.2: **^{137}Cs Activity in Lady Wood whole soil cores - temporal and spatial variation.**

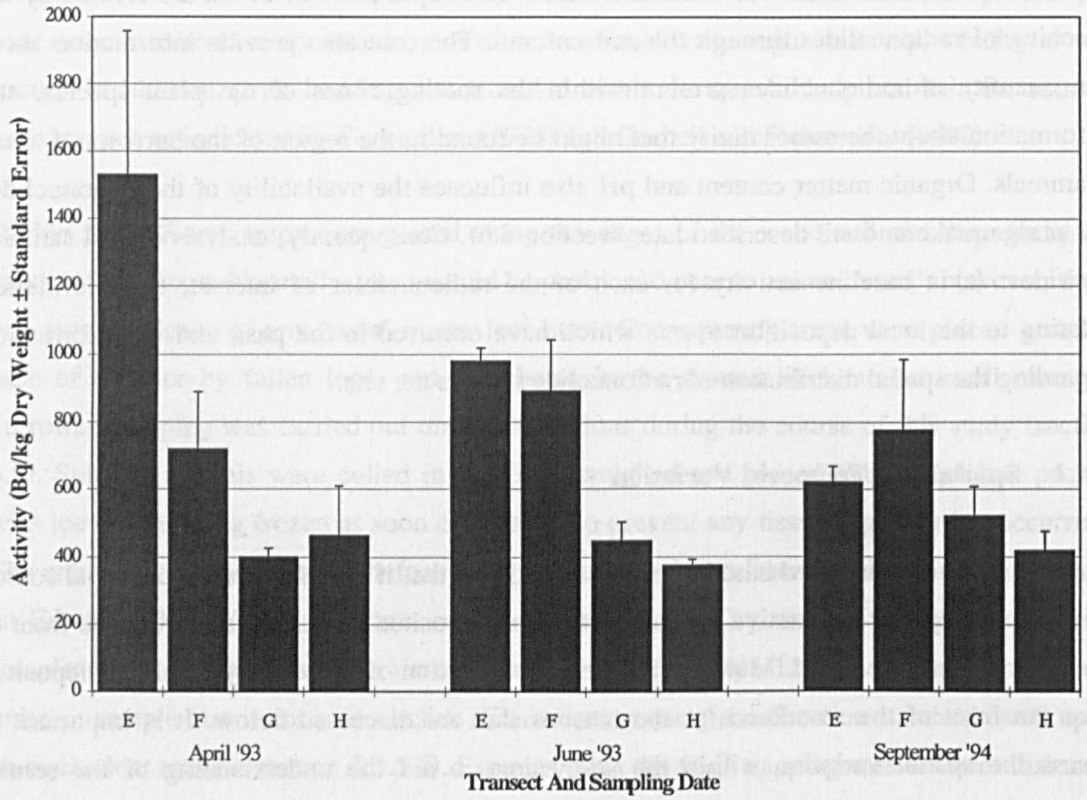
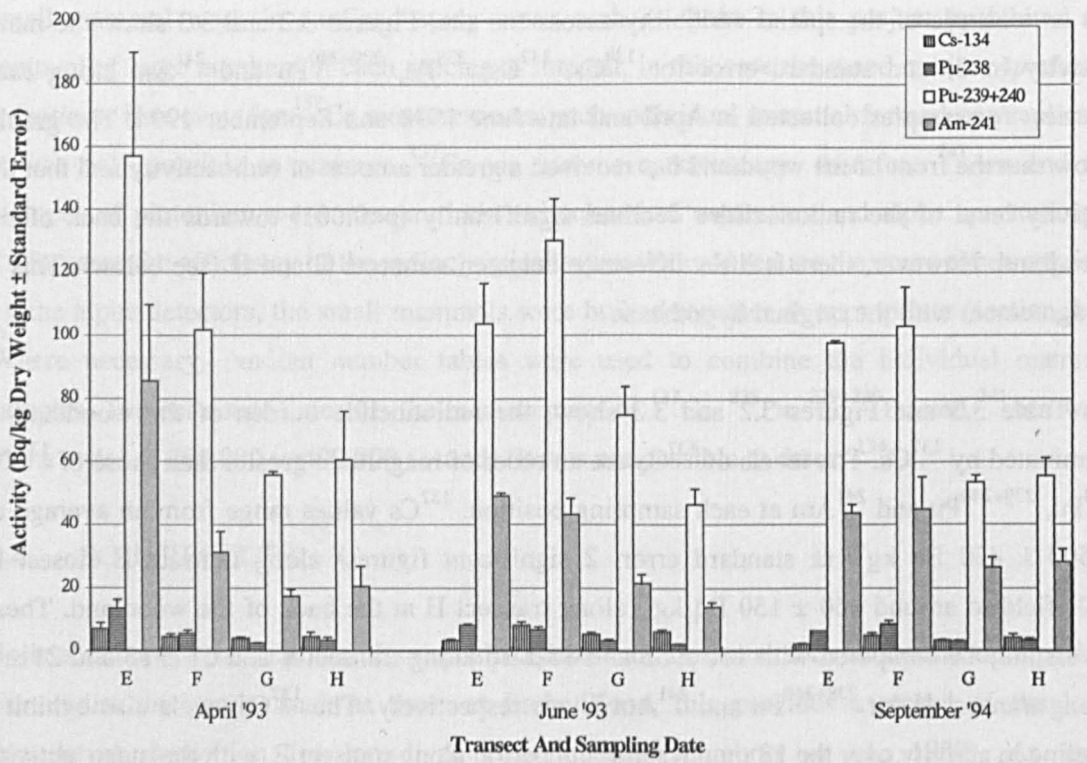


Figure 3.3: **^{134}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am Activities in Lady Wood whole soil cores - temporal and spatial variation.**



declining from 1,500 Bq kg^{-1} to 620 Bq kg^{-1}. This decline to approximately half the original level is seen for all five radionuclides, and in all cases except ^{137}Cs and $^{239+240}Pu$ this decline is statistically significant ($p<0.05$). The large standard error figures indicate the heterogeneity of soils as a sampling medium which has been commented on by Cawse (1980) and Eakins *et al.* (1981). For ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am, between 65 and 70% of the total deposited activity within the woodland is found along transects E and F reflecting the increased deposition along the front edge. ^{134}Cs is different in this respect with a more even distribution across the transects (52% of the total activity along transects E and F). Reasons for this are discussed later.

Table 3.3: Mean whole core activity (Bq kg^{-1}) for Lady Wood Soils (± s.e.), n=3.

Date	*Transect*	*^{134}Cs*	*^{137}Cs*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*
April	E	6.8 ± 1.8	1500 ± 430	13.0 ± 2.8	160 ± 33.0	86 ± 25
1993	F	4.4 ± 0.8	720 ± 170	5.5 ± 1.1	102 ± 18.0	31 ± 6.4
	G	3.9 ± 0.4	400 ± 25	2.5 ± 0.2	56 ± 1.2	17 ± 2.2
	H	4.4 ± 1.4	460 ± 150	3.4 ± 1.0	61 ± 15.0	21 ± 7
June	E	3.2 ± 0.1	1010 ± 25	8.2 ± 0.8	106 ± 22.0	50 ± 0.5
1993	F	7.0 ± 1.3	870 ± 260	6.4 ± 1.2	120 ± 18.0	40 ± 6.2
	G	4.9 ± 1.0	450 ± 90	3.4 ± 0.8	74 ± 16.0	21 ± 4.7
	H	5.2 ± 0.4	390 ± 7.0	2.4 ± 0.2	50 ± 6.0	15 ± 1.5
September	E	2.5 ± 0.1*	650 ± 66	6.4 ± 0.1	98 ± 0.5	45 ± 4.2
1994	F	4.3 ± 1.0	750 ± 350	7.9 ± 1.3	96 ± 18.0	42 ± 17.0
	G	3.0 ± 0.4	520 ± 150	3.1 ± 0.4	53 ± 3.8	25 ± 5.3
	H	3.3 ± 1.3	430 ± 92	3.8 ± 1.0	59 ± 7.4	30 ± 6.2

* Limit of Detection values used during calculations

3.4.1.1 Deposition Values, Bq m^{-2}

Table 3.4 presents data for the deposition (Bq m^{-2} to 2 significant figures) of ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am along each transect and compares the values obtained with two other studies. Cawse (1980) and Jones *et al.* (1996) collected soil cores from Lady Wood as part of a much larger project determining the levels of ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in soils around Sellafield. All of these samples refer to the deposition to a depth of 30 cm down the soil profile. The range of values obtained in 1993 shows reasonable agreement with those presented by Jones *et al.* (1996). As expected the highest deposition to the woodland occurs along transect E and this is reflected in the figure for transect E compared to the other three transects and the results presented by Jones *et al.* (1996). The results from the main area of the woodland covered by transects F, G and H, show the closest similarity with the Jones *et al.* (1996) study. However, the samples collected in 1978 by Cawse (1980) are an order of

magnitude lower for ^{137}Cs and ^{238}Pu than the samples collected in the present study and by Jones *et al.* (1996). The $^{239+240}Pu$ figures, although lower, are of a similar order in all three studies. Given that the Cawse (1980) samples were collected pre-Chernobyl, it is not surprising that ^{137}Cs deposition values have increased. Estimates for the contribution of ^{137}Cs from the Chernobyl plume to the grassland areas within Cumbria range from 3.5 kBq m^{-2} through to 10 kBq m^{-2} (Cambray *et al.*, 1987), with a wide variation even within a small sampling area (for example 3.5 to 8.7 kBq m^{-2} as reported by Clark and Smith (1988)). The amount of rainfall in a given area during May 1986 as the Chernobyl plume passed over the United Kingdom is largely responsible for the wide variation in measurements (Cambray *et al.*, 1987). It is therefore feasible that the contribution of ^{137}Cs from Chernobyl to Lady Wood is in the order of 10 kBq m^{-2} (see below).

Table 3.4: Comparison of deposition values (Bq m^{-2}) from samples collected in April 1993 with previous studies.

Transect	*Sample Date*	*^{134}Cs*	*^{137}Cs*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*	*Reference*
E	1993	250	53,000	470	5,500	2,900	This study
F	1993	230	36,000	280	5,100	1,600	"
G	1993	260	26,000	160	3,600	1,100	"
H	1993	320	32,000	230	4,000	1,400	"
Lady Wood (top 15 cm)	1978		6,100	93	3,500	---	Cawse, 1980
Lady Wood (30 cm cores)	1987	---	---	180	7,000	1,300	Jones *et al.*, 1996
Lady Wood (30 cm cores)	1987	---	39,000	---	---	---	Cawse, (unpublished data)

3.4.1.2 Isotopic and Nuclide Ratios

Table 3.5 shows the isotopic and nuclide ratios for the soil samples collected in the present study during April 1993. The ^{238}Pu:$^{239+240}Pu$ ratios across transects F, G and H are comparable with the expected range of 0.02 to 0.05 reported by Jones *et al.* (1996) for areas of high plutonium deposition. This range in the isotopic ratio is typical for plutonium released from uranium of low irradiation and therefore suggests that the bulk of the plutonium deposition occurred during the early years of Sellafield's operation. The higher ^{238}Pu:$^{239+240}Pu$ ratio for transect E reflects the fact that for ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am, transect E behaves in a different manner to the rest of the woodland. Further evidence of these differences is provided by the ^{137}Cs:^{241}Am nuclide ratio. Viewing this nuclide ratio in Table 3.5, it can be seen that transect E is noticeably different from other transects within the woodland. This suggests that either there is a lower deposition rate of ^{137}Cs or an enhanced ^{241}Am deposition along transect E. The latter is favoured in this case because of the so-called 'edge effect' where there is an

enhanced deposition of airborne particulates as the direct consequence of an interruption in the air flow, in this case caused by the trees and tree canopy (see below).

Table 3.5: Mean isotopic and nuclide ratios for Lady Wood whole core soils (± standard error) from April 1993 sampling, n=3.

Transect	*Isotopic Ratios*		*Nuclide Ratios*	
	$^{137}Cs:^{134}Cs$	$^{238}Pu:^{239+240}Pu$	$^{137}Cs:^{239+240}Pu$	$^{137}Cs:^{241}Am$
E	223.0 ± 29.1	0.086 ± 0.001	9.5 ± 1.2	17.8 ± 0.9
F	157.8 ± 22.4	0.051 ± 0.002	6.9 ± 0.5	22.6 ± 0.7
G	99.7 ± 6.0	0.039 ± 0.004	7.2 ± 0.3	23.6 ± 2.3
H	116.5 ± 41.2	0.051 ± 0.003	7.6 ± 0.9	23.6 ± 3.0

3.4.1.3 Results from spatial and temporal ANOVA

Table 3.6 shows the results of balanced two factor Analyses of Variance for the transect and temporal data of each radionuclide (^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am) respectively. Probability values marked by *, ** and *** are significantly different at $p<0.05$, $p<0.01$ and $p<0.001$ respectively. For all source categories with significant F-ratios, a 5% Least Significant Difference value (LSD) was calculated using Bonferroni's method. The LSD results are also reported in Table 3.6.

The ^{134}Cs Analysis of Variance (ANOVA) results (Table 3.6) contrast with the other four radionuclides because ^{134}Cs activity does not exhibit a significant ($p>0.05$) decline in activity from the front to the back of the woodland. Given that 52% of the total ^{134}Cs activity was present along transects E and F, compared with the 65 to 70% for the other four radionuclides, the change in activities between each transect is highly significant ($p<0.001$) for ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am. Examination of the 5% LSD values against the mean values for each transect shows that transects E and F are significantly different from G and H. This follows the original hypothesis for the spatial distribution of the radionuclides within the woodland.

Since Sellafield only releases small quantities of ^{134}Cs as liquid effluent and to the atmosphere (section 1.3.1), it is likely that the majority of ^{134}Cs was deposited as a result of the Chernobyl accident (section 1.3.3). If this is assumed, then the pattern of deposition for ^{134}Cs might well be different to the other radionuclides. Given that the majority of radionuclide deposition from Chernobyl occurred as a result of precipitation scavenging, a more uniform distribution across the woodland would be expected. A similar pattern of deposition would also be expected for ^{137}Cs released from Chernobyl and there is no evidence to the contrary. However, because ^{137}Cs is routinely discharged from Sellafield (section 1.3.1) both to the atmosphere and in liquid effluent, the deposition to woodland will be by dry deposition or sea to land transfer. In either case, the quantity of radionuclide deposition will decline with increasing distance from

Table 3.6: **Results from a balanced two factor Analysis of Variance. Test for spatial and temporal changes in whole soil core ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities respectively.**

^{134}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	2	24.790	12.395	4.62	0.020*	1.72
Transect	3	16.531	5.510	2.05	0.133	
Season*Transect	6	47.329	7.888	2.94	0.027*	3.61
Error	24	64.425	2.684			
Total	35	153.075				

^{137}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	2	225467	112733	1.42	0.262	
Transect	3	2367226	789075	9.93	0.000***	358.7
Season*Transect	6	1115277	185880	2.34	0.064	
Error	24	1906490	79437			
Total	35	5614460				

^{238}Pu:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	2	7.088	3.544	1.09	0.351	
Transect	3	259.791	86.597	26.73	0.000***	2.29
Season*Transect	6	91.587	15.265	4.71	0.003**	5.83
Error	24	77.742	3.239			
Total	35	436.209				

$^{239+240}Pu$:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	2	1687.8	843.9	1.48	0.249	
Transect	3	30579.2	10193.1	17.83	0.000***	30.43
Season*Transect	6	7360.7	1226.8	2.15	0.085	
Error	24	13721.8	571.7			
Total	35	53349.5				

^{241}Am:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	2	274.4	137.2	0.65	0.531	
Transect	3	8940.6	2980.2	14.1	0.000***	18.51
Season*Transect	6	3600.4	600.1	2.84	0.031*	32.05
Error	24	5072.5	211.4			
Total	35	17887.9				

the source. Furthermore, the woodland canopy will be more effective at intercepting airborne material along the front edge (see below). As a result, the spatial distribution for ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in the soil and leaf litter will be produced as a result of dry deposition followed by precipitation scavenging and leaching of material from the leaf canopy. Thus for ^{137}Cs, material released from Sellafield will be deposited preferentially along the front edge of

the wood (i.e. transect E). However, the inventory of ^{137}Cs across the site will have been modified as a result of the Chernobyl accident and through the continuing Sellafield discharges both to the atmospheric and marine environments. Hence the higher deposition values presented in Table 3.4 for the present study and Jones *et al.* (1996) compared to those from Cawse (1980).

3.4.1.4 Chernobyl derived radiocaesium

The ANOVA (Table 3.6) indicates that ^{134}Cs exhibits significant ($p<0.05$) temporal variation over the three sampling occasions. This was unexpected. Although soils are not static systems, it is unusual for radionuclide concentrations to change over such relatively short time periods. Many authors have reported that soils act as a 'long term' repository or sink for radionuclides (Adriano *et al.*, 1981; Cremers *et al.*, 1990; Thiry and Myttenaere, 1993), although the retention of radionuclides within the soil matrix is highly dependent upon a number of factors, for example: soil composition, rainfall, organic content *etc.* (Andolina and Guillitte, 1990; Shenber and Eriksson, 1993; Sposito, 1989). Examination of the 5% LSD values for ^{134}Cs indicates that a significant change occurred between the samples collected in June 1993 and September 1994. The change in ^{134}Cs can be explained as a function of radioactive decay. Since ^{134}Cs has a short half-life (2.065 years), it is possible that radioactive decay could reduce the radioactivity present providing that there is minimal or no further input of the radionuclide. If the ^{134}Cs is mainly derived from Chernobyl fallout as its spatial distribution implies, then decline in levels of ^{134}Cs would be expected. The interval between collections in 1993 and 1994, about 15 months, is approximately 60% of one half-life and would lead to a reduction in ^{134}Cs by a factor of 1.5. The decline in mean activity of ^{134}Cs over the 15 month period was from 5.8 Bq kg^{-1} to 3.8 Bq kg^{-1}, almost exactly by a factor of 1.5 suggesting that the ^{134}Cs decline is indeed due to radioactive decay.

If the ^{134}Cs data are extrapolated back to 1986 when the Chernobyl deposition occurred, the contribution of Chernobyl derived caesium can be estimated. Table 3.7 contains the extrapolated data for samples collected during April and August 1993. These estimates are consistent with the absolute radiocaesium deposition actually measured in the locality of Sellafield in 1986 (Fulker, 1987; Rudge, 1989).

It is not unreasonable to assume that the ^{134}Cs levels measured in Lady Wood derive from the Chernobyl deposition because only small quantities are released from the Sellafield complex and these are mainly to the aquatic environment. Furthermore, the contribution of ^{134}Cs from weapons testing will be negligible as a result of radioactive decay. The isotopic ratio, ^{134}Cs:^{137}Cs measured in the soil cores as >100 (Table 3.5) provides collateral evidence that a large component of the radiocaesium present in the woodland is attributable to the Chernobyl

Table 3.7: **Caesium data from whole and sectioned Lady Wood soil cores, decay corrected to 2nd May 1986.**

Transect	*Date*	*^{134}Cs Bq kg^{-1}*	*^{137}Cs Bq kg^{-1}*	*^{137}Cs:^{134}Cs*	*Reference*
Lady Wood					
E	April 1993	74 ± 34	1800 ± 500	25.1 ± 3.3	This study
F		48 ± 9.2	840 ± 200	17.7 ± 1.7	"
G		43 ± 4.5	470 ± 30	11.2 ± 0.7	"
H		48 ± 15	550 ± 170	13.1 ± 4.6	"
E 0-3 cm	August 1993	170 ± 96	3040 ± 15	17	This study
E 3-6 cm		58 ± 55	1700 ± 5.8	30	"
E 6-9 cm		16 ± 20	640 ± 6.4	39	"
F 0-3 cm		160 ± 61	1900 ± 13	12	"
F 3-6 cm		32 ± 18	550 ± 5.5	17	"
F 6-9 cm		8.2 ± 3.2	92 ± 3.9	11	"
G 0-3 cm		140 ± 29	900 ± 12	7	"
G 3-6 cm		34 ± 14	450 ± 7.1	13	"
G 6-9 cm		7.2 ± 5.5	160 ± 2.9	23	"
H 0-3 cm		100 ± 29	910 ± 8.2	9	"
H 3-6 cm		18 ± 11	340 ± 3.6	19	"
H 6-9 cm		4.7 ± 4.6	140 ± 2.8	29	"
Drigg	June 1986				Rudge, 1989
Grassland					
0-4 cm		113 ± 0.6	449 ± 1.1	4	"
4-6 cm		19.3 ± 0.3	222 ± 1.4	12	"
6-8 cm		12.7 ± 0.3	162 ± 0.9	13	"
8-10 cm		5.0 ± 0.3	63.4 ± 0.9	13	"

April 1993 samples were whole cores, n=3, standard error reported.
In August 1993, the sectioned cores were produced by bulking sections from five cores, resulting in one sample for analysis, therefore a 2 σ counting error is reported.
Drigg values obtained from Rudge (1989) with error term reported as standard deviation.

incident because a ratio in the order of 20 to 30 would be expected for Sellafield emissions alone (Hamilton and Clark, 1984; Rudge, 1989). Consequently, using the assumption that the ^{134}Cs is derived from Chernobyl, an estimate of the ^{137}Cs attributable to Chernobyl can be made by multiplying the decay corrected ^{134}Cs values by the isotopic ratio measured locally in 1986 at the time of the Chernobyl deposition. The caesium levels around Sellafield were monitored with high volume air samplers located at Drigg (OSGR: SD 058990) and Holmrook (OSGR: SD 078997) and from grass and soil samples collected by Fulker (1987). These measurements provide an average ^{137}Cs:^{134}Cs isotopic ratio for the local area of 1.6, therefore it is estimated that about 10% of the ^{137}Cs is derived from Chernobyl. This equates to an average deposition throughout the woodland of approximately 4,500 Bq m^{-2} using the whole core data. This estimated value, although an upper limit, produces a believable level of ^{137}Cs deposition for the woodland and is comparable to measurements made in 1986 (Cambray *et al.*, 1987; Clark and Smith, 1988). However, the contribution of the estimated

^{137}Cs to the measurements of ^{137}Cs by Cawse (1980) only accounts for about 30 to 40% of the total ^{137}Cs measured within the body of the woodland and suggests that the remainder of the ^{137}Cs is Sellafield-derived and provides a further indication of the effectiveness of the canopy at intercepting airborne particulate material.

3.4.1.5 Temporal variation of ^{134}Cs, ^{238}Pu and ^{241}Am

For ^{134}Cs, ^{238}Pu, and ^{241}Am the ANOVA Table (3.6) also indicates that there is an interaction whereby the measured activities for each transect are behaving differently over the sampling period. In fact, analysis of the 5% LSD values shows that, in all these cases, it is the behaviour of transect E which exhibits a significant decline in the activity of ^{134}Cs, ^{238}Pu, and ^{241}Am over the sampling period. To confirm that this decline is due to changes in transect E, an Analysis of Variance was undertaken without including the data from transect E. Table 3.8 summarises the ANOVA output for each radionuclide.

Not surprisingly, all the radionuclides with the exception of ^{134}Cs exhibit very significant differences between the three transects ($p<0.01$). As suspected, after the removal of the data from transect E there were no significant temporal or ANOVA interaction effects. Possible reasons for the observed change in transect E activities are discussed in the following section.

Table 3.8: Summary showing F ratios and probabilities determined by an Analysis of Variance for ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am for transects F, G and H only.

	^{134}Cs		^{137}Cs		^{238}Pu		$^{239+240}$Pu		^{241}Am	
	F	P	F	P	F	P	F	P	F	P
Season (S)	6.57	0.007**	0.14	0.14	1.30	0.297	2.65	0.098	2.89	0.082
Transect (T)	2.63	0.099	8.83	0.002**	25.51	0.000***	30.77	0.000***	12.24	0.000***
S*T	0.53	0.717	0.39	0.810	1.27	0.317	1.66	0.204	0.85	0.511

3.4.2 Radionuclide Distribution in Vertical Soil Core Profiles

Data from the 9 cm sectioned cores are presented in Figures 3.4 to 3.7. These figures show a marked decline in the activity for ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am with increasing depth. This decline in activity levels was expected and has been reported by a number of authors (Eakins *et al.*, 1981; Cawse, 1983; Cawse and Horrill, 1986). The top 6 cm contained the bulk of the radioactivity (approximately 90%) for the 9 cm cores. Several studies have described that the bulk of the activity, particularly for ^{137}Cs, can be found in the top 15 cm (around 75% of the total inventory of the 30 cm core (Cawse, 1983; Cawse and Horrill, 1986)). Just as with the whole core samples, ^{137}Cs levels dominate the profiles being one order of magnitude greater than the levels of ^{134}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am. This is around

Figure 3.4: ^{137}Cs Activity in Lady Wood soils - 9 cm depth profiles (August 1993).

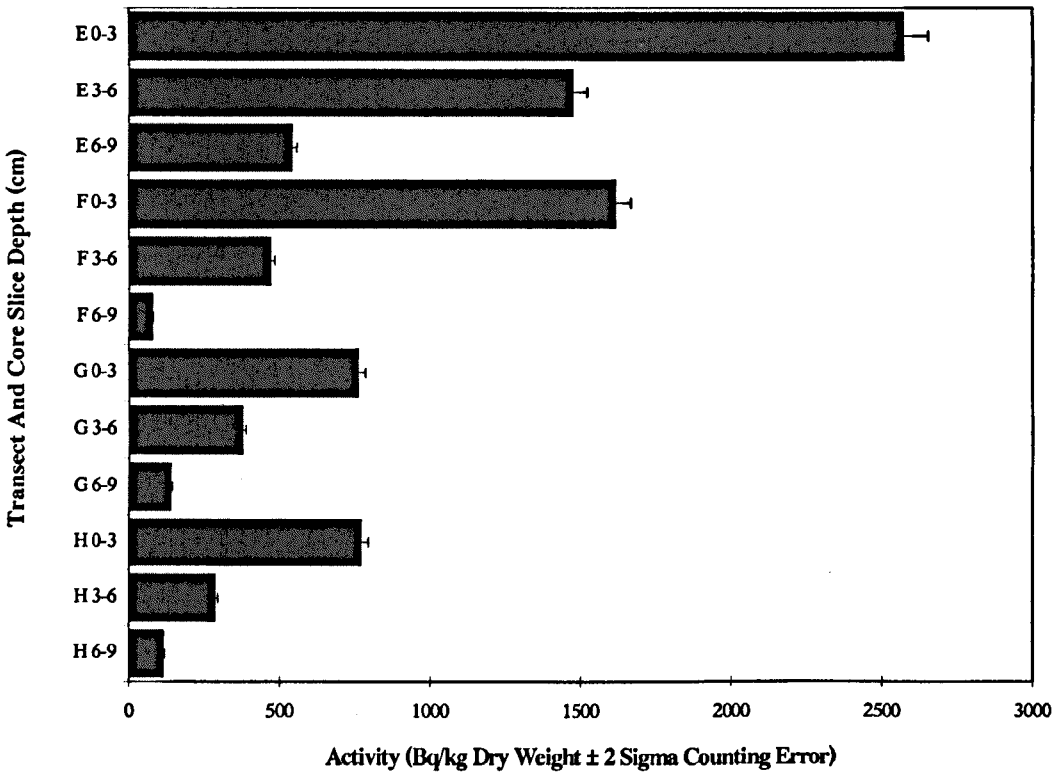
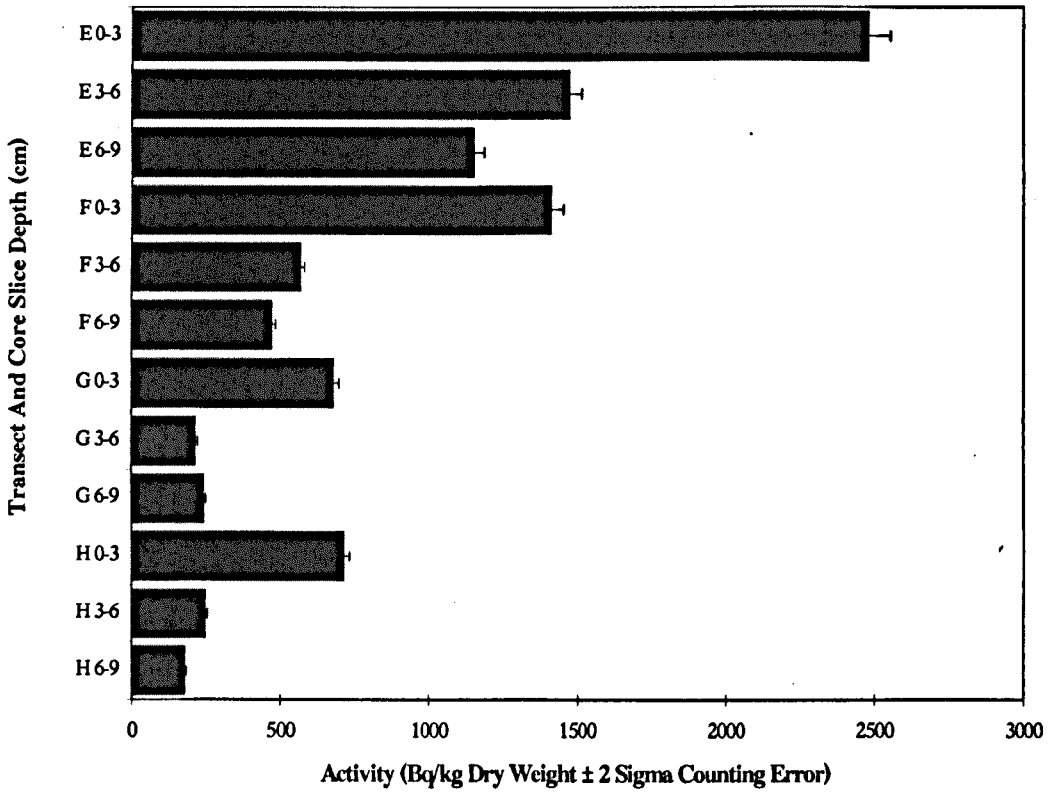


Figure 3.5: ^{137}Cs Activity in Lady Wood soils - 9 cm depth profiles (September 1994).



E, F, G and H = Transects E, F, G and H respectively.

Figure 3.6: **^{134}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am Activities in Lady Wood soils - 9 cm depth profiles (August 1993).**

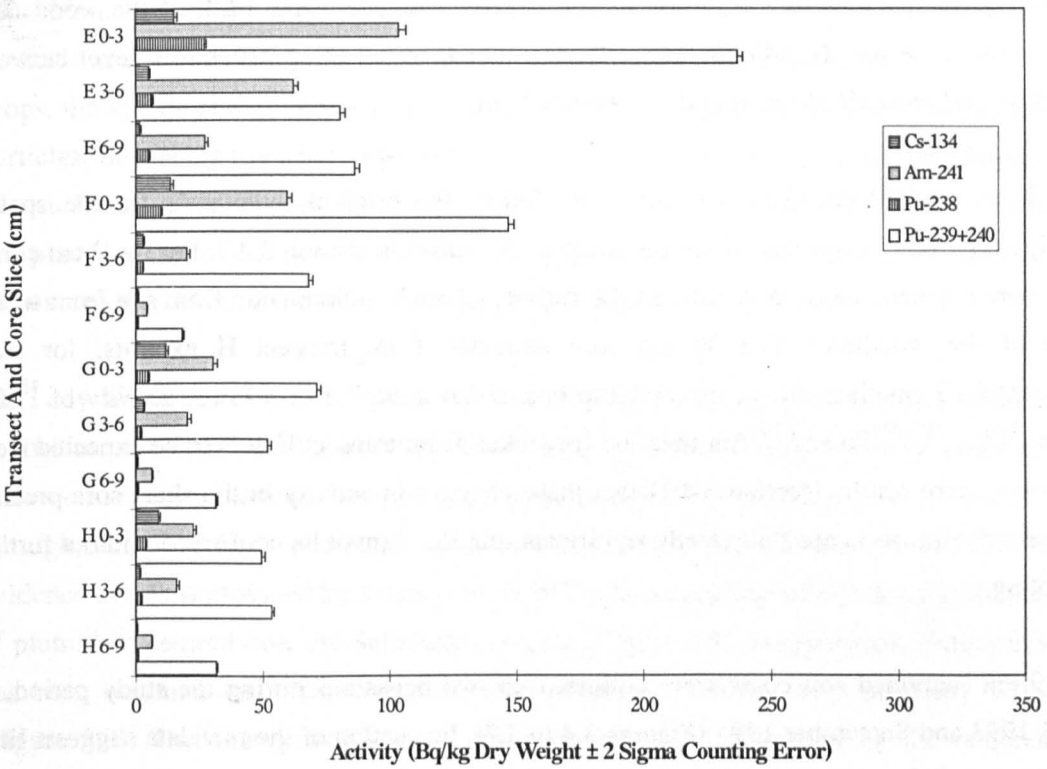
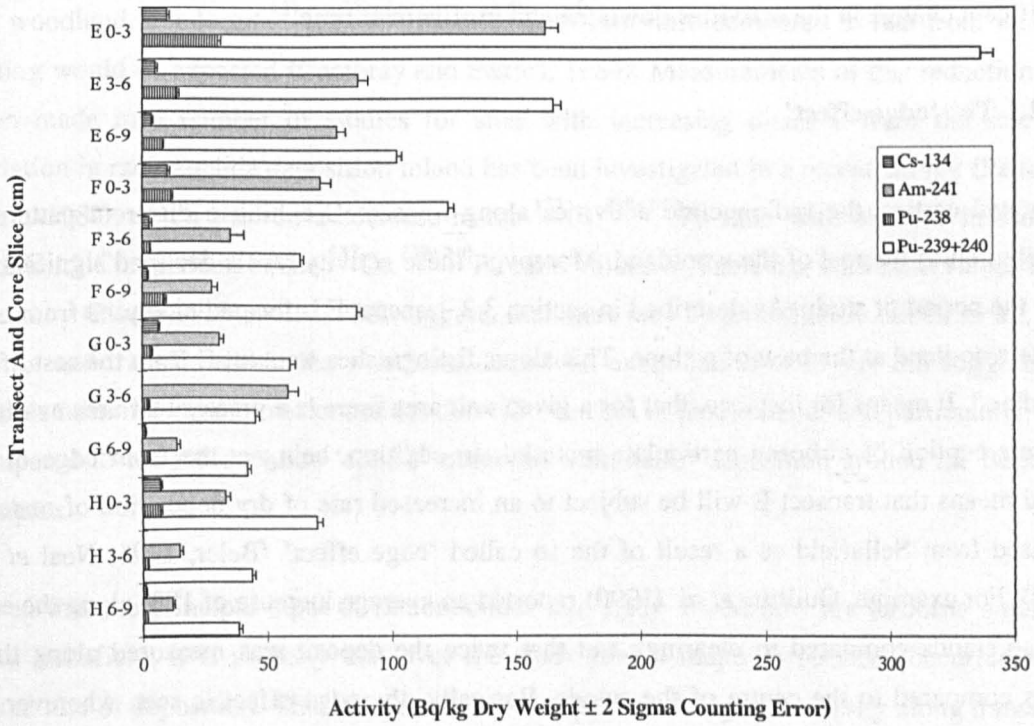


Figure 3.7: **^{134}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am Activities in Lady Wood soils - 9 cm depth profiles (September 1994).**



E, F, G and H = Transects E, F, G and H respectively.

2,500 Bq kg^{-1} for the top 3 cm along transect E to 750 Bq kg^{-1} along H. Using ^{137}Cs values, this reflects a deposition value across the site for the top 3 cm of 44,000 Bq m^{-2} along transect E which then declines to a value of 16,500 Bq m^{-2} along H at the back of the wood. The deposition value for all radionuclides also declines down all the profiles to a level between one third and one half of the activity in the top 3 cm.

The 9 cm and 50 cm sectioned core data follow the original hypothesis for the spatial distribution of radionuclides within the wood as described in section 3.4.2. For the 9 cm cores, each core segment shows a decline in the activity of each radionuclide from the front to the back of the woodland. The 50 cm core extracted from transect H exhibits, for each comparable 5 cm slice except the very top one, either a similar or a lower activity of ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am than the core taken from transect F. It is to be expected from the whole core results (section 3.4.1) that these changes in activity in the short core profiles between the transects are statistically significant, but this cannot be confirmed without further sampling.

The 9 cm sectioned soil cores were collected on two occasions during the study period, in April 1993 and September 1994 (Figures 3.4 to 3.7). Inspection of the raw data suggests little temporal variation across the site with the exception of the 6 to 9 cm core slice. In this slice the activity of each radionuclide was higher in all of the 1994 samples. This could be an indication of radionuclides being leached down the soil profile. Alternatively it could simply be due to random variation. Further sampling would be required to confirm if there is a significant change in the activities down the soil profile over time.

3.4.2.1 The 'edge effect'

As stated earlier, the radionuclide activities along transect E exhibit a different pattern of distribution to the rest of the woodland. Moreover, these activity levels declined significantly over the period of study. As described in section 3.2, transect E is located along the front edge of the woodland at the base of a slope. This slope distinguishes transect E from the rest of the woodland. It means for instance, that for a given unit area there is a greater leaf area available for interception of airborne particulate material. In addition, being at the front edge of the wood means that transect E will be subject to an increased rate of dry deposition of material released from Sellafield as a result of the so called 'edge effect' (Beier, 1991; Neal *et al.*, 1994). For example, Guillitte *et al.* (1990) reported an average increase of 18% along the edge of tree stands compared to clearings and that twice the deposit was measured along these edges compared to the centre of the stands. Basically, the edge effect is seen wherever the flow of wind is interrupted by an obstacle. In the case of a woodland the flow of air is slowed and there is increased turbulence as the air passes between the trees. The flow rate and

turbulence changes will be affected by the density and distribution of the trees within the woodland. These changes in wind speed and turbulence will greatly affect the rate of dry deposition. As briefly explained in section 3.1, dry deposition involves the deposition of particles as a result of diffusion, impaction, interception and sedimentation. If the wind speed drops, the kinetic energy of the air decreases, leading to a greater rate of sedimentation of the particles. Increasing the wind turbulence increases the chance of a particle being impacted on to a surface, such as the leaves. The chance of a particle being intercepted by the surface of the plant will also increase. Generally it holds true that as the wind speed decreases or turbulence increases, so the deposition rate of particulates will increase.

For Lady Wood this means that along transects E and F there will generally be a much greater deposition rate compared to G and H. The original hypothesis for this site proposed that the dominant source of radioactive material was from airborne emissions from Sellafield either through routine operations or from accidents such as the Windscale pile fire in 1957. Further evidence of this is provided by Jones *et al.* (1996) who showed that there is a significant spike of plutonium centred over the Sellafield complex (Figure 3.8). Furthermore, similar evidence has been obtained for ^{137}Cs with peak levels observed actually over the Lady Wood site (Figure 3.9). This then provides clear evidence that aerial deposition of the radionuclides released from the Sellafield complex is the most dominant mechanism.

It is possible that the woodland receives an input from sea to land transfer due to its proximity to the coastline. If sea to land transfer was to play an important role in the deposition across the woodland, a reduced ^{137}Cs:$^{239+240}Pu$ or ^{137}Cs:^{241}Am ratio compared to that from weapons testing would be expected (Cambray and Eakins, 1980). Measurements of this reduction have been made in a number of studies for sites with increasing distance from the sea. This variation in radionuclide deposition inland has been investigated in a recent survey (Pattenden *et al.*, 1987) which showed a decrease in the ^{137}Cs:$^{239+240}Pu$ ratio with distance inland near Sellafield. Comparison of the ^{137}Cs:$^{239+240}Pu$ ratio values in Table 3.5 with ratio values found inland by Cawse and Horrill (1986) suggests that there may be an influence, albeit small, from sea to land transfer within the woodland. However, examination of Figure 3.8 suggests that there is minimal input of radionuclides derived from sea to land transfer and particularly when compared to the radionuclide 'spikes' observed from aerial deposition around the Sellafield complex.

Given that the principal input of radionuclides into Lady Wood now are airborne emissions from Sellafield, it is unlikely that over the study period major differences occurred in the actual rate of deposition. This still leaves the unexpected changes in activity along transect E. These changes could be due to natural variation in the soil coring areas. Given that the samples were taken from a slope, differential movement of the radionuclides as solutes

through the soil or soil creep as erosional processes occur could cause variations in the radionuclide deposit. Another important feature is the change in the gradient of the slope. In general, samples collected from the more shallow slope contained between 20 and 50% more radioactivity. This is partly due to the greater accumulation of leaf litter and eroded material from the steeper parts of the slope. Since the cores were not taken from exactly the same locations, it is suspected that differences in the radionuclide activity are a result of variation in gradient. A more detailed sampling programme would be required to confirm this.

Two 50 cm cores were extracted in addition to the normal 9 cm cores to examine the soil profile distribution in more detail. These cores were taken from transects F and H to assess any differences in the core inventory due to the spatial variation of the radionuclide deposition, particularly in terms of activity levels. The results are shown in Figures 3.10 and 3.11. As these cores were not a major component of this study, the soil material was bulked into 5 cm slices. The cores lack a detailed resolution making it difficult to distinguish any short term deposition events. However, the results show quite clearly that there is a continued decline in the activity levels below 9 cm. Approximately 25 to 30 cm down, the levels of activity for each of the radionuclides fall to a level comparable to present background activity in the surface soils of the comparison site in Cheshire (section 3.4.3). The decline in core activity follows an exponential curve. For the 50 cm depth from transect F, r^2 was >0.9 for ^{137}Cs, $^{239+240}Pu$, ^{241}Am and was >0.6 for ^{134}Cs and ^{238}Pu. The lower r^2 for ^{134}Cs and ^{238}Pu reflects the number of values below the limit of detection. This is almost the same for transect H but only ^{137}Cs and $^{239+240}Pu$ provide a good fit (r^2>0.9) to an exponential curve.

The most important aspect of these profiles is they show that greater than 85% of each of the radionuclides is present in the top 20 cm. Schell and Tobin (1990) also report that in woodland soils in the Great Britain, Japan and Pennsylvania, USA, greater than 90% of the ^{137}Cs is in the top 20 cm. Since the predominant rooting depth for the root mass of most grass and herbaceous species is less than 20 cm, the bulk of the radionuclides is theoretically plant-available, depending upon the soil characteristics that affect the mobility of radionuclides in soil (section 1.4.1).

3.4.2.2 Soil characterisation

Although the scope of this study did not allow for a detailed examination of the factors that affect the mobility, and therefore availability, of radionuclides for plant uptake, it is useful to refer to the more important ones. As explained in section 1.4.1 organic matter content, pH and clay content are some of the more important soil characteristics (Bergeijk *et al.*, 1992; Cawse and Turner, 1982; Livens and Rimmer, 1988; Morgan 1990; Shenber and Eriksson, 1993),

Figure 3.8: **$^{239+240}$Pu deposition in the vicinity of Sellafield, based on Ordnance Survey grid references.**

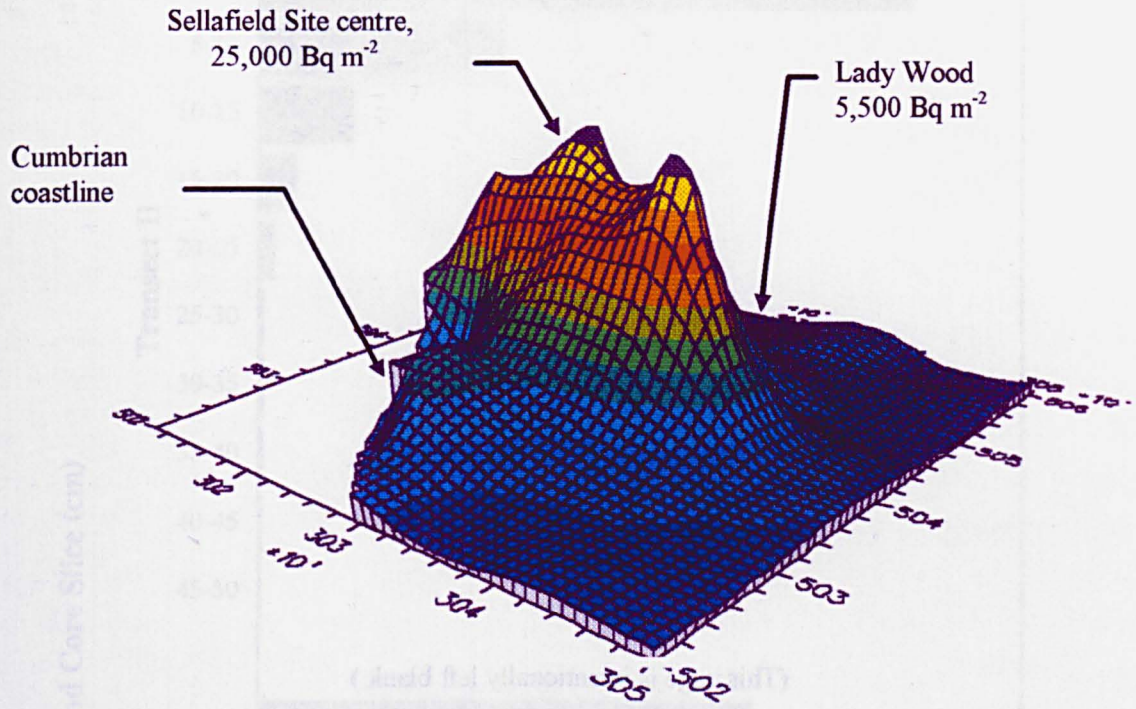
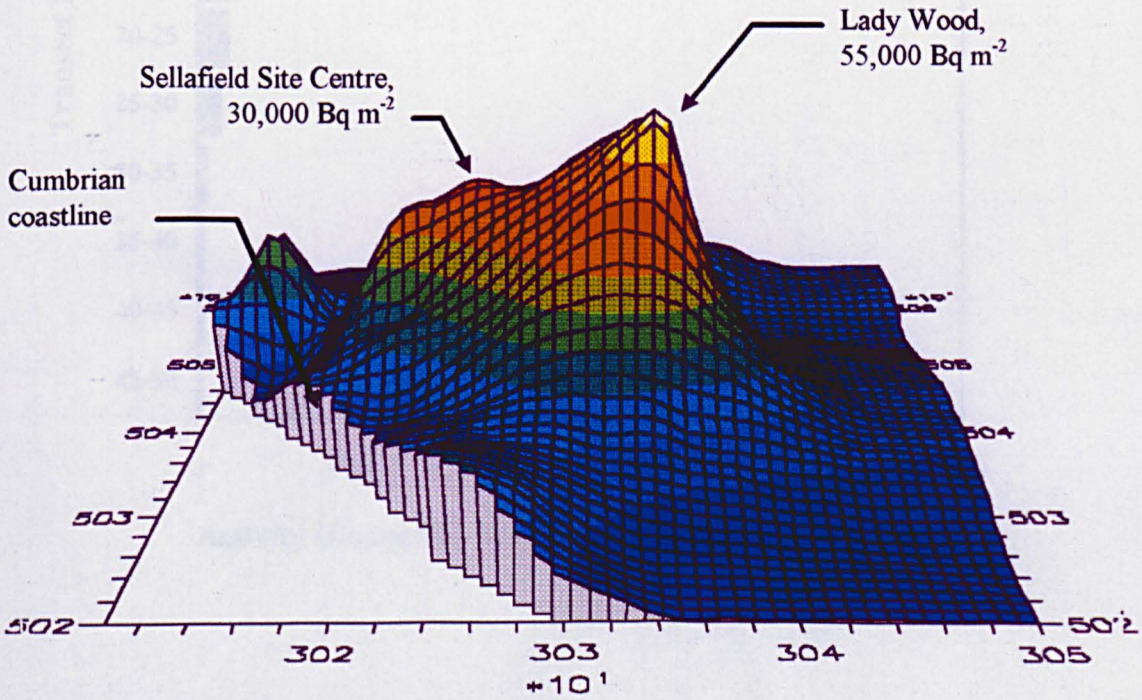


Figure 3.9: **^{137}Cs deposition in the vicinity of Sellafield, based on Ordnance Survey grid references.**



Data used to generate graphs were obtained from this study, Jones *et al.* (1996) and Jones (pers. comm.).

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Figure 3.10: ^{137}Cs Activity in Lady Wood soils - 50 cm core profiles.

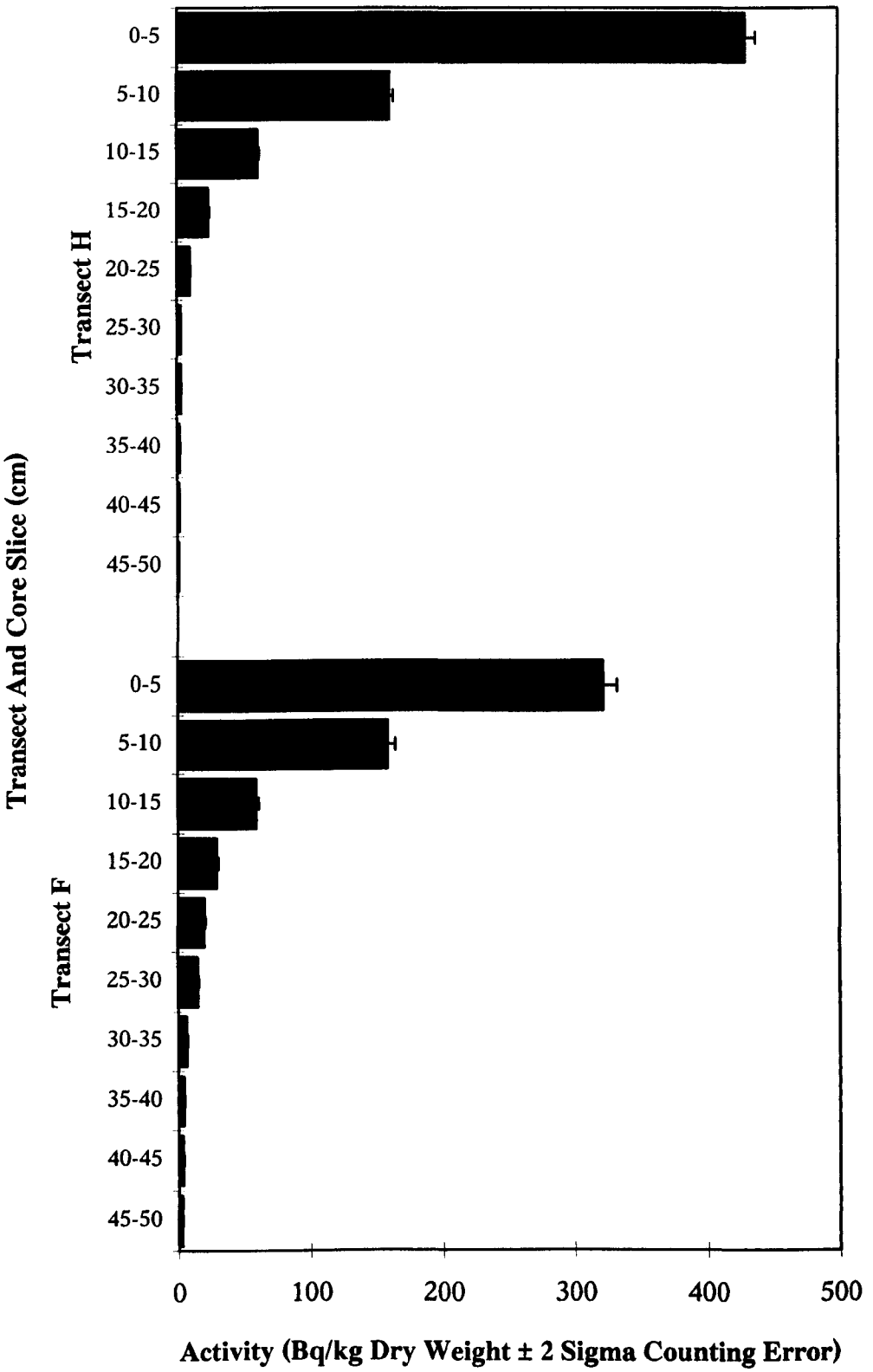


Figure 3.11: ^{134}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am Activities in Lady Wood soils - 50 cm core profiles.

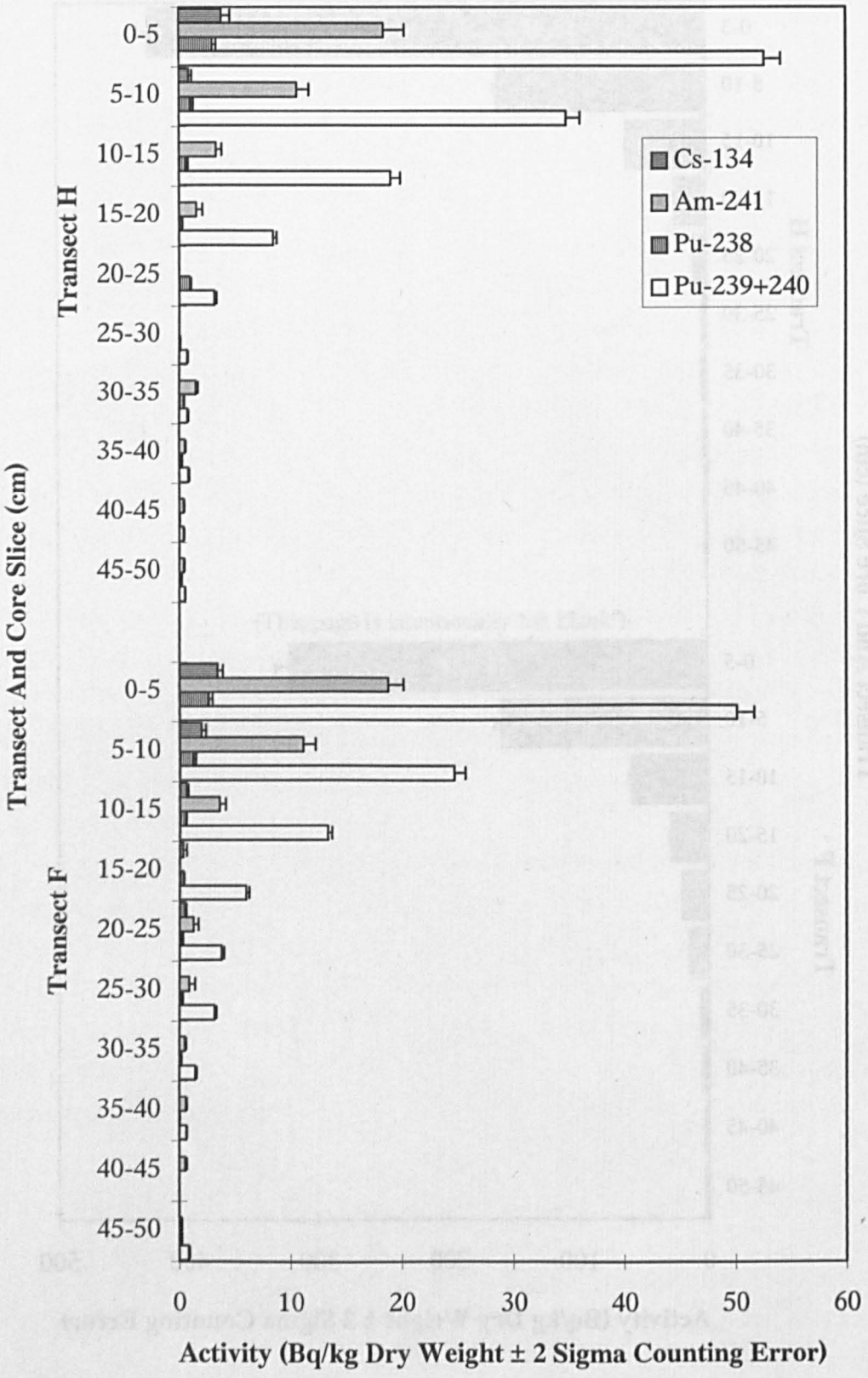


Table 3.9: Soil parameter results for Lady Wood soils (± standard error), n=2.

Sampling Location	*pH*	*% Loss on Ignition**	*Particle Size Distribution (%)***		
			Sand (>20 μm)	*Silt (2-20 μm)*	*Clay (<2 μm)*
Transect E	3.8 ± 0.14	19.4 ± 2.3	48.8 ± 1.1	41.5 ± 1.1	13.0 ± 3.2
Transect F	3.4 ± 0.18	18.3 ± 1.5	47.8 ± 6.2	36.0 ± 3.7	16.3 ± 2.6
Transect G	3.4 ± 0.1	13.8 ± 1.0	43.4 ± 7.0	46.3 ± 12.0	10.3 ± 5.0
Transect H	3.4 ± 0.22	13.4 ± 0.8	34.6 ± 1.3	45.6 ± 0.4	19.8 ± 0.9
E 0-3 cm	n/a	32.2 ± 3.4	n/a	n/a	n/a
E 3-6 cm	n/a	21.6 ± 6.2	n/a	n/a	n/a
E 6-9 cm	n/a	17.3 ± 3.3	n/a	n/a	n/a
F 0-3 cm	3.4 ± 0.18	28.5 ± 6.6	44.2 ± 7.8	42.6 ± 8.0	13.2 ± 0.2
F 3-6 cm	3.2 ± 0.02	17.9 ± 7.2	38.8 ± 1.6	43.1 ± 1.8	18.1 ± 0.2
F 6-9 cm	3.3 ± 0.08	12.9 ± 6.1	32.1 ± 7.8	45.5 ± 5.4	22.5 ± 2.4
G 0-3 cm	n/a	17.9 ± 2.7	n/a	n/a	n/a
G 3-6 cm	n/a	10.9 ± 1.6	n/a	n/a	n/a
G 6-9 cm	n/a	9.4 ± 3.1	n/a	n/a	n/a
H 0-3 cm	3.6 ± 0.28	14.2 ± 3.4	33.2 ± 0.9	44.2 ± 1.5	22.6 ± 0.6
H 3-6 cm	3.5 ± 0.13	10.8 ± 0.1	36.8 ± 3.6	43.0 ± 1.7	20.3 ± 1.8
H 6-9 cm	3.5 ± 0.05	9.0 ± 2.3	37.3 ± 1.4	45.6 ± 0.9	17.0 ± 2.4
Cheshire Site	4.3 ± 0.14	14.5 ± 0.3	n/a	n/a	n/a
0-3 cm	n/a	14.2 ± 0.6	n/a	n/a	n/a
3-6 cm	n/a	8.8 ± 2.0	n/a	n/a	n/a
6-9 cm	n/a	9.5 ± 0.9	n/a	n/a	n/a

* Loss on ignition (LOI) results averaged for each transect over the three sampling occasions.
** Particle size divisions after Russell (1973).
n/a - No analysis carried out.

although these factors can be modified by the effects of climatic conditions (primarily rainfall) and bioturbation. A limited number of samples were analysed for pH, organic matter content and particle size distribution; the results are presented in Table 3.9.

As already discussed (section 1.4.1), the interaction of radionuclides with soils is complex and there is no one dominant factor controlling the adsorption to the soil and availability for uptake. The radionuclides reach the soil through direct deposition, canopy interception followed by washoff and leaf fall. Subsequent radionuclide passage into the soil is controlled by further leaching of the radionuclides from the leaf litter down into the mineral soil and also through the processes of decomposition (Sauras *et al.*, 1994). Some direct incorporation of the leaf litter and other plant debris into the soil may also be achieved by the burrowing activity of some animal species. Within Lady Wood, the radionuclide distribution within soil profiles described previously, will have been produced primarily through leaching and vertical migration of the radionuclides. Investigating the soil/radionuclide interactions is in itself a major study beyond the scope of the current project. However, some knowledge of the major

factors controlling the mobility and, therefore, bioavailability of the radionuclides is beneficial in determining transfer factors from the soil to plants (section 3.6).

Section 3.2 has already described Lady Wood's soil type as that of a juvenile podzol and inspection of the extracted soil cores has shown the stratified layers typical of this classification. The results presented in Table 3.9 confirm that there is a low pH and low clay content in the mineral soil, again typical of this type of podzol (Melin *et al.*, 1994; White, 1987).

Several authors report that the majority of the radionuclide inventory, particularly for ^{137}Cs and plutonium, is found bound within the upper organic horizons within the soil profile under forests (Burmann, 1993; Burmann *et al.*, 1994; Cook *et al.*, 1984; Kuhn *et al.*, 1984; Thiry *et al.*, 1994). In general, the retention of radionuclides within the upper layers of the soil cores indicates that they are strongly adsorbed to, or complexed with, the organic matter in the upper soil layer. Table 3.9 shows that the measured organic matter content is higher at the soil surface. This was predicted prior to coring since, at depth, most of the organic matter will have been decomposed, but for this site the amount of organic matter is controlled by the decomposition of the leaf litter. Since there is a slow decomposition of the leaf litter because of the nature of pine needles and the acidic conditions prevailing, there is a large surface area and quantity of organic matter available for absorption of radionuclides. It has been reported that caesium particularly is considered available for root uptake by plants from these organic soil horizons and that the exchangeable fraction can readily leach from the upper region of the mineral soil (Melin *et al.*, 1994). Even so, a proportion of the radionuclides will become strongly bound to the organic fraction, mainly through the complexation of radionuclides with the organic matter. In this case, it is only during the decomposition of the organic matter that these radionuclide ions will be released and become available for either uptake into plants/animals or for further leaching down the soil profile (Melin *et al.*, 1994). Plutonium, in particular, has been shown to readily form complexes with organic matter (Bulman, 1983; Cook *et al.*, 1994; Litaor *et al.*, 1994; Livens and Rimmer, 1988). Analysis of the soil cores by sectioning the soil horizons, rather than at fixed specified depths, would have provided more detailed information on the distribution of the radionuclides within the vertical profile.

The soil pH was low but reasonably uniform across Lady Wood. At low pH most metal ions become more soluble thus making the radionuclides more bioavailable. Countering this, however, is the fact that stable metals, in particular sodium and potassium, will also become more available and, since these compete directly for uptake into plants, there may or may not be an actual increase in the transfer of radionuclides to plants. For example, Kuhn *et al.* (1984) report that as pH decreases, transfer coefficients for ^{137}Cs increase, resulting in greater uptake. As mentioned previously, pH is also very important for this site because the low pH

limits the rate of decomposition. In doing so the availability of radionuclides adsorbed to the leaf litter is reduced and this gives rise to a time lag between the deposition of the radionuclides to the canopy and their subsequent release for uptake into plants/animals or for further leaching.

The low clay content (Table 3.9) is again typical of coniferous forest soils. The effect of clays on the availability of radionuclides, particularly ^{137}Cs again, has been well characterised for different soil types, especially those found in agricultural land. Typically, clays bind radionuclides irreversibly. Therefore, soils with high clay content produce the greatest fixation of ^{137}Cs (Squire and Middleton, 1966). The fixation of the ^{137}Cs into a non-exchangeable form will occur gradually over time and is therefore dependent upon factors such as the amount of clay present; time available for sorption; organic matter content and the competition by other ions for the binding sites. Plutonium activity has been shown to increase with decreasing particle size (Litaor *et al.*, 1994) and also to be held in the upper soil layers. The latter confirms the findings in the sectioned cores in the current study, where approximately 70% of the plutonium inventory is located within the top 10 cm (this section).

Particle size within the soil also determines the drainage and aeration of the soil. In this case, the particle size is evenly distributed and has produced a relatively free draining soil. One feature of free draining soils is that the vertical transportation of radionuclides may occur more readily, partly because of the presence of channels through the soil and partly because free water movement means a limited residence time within the soil for adsorption to occur. This is perhaps why plutonium is seen down to 15 to 20 cm within the present study, whereas several studies report that 90% of the plutonium in soils is located within the top 10 to 12 cm (Cook *et al.*, 1984; Litaor *et al.*, 1994). Litaor *et al.* (1994) also reports varying depth profiles for plutonium which are dependent upon particle size and the presence of macropores, usually produced as a result of the decay of plant roots. For Lady Wood, there is a large tree root mass just below the soil surface; this could aid the movement of the radionuclides within the soil, firstly by direct uptake into the trees and secondly by providing potential transfer paths for the movement of the radionuclides down through the soil profile under the influence of water flow.

3.4.3 Comparison of Soil Samples Collected in Cheshire to those from Lady Wood

Table 3.10 compares the soil activity data for Lady Wood with the results from the Cheshire site. Not surprisingly the data clearly show that the radionuclide contamination in Lady Wood is greater than the Cheshire site. For example, ^{137}Cs levels are between 20 and 120 times higher, depending upon the transect, than those for soil samples collected from Cheshire. The Cheshire site receives negligible input of Sellafield-derived radionuclides. However, ^{134}Cs,

^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am values for the Cheshire site do provide a measure of the background levels of anthropogenic radionuclides released during the nuclear weapons testing of the 1950s, early 1960s and from the Chernobyl accident in 1986. The levels of ^{137}Cs activity in Cheshire are in agreement with data collected from Woolston Eyes, Warrington in Cheshire by Rudge (1989). The Cheshire site also compares favourably to data reported by Cawse and Horrill (1986) with regard to plutonium. Cawse and Horrill (1986) determined ^{137}Cs and $^{239+240}Pu$ activities in a range of soil samples collected across the UK in 1977. For the general region incorporating Cheshire, values ranging from 0.22 Bq kg^{-1} to 0.37 Bq kg^{-1} were recorded. Also, by comparing the plutonium data to a Welsh control site used in this current work (Chapters 5 and 6), where $^{239+240}Pu$ values of 1 to 2 Bq kg^{-1} were measured, a reasonable measure of the 'background' levels of plutonium attributable to weapons testing is

Table 3.10: A comparison of the ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in soil cores. Results from soils at Lady Wood and Cheshire.

Transect/Site	*^{134}Cs*	*^{137}Cs*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*
Whole Cores					
E	3.7 ± 0.4	1010 ± 15	8.3 ± 0.4	106 ± 12	50 ± 0.3
F	7.5 ± 0.8	880 ± 150	6.4 ± 0.7	120 ± 10	40 ± 3.4
G	5.2 ± 0.6	460 ± 53	3.4 ± 0.5	74 ± 9.0	21 ± 2.7
H	5.6 ± 0.2	390 ± 4.1	2.5 ± 0.1	50 ± 3.5	15 ± 0.9
Cheshire	<0.15**	12 ± 1.5	0.015 ± 0.01	0.42 ± 0.05	0.25 ± 0.05
Sectioned Cores					
E 0-3 cm	15 ± 1.3	2600 ± 82	27 ± 0.7	240 ± 2.1	103 ± 2.9
E 3-6 cm	5.1 ± 0.5	1500 ± 47	6.2 ± 0.5	80 ± 1.9	62 ± 1.8
E 6-9 cm	1.4 ± 0.6	540 ± 17	5.0 ± 0.5	86 ± 2.0	27 ± 1.3
F 0-3 cm	13 ± 1.1	1600 ± 51	9.8 ± 0.6	150 ± 2.5	60 ± 2.0
F 3-6 cm	2.8 ± 0.5	470 ± 15	2.6 ± 0.4	68 ± 1.8	20 ± 1.3
F 6-9 cm	0.7 ± 0.3	78 ± 2.7	0.8 ± 0.1	18 ± 0.5	4.2 ± 0.3
G 0-3 cm	12 ± 1.0	760 ± 24	4.9 ± 0.5	71 ± 1.9	30 ± 1.7
G 3-6 cm	3.0 ± 0.6	380 ± 12	2.1 ± 0.3	52 ± 1.6	20 ± 1.6
G 6-9 cm	0.6 ± 0.3	140 ± 4.6	0.9 ± 0.1	32 ± 0.6	6.3 ± 0.4
H 0-3 cm	8.9 ± 0.7	770 ± 25	3.8 ± 0.4	49 ± 1.4	22 ± 1.2
H 3-6 cm	1.6 ± 0.3	290 ± 9.3	2.1 ± 0.2	53 ± 1.0	16 ± 1.1
H 6-9 cm	0.4 ± 0.3	110 ± 3.9	0.9 ± 0.1	32 ± 0.6	6.1 ± 0.3
Cheshire 0-3 cm	0.2 ± 47	17 ± 3.6	0.02 ± 72.55	0.69 ± 10	0.70 ± 17
Cheshire 3-6 cm	<0.17**	14 ± 3.7	0.01 ± 172.13	0.46 ± 15	0.30 ± 25
Cheshire 6-9 cm	<0.15**	8.4 ± 4.2	0.02 ± 74.36	0.37 ± 17	0.30 ± 25

Whole core data reported as mean activities (Bq kg^{-1} dry weight) ± standard error, n=3.
Sectioned cores reported ± 2σ counting errors.
Lady Wood data taken from June 1993, Cheshire data from August 1993.
** Limit of Detection values.

provided. The differences in radionuclide levels between the two reference sites are likely to reflect variation in rainfall, influence of sea to land transfer (at the Welsh site) and differences in soil characteristics.

Results of a One-way Analysis of Variance for whole core samples collected in June (Lady Wood) and August (Cheshire) 1993, show a significant difference ($p<0.01$) between each of the transects in Lady Wood and the Cheshire site. The ANOVA output is shown in Table 3.11 and includes a 5% LSD value calculated using Bonferroni's method for comparison with the mean whole core soil activities given in Table 3.9.

Table 3.11: Results for ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am from a One-way Analysis of Variance on Lady Wood and Cheshire whole core soil samples.

^{134}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	4	59.83	14.96	10.14	0.013*	2.19
Error	5	7.37	1.47			
Total	9	67.20				

^{137}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	4	1296759	324190	10.57	0.012*	315.86
Error	5	153389	30678			
Total	9	1450148				

^{238}Pu:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	4	84.99	21.25	19.16	0.003**	1.90
Error	5	5.54	1.11			
Total	9	90.53				

$^{239+240}Pu$:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	4	18399	4600	10.84	0.011*	32.13
Error	5	2121	424			
Total	9	20520				

^{241}Am:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	4	3149.30	787.30	31.12	0.001***	9.07
Error	5	126.50	25.30			
Total	9	3275.80				

3.4.4 Summary of Soils Results

Soils in Lady Wood show evidence of the accumulation of radionuclides due to atmospheric deposition over an extended period of time. Accumulated deposits lie within the range 250 to 320 Bq m^{-2} (^{134}Cs), 2,600 to 53,000 Bq m^{-2} (^{137}Cs), 160 to 470 Bq m^{-2} (^{238}Pu), 3,600 to 5,500 Bq m^{-2} ($^{239+240}Pu$) and 1,100 to 2,900 Bq m^{-2} (^{241}Am). These levels of accumulated deposition are broadly consistent with measurements made in the immediate vicinity by other researchers.

Deposits of plutonium are characterised by low values of the ratio ^{238}Pu:$^{239+240}Pu$, in the range 0.039 to 0.086, which are characteristic of plutonium produced from uranium of low irradiation and indicate that much of the deposition results from the early years of operation of the Sellafield site. Much of the ^{134}Cs deposited at the site appears to originate from the Chernobyl deposition in 1986, and this would imply that Chernobyl could account for up to 4,500 Bq m^{-2}, or about 10-20%, of the ^{137}Cs deposit.

All radionuclides, with the exception of ^{134}Cs, show a consistent fall in accumulated deposit from the 'front' to the 'back' of the wood; this is attributed to the 'edge effect' whereby greater deposition occurs at the leading edge of the woodland which is first encountered by aerosols dispersing from the Sellafield site. The more uniform deposition pattern for ^{134}Cs is consistent with the hypothesis that ^{134}Cs deposits largely arise from wet deposition when the Chernobyl 'cloud' reached Cumbria in early May 1986.

3.5 LEAF LITTER RESULTS

Section 3.1 described the role of leaf canopies in intercepting airborne particulates. In Lady Wood, leaves form the primary route for capture and subsequent relocation of radionuclides to the forest floor where they may become available for transfer into indigenous food chains. The leaf canopy is very dense, so direct deposition of particulate material to the forest floor is difficult. Recent work on spruce canopies in wind tunnel experiments showed that over 99% of the particulate material was deposited in the tree canopy (Ould-Dada, 1996). Several studies are now investigating in detail the atmospheric deposition of radionuclides to woodland canopies, particularly using wind tunnel experiments (Kinnersley *et al.*, 1994; Shaw *et al.*, 1994) resulting from the interest generated following the Chernobyl accident in 1986. Data from these studies are currently being used to generate predictive models for understanding the effects of accidental releases and the application of counter-measures.

The canopy plays a vital role in the accumulation of airborne pollutants. This is affected by a wide range of factors, some determined by the canopy/wood itself, such as age, structure and

leaf area index (Draaijers *et al.*, 1992; Guillitte *et al.*, 1990), and some determined by climatic conditions and season of the year at the time of deposition (Raitio and Rantavaara, 1994). Several studies have investigated the effects of foliar structure and seasonal variation on the accumulation of ^{137}Cs (Antonopoulos-Domis *et al.*, 1990; Ertel and Ziegler, 1991; Kinnersley *et al.*, 1994; Raitio and Rantavaara, 1994). Differences in deposition to the canopy can account for some of the variation seen in the soil activities measured below the stand mainly as a result of differences in throughfall and wash-off variations within the tree canopy (Beier *et al.*, 1993; Hansen *et al.*, 1994). For example, several authors have observed high spatial variation in soil samples (Bunzl *et al.*, 1989; Guillitte *et al.*, 1990) which was attributable to variation in the woodland structure. In the current study, there was an overall spatial variation in the measured radionuclide activities across the whole site and, as above, this can be explained by the deposition across the canopy. In this case, it is not due to a variation in the age/structure or humus variation within the woodland but rather to the presence of a long term point source of activity release coupled with direct interception of the canopy (see below).

3.5.1 Radionuclide movement and transfer within the canopy

Within the canopy radionuclides may enter the leaves via stomata or cuticle, or may be bound to the external leaf surface. It has been shown, for example, that caesium is very readily absorbed by leaves following deposition (Kirchmann *et al.*, 1993). Once deposition to the canopy has occurred, relocation to the forest floor may happen as a consequence of rainfall leaching the particulate material from the canopy or as a result of leaf fall. In either case, radionuclides relocate to forest floor where they are deposited on to the leaf litter layer. Retention times for intercepted radionuclides deposited on to leaf surfaces and then subsequently weathered have been calculated by a number of authors. Witherspoon and Taylor (1969) assessed the half-life of ^{134}Cs on oak and pine trees using a fallout simulant. Unsurprisingly, oak leaves lost ^{134}Cs much more rapidly than pine needles. Sombre *et al.* (1990a) also determined the retention time for ^{134}Cs on spruce trees using thermo-generated aerosols under controlled conditions. Both studies determined that there are two components to the retention curves. The initial component showed a removal half time of five days, whereas the removal half time for the second component was 50 days. Both measurements were confirmed through the analysis of the retained ^{134}Cs on the needles. This is not directly applicable to Lady Wood because there is a continuing input of activity from Sellafield but it does provide some idea of the timescale for radionuclide movement following deposition to the canopy. There may also be seasonal effects which were not recorded during these aerosol experiments. Also, the effects of the two components in the retention curves means that, in the short term, deposited material is absorbed into the leaves, or washed off during rainfall. In the longer term, the incorporated radionuclides are leached out from the leaves, giving rise to the long term availability of the radionuclides to the forest floor where they may subsequently be

taken up into plants and higher trophic levels. Alternatively, rainfall may continue to leach the radionuclides from the leaf litter layer, facilitating their penetration into the soil.

Following the absorption of radionuclides into the leaf, translocation within the trees can occur. Isotopes of caesium, in particular, are readily translocated along with potassium and it has been indicated that caesium accumulates in the wood before the onset of winter. The translocation of radionuclides differs depending upon which nuclide is involved. It is expected, for example, that the actinides will tend to accumulate in the older tissue prior to shedding of the leaves because the actinides are not utilised by the tree nor are they analogous to any other metabolically important ions and are therefore effectively a waste product to be discarded.

In this study no attempt was made to examine the activity on, or in, the leaves whilst they remained in the canopy nor were the trees analysed to determine the presence and/or distribution of the radionuclides. It was assumed that the radionuclide contamination of interest, as regards food chain transfer, was that actually deposited to the leaf litter and upper soil layers.

3.5.2 Decomposition of leaf litter

It was also assumed that, because of the limited vegetation growth, the primary transfer of radionuclides into the food chains within Lady Wood occurs as a result of decomposition of the leaf litter layer. Much of the invertebrate and small mammal activity that occurs within the wood revolves around the leaf litter. The majority of invertebrate species caught in pitfall traps (section 3.7) belong to either predatory or detritivore groups. Obviously the latter are involved with decomposition and will ingest radionuclides either bound to, or within, the leaf litter. This forms a direct route for the inclusion of radionuclides within food chains. In addition to invertebrate detritivores, soil fungi (basidiomycetes) and other micro-organisms play a very important role in the decomposition of leaf litter. Rommelt *et al.* (1990) and Yoshida and Muramatsu (1994) described the transfer of ^{134}Cs, ^{137}Cs and ^{90}Sr from leaf litter to basidiomycetes within forest ecosystems and reported a rapid uptake of radiocaesium into the fruiting bodies, providing another direct route for the transfer of caesium through to higher trophic levels. Recent measurements of ^{137}Cs and ^{241}Am activity in the fruiting bodies of fungi collected in October 1995 showed a range in activities from 100 to 2,500 Bq kg^{-1} for ^{137}Cs and <1 to 15 Bq kg^{-1} for ^{241}Am depending upon the species involved (Toal, pers. comm.).

Clint *et al.* (1990) investigated the role of microbes and their effects on the release of ^{137}Cs from plant litter, although the study utilised heather plants, *Calluna vulgaris*, rather than

woodland leaf litter. However, the principles should be similar to the breakdown of Lady Wood leaf litter because heather produces a standing crop of litter which contains many nutrients for new plant growth and, as in Lady Wood, the nutrients will be removed from the litter by leaching and decompositional processes. One of the main points raised by Clint *et al.* (1990) was that the micro-organisms themselves can accumulate the radionuclides, thus retaining them within the food chain. Within Lady Wood, much of the decomposition is carried out through the growth of fungi, particularly those of the Actinomycete and Basidiomycete families, since these are better able to survive the low pH. The importance of the retention of radionuclides by those species undertaking decomposition should not be underestimated, for example Olsen *et al.* (1990) estimated that up to 30% of the ^{137}Cs present within the soil may actually be located within fungal mycelia. Witkamp and Barzansky (1968) also suggested that caesium may be immobilised by fungi. This is somewhat dependent upon the actual fungal species, for example Grueter (1971) found selective enrichment of ^{137}Cs within the fruiting bodies of different basidiomycete species and this was later attributed to different feeding habits or depths to which the mycelia penetrate (Randa *et al.*, 1990). What is important for Lady Wood is that fruiting bodies of fungi are used as an integral part of the diet of small mammals and some invertebrate species. Bakken and Olsen (1990) showed that where the caesium was translocated within the fungi to the fruiting bodies, uptake to higher organisms was possible. Consequently, the leaf litter forms an integral part of the woodland ecosystem and, particularly within Lady Wood.This is because the limited vegetation growth due to the dense tree canopy, means that litter is a major component in the accumulation, recycling and transfer of radionuclides through the woodland food chains.

3.5.3 Spatial and Temporal Variation

As with the soils, it is important to understand the spatial and temporal variation in the leaf litter activity of each of the nuclides, ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am, in order to understand how the radionuclides may affect the higher trophic levels of a food chain. However, unlike the soils, it was expected that the leaf litter would show temporal variation over the course of the study. Samples were therefore collected initially on a monthly and then a bi-monthly basis. Samples were collected along the same transects as soils and a similar spatial distribution of radionuclides was hypothesised.

Mean activities (n=2) and standard deviation for the samples are presented in Figures 3.12 to 3.16. Figure 3.12 presents ^{134}Cs data. However, because Sellafield only releases small quantities of ^{134}Cs during routine operations and these are mainly to the marine environment (section 1.3.1), concentrations were difficult to measure within the counting time available. Consequently, the raw data contain several samples whose activities are reported as being less than the limit of detection (LOD). These data have been used in the presentation of the results

Figure 3.12: ^{134}Cs **Activity in leaf litter samples - temporal and spatial variation.**

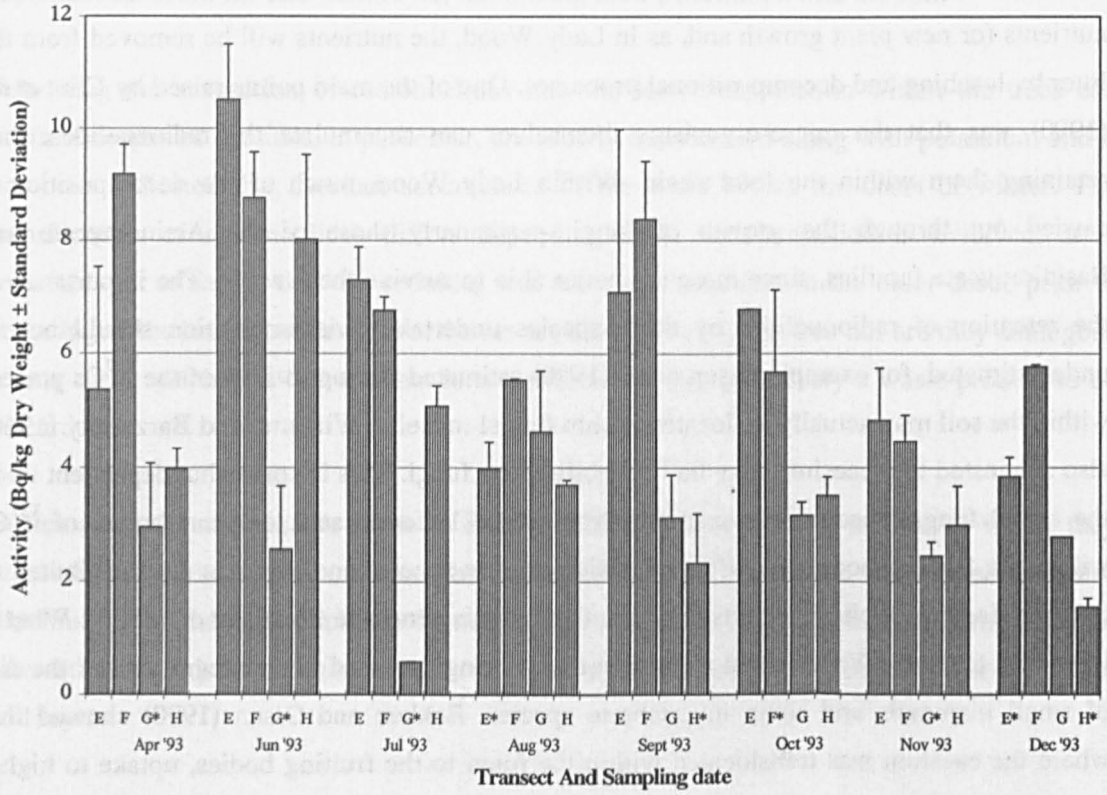
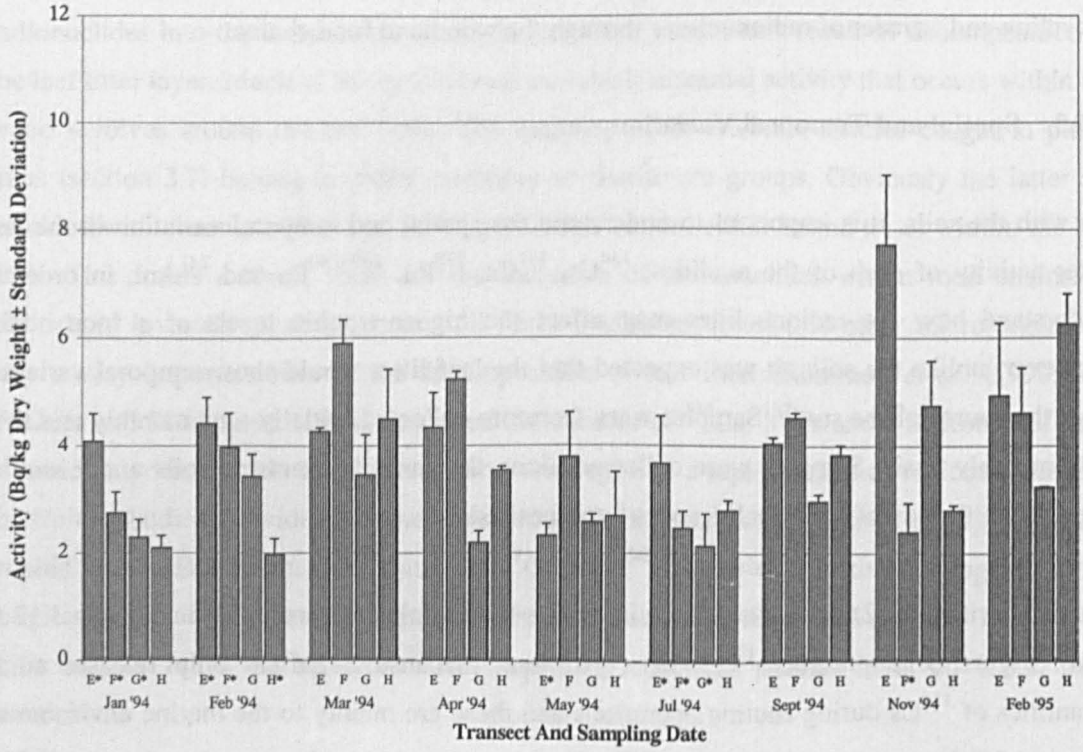


Figure 3.12: **(Continued.)**



* Indicates where activities below the limit of detection have been used in the calculation of the mean (see text).

Figure 3.13: ^{137}Cs Activity in leaf litter samples - temporal and spatial variation.

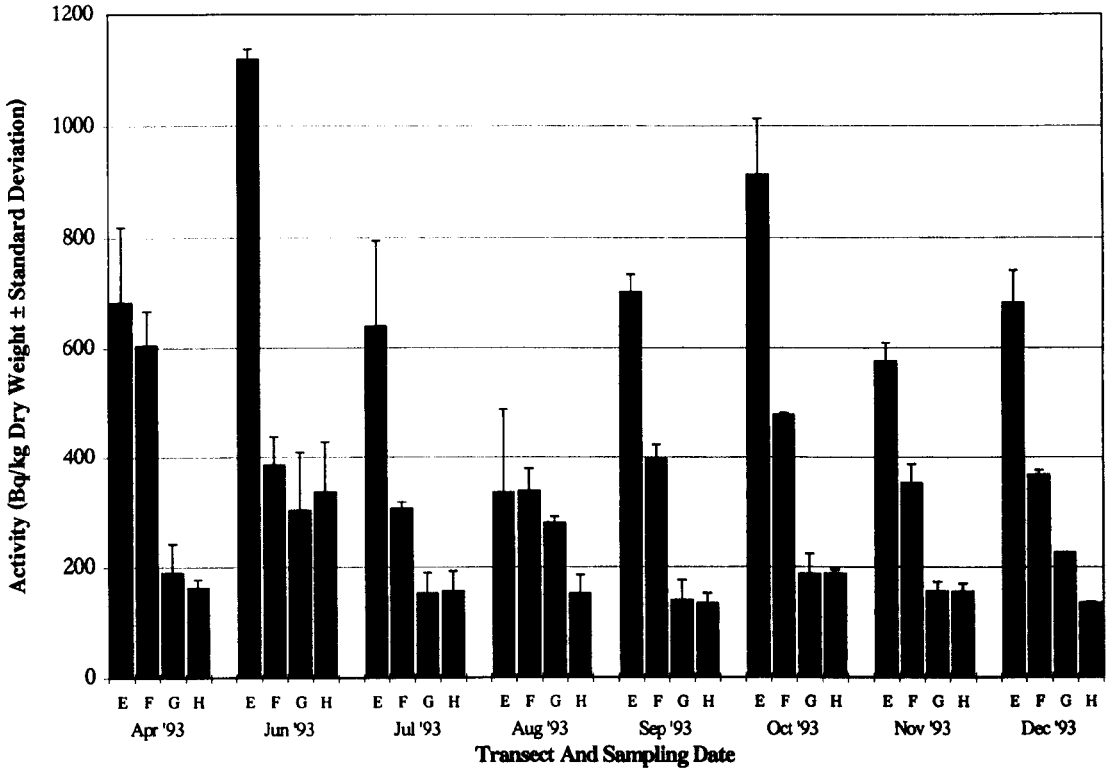


Figure 3.13: (Continued.)

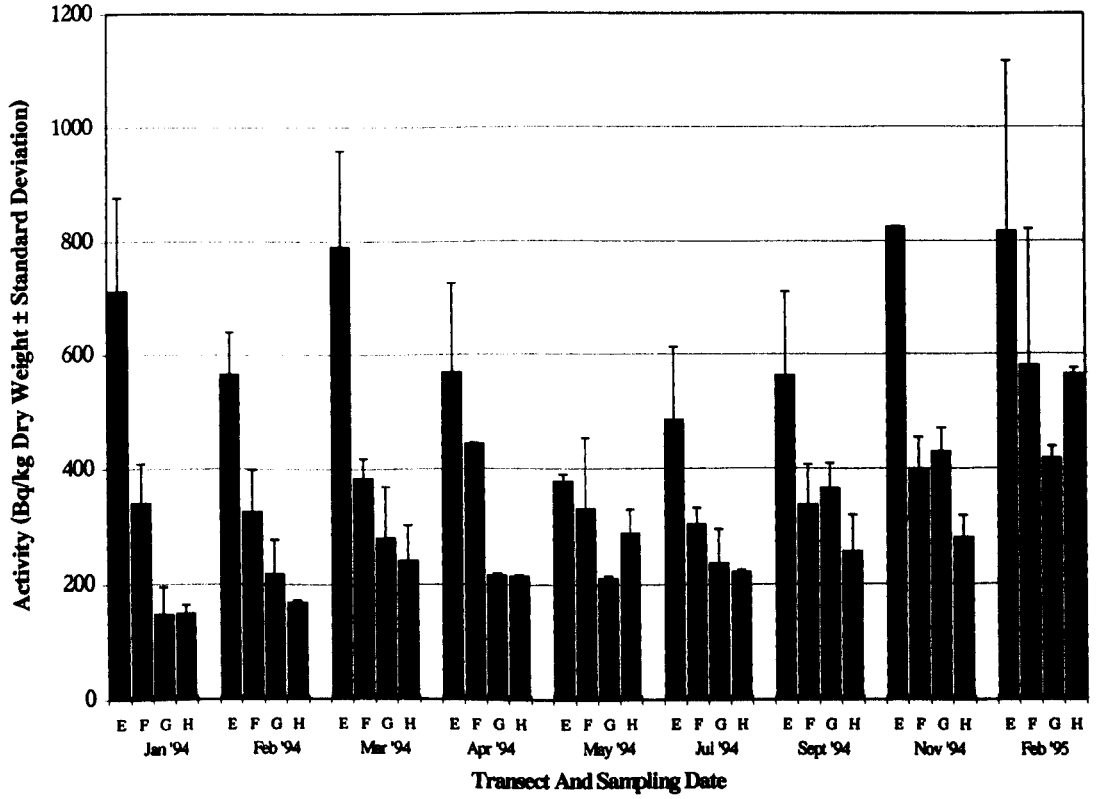


Figure 3.14: ^{238}Pu Activity in leaf litter samples - temporal and spatial variation.

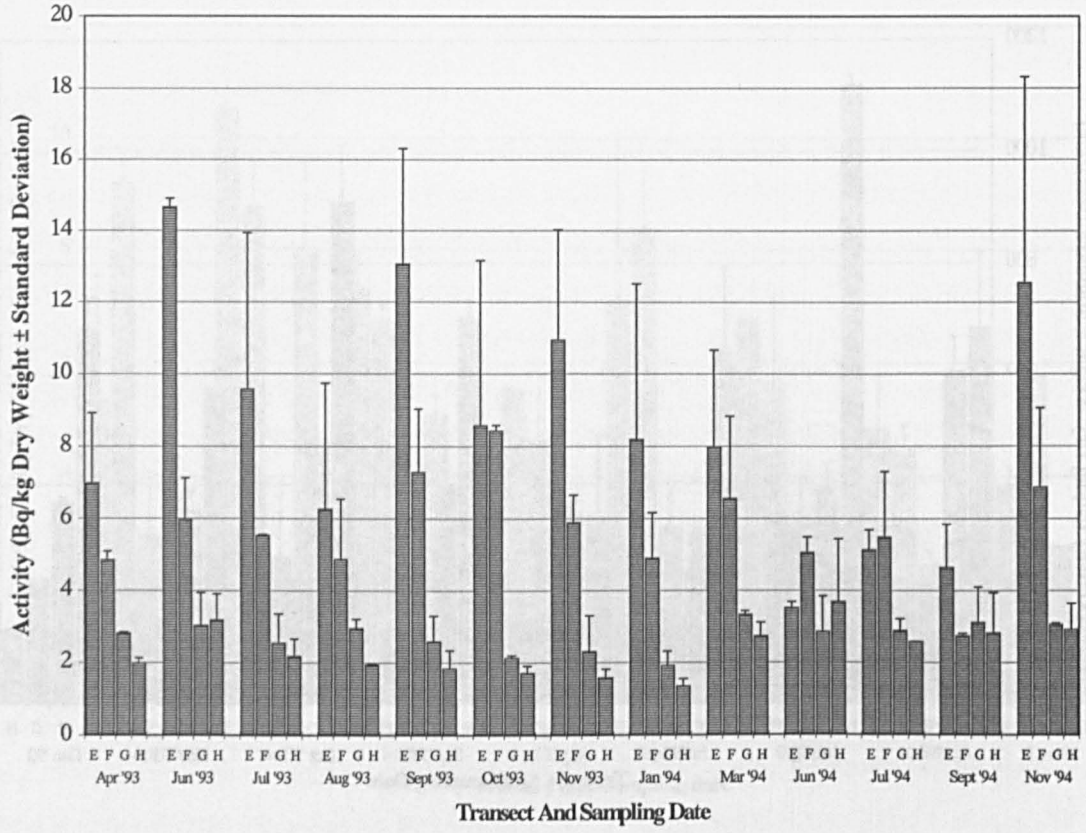


Figure 3.15: $^{239+240}$Pu Activity in leaf litter samples - temporal and spatial variation.

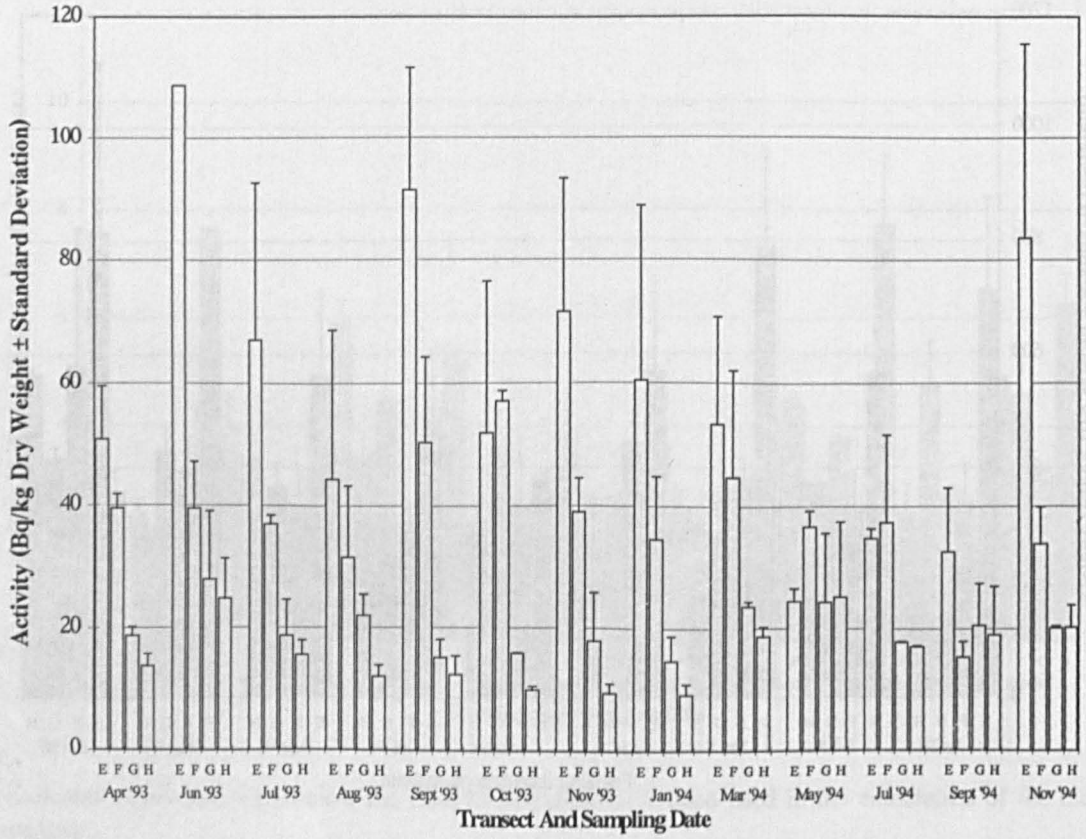


Figure 3.16: ^{241}Am Activity in leaf litter samples - temporal and spatial variation.

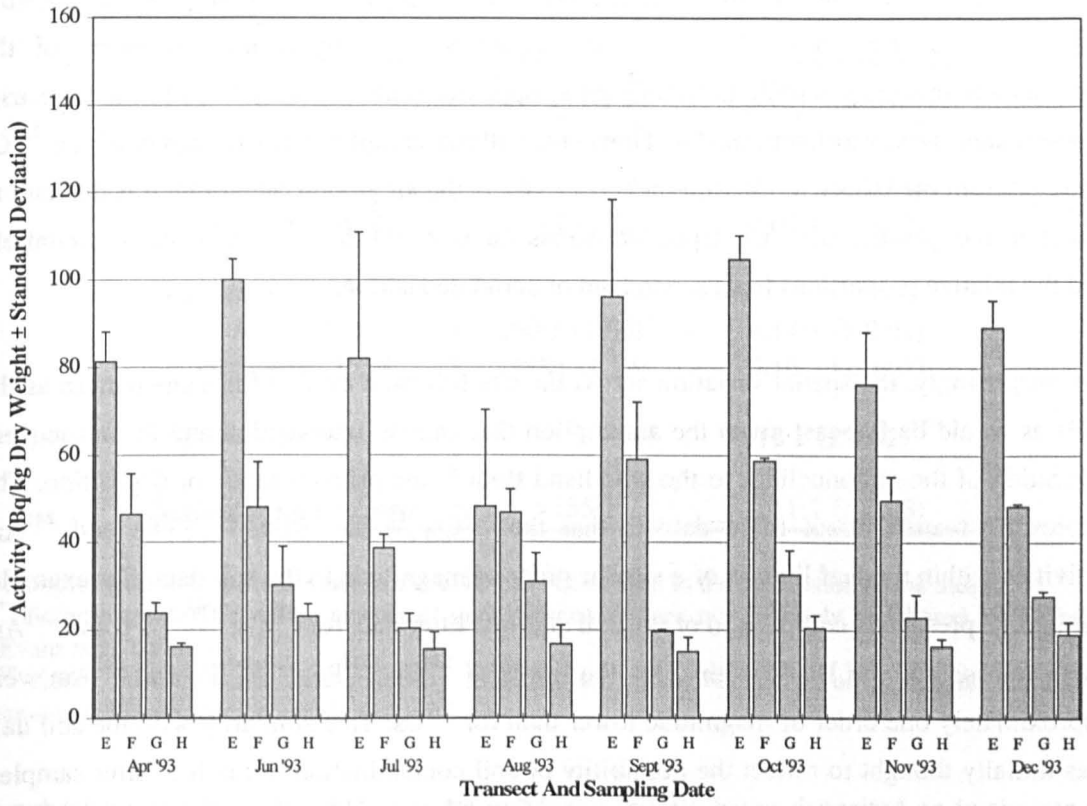
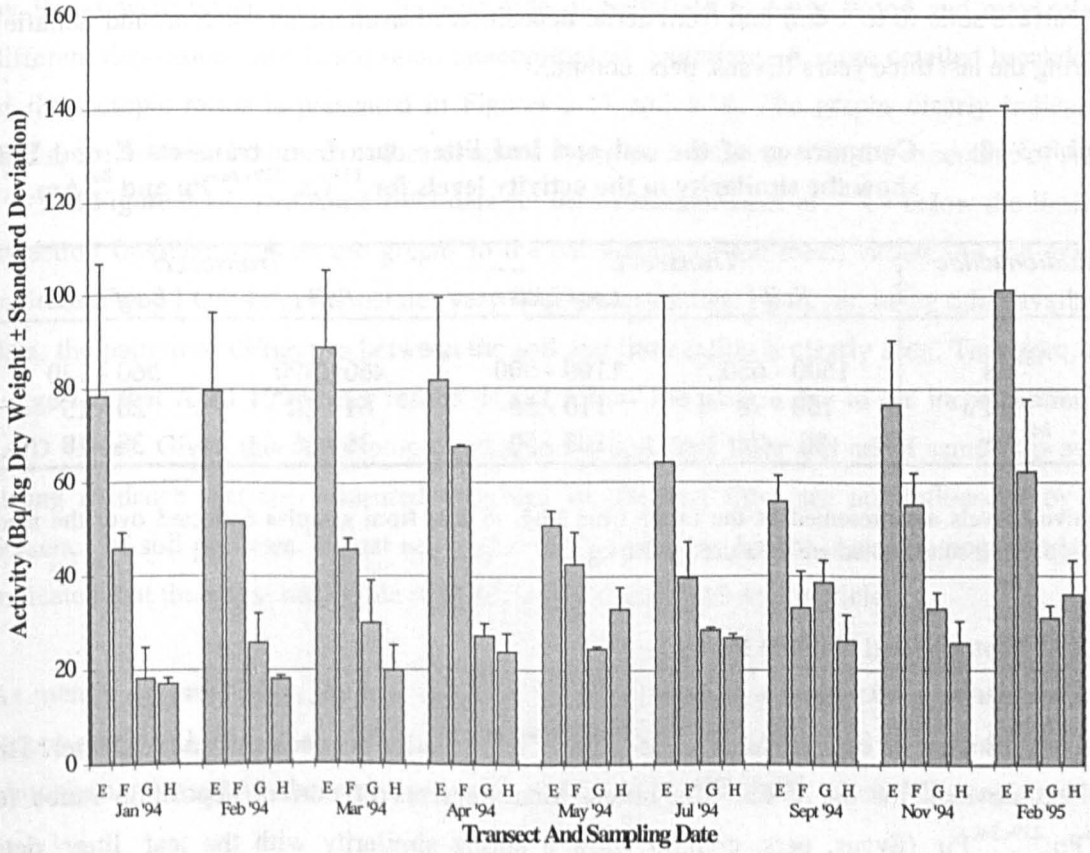


Figure 3.16: (Continued.)



but it should be noted that those samples marked with * are conservative estimates of the reported activity. Because of the effectiveness of the canopy interception, it is still possible that the ^{134}Cs released to the atmosphere could form a significant component of the radionuclide inventory within the woodland as opposed to the return of ^{134}Cs to the terrestrial environment by sea to land transfer. However, without complete measurements of the ^{134}Cs released from the stacks, or measurements of ^{134}Cs in the air around Sellafield, it is difficult to ascertain the quantity of ^{134}Cs deposited across Lady Wood that is attributable to Sellafield and the relative proportions from sea to land or aerial deposition.

Not surprisingly, the spatial variation across the site followed exactly the same pattern as the soils as would be forecast given the assumption that canopy interception and the subsequent relocation of the radionuclides to the woodland floor is the primary mode of deposition. The interesting feature about these data is that the ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities within the leaf litter is of a similar order of magnitude to the soil data. For example, Table 3.12 presents a comparison of the soil and leaf litter data for ^{137}Cs, $^{239+240}Pu$ and ^{241}Am along transects E and H. As with soils, the levels of ^{134}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am were approximately one order of magnitude lower than for ^{137}Cs. This similarity with the soil data was initially thought to reflect the possibility of soil contamination in the leaf litter samples. However, by comparing the isotopic ratios for ^{137}Cs:^{134}Cs and ^{238}Pu:$^{239+240}Pu$ (Table 3.13) for soils and leaf litter, it is clear that the soil contains material of a different composition to that of the leaf litter. For comparison, Table 3.13 also contains unpublished data on isotopic ratios in surface soils (0 to 1 cm) and from aerial deposition measurements taken around Sellafield during the last three years (Evans, pers. comm.).

Table 3.12: Comparison of the soil and leaf litter data from transects E and H to show the similarity in the activity levels for ^{137}Cs, $^{239+240}Pu$ and ^{241}Am.

Radionuclide	*Transect E*		*Transect H*	
	Soil	*Leaf Litter*	*Soil*	*Leaf Litter*
^{137}Cs	1500 - 650	1100 - 300	460 - 390	560 - 130
$^{239+240}Pu$	160 - 98	110 - 25	61 - 50	20 - 15
^{241}Am	86 - 45	105 - 50	30 - 15	38 - 18

Activity levels are presented as the range from high to low from samples collected over the study period. All results quoted are measured in Bq kg^{-1}.

3.5.3.1 Isotopic and Nuclide Ratios

Table 3.13 shows a clear difference for ^{238}Pu:$^{239+240}Pu$ ratios between soil and leaf litter. This is less obvious for the ^{137}Cs:^{134}Cs ratios. Measurements of current deposition ratios for ^{238}Pu:$^{239+240}Pu$ (Evans, pers. comm.) show a strong similarity with the leaf litter data,

Table 3.13: **Comparison of average isotopic ratios for April 1993 and September 1994 soil and leaf litter samples across the study site (± standard error), n=8.**

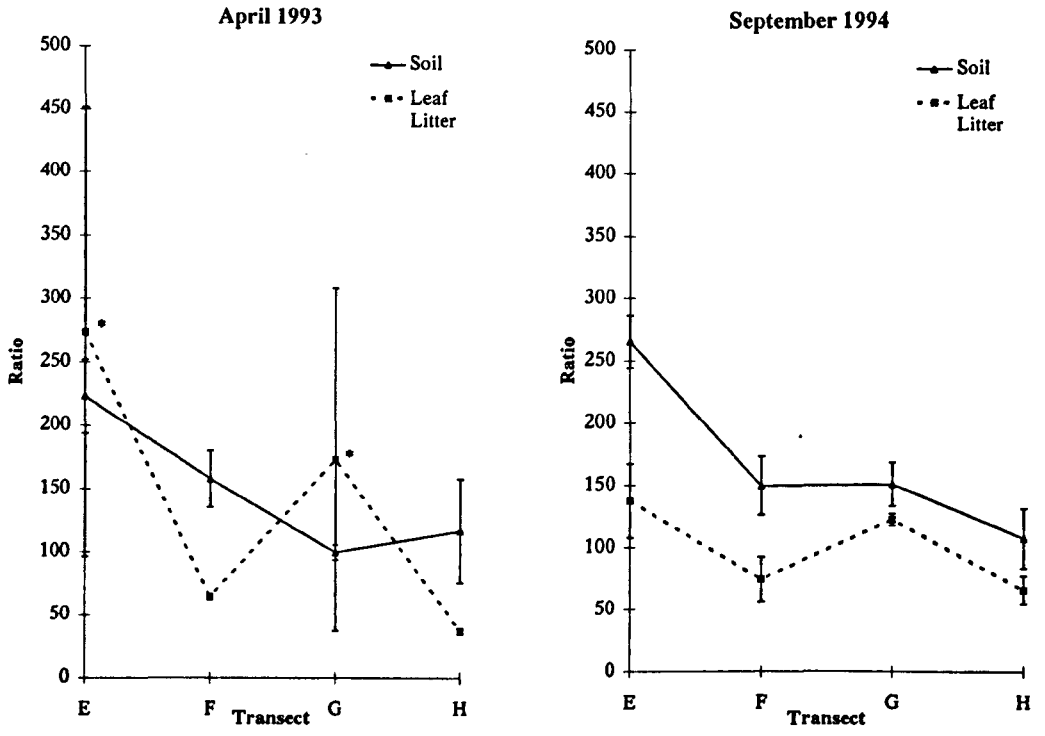
	Isotopic Ratios	
	^{137}Cs:^{134}Cs	^{238}Pu:$^{239+240}Pu$
Soil:		
April 1993	149:1 (± 19)	0.06:1 (± 0.01)
September 1994	169:1 (± 20)	0.07:1 (± 0.01)
Leaf Litter:		
April 1993	160:1 (± 80)*	0.14:1 (± 0.01)
September 1994	100:1 (± 14)	0.16:1 (± 0.01)
Deposition Samples	**	0.16:1 (± 0.02)***
Surface Soils (0-1 cm)	>155:1	0.11:1 (± 0.03)

* Samples with ^{134}Cs activities below the LOD have been used in the calculation of the mean.
** Unfortunately ^{134}Cs measurements of aerial discharges are not routinely monitored by Sellafield (Evans pers. comm.).
*** Aerial measurements were made by assessing the direct deposition of particulate material to containers (Evans, pers. comm.), n=4.

emphasising the difference between the material currently being deposited on to the canopy and the activity in the soil. However, higher surface soil ratios were measured by Evans and may reflect differences in sampling location and methods. For example, the samples collected by Evans were taken from the opposite side of Sellafield to Lady Wood and may reflect different deposition rates based upon meteorological conditions. A more detailed breakdown of the isotopic ratios is presented in Figures 3.17 and 3.18. The graphs clearly indicate a difference between both ratio values obtained along each transect with the exception of April 1993 in Figure 3.17. The April 1993 data set uses measurements of ^{134}Cs below the limit of detection (marked by * on the graph) in the calculation of the mean values. As the graphs indicate, using LOD values generates very large uncertainties. However, using other available data, the pattern of difference between the soil and litter ratios is clearly seen. Therefore it is suggested that April 1993 litter results do not follow the pattern due to the incorporation of LOD values. Given this, the isotopic ratios in the soil, leaf litter and aerial samples provide strong evidence that the measured activities for the leaf litter are not influenced by the presence of soil particles. Visual inspection of the leaf litter before it was homogenised also indicated that there was negligible surface contamination with soil particles.

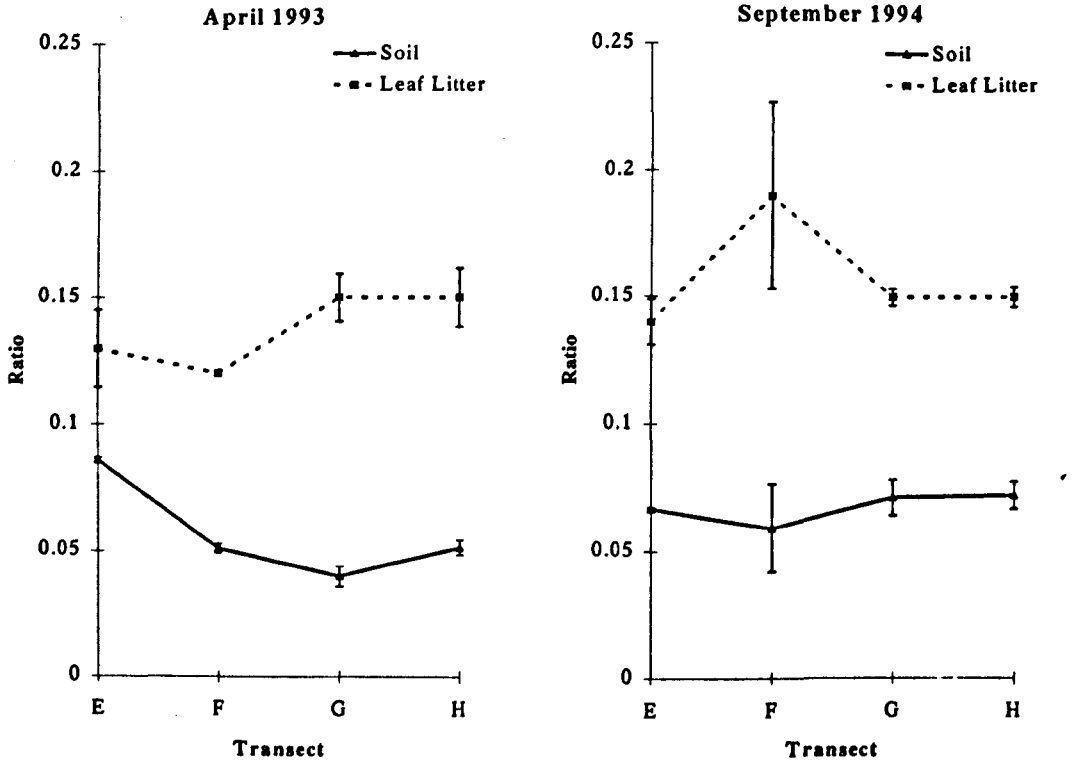
As mentioned previously, the low soil ^{238}Pu:$^{239+240}Pu$ ratio of around 0.06 is consistent with recent studies (Jones *et al.*, 1996; McCarthy and Nicholls, 1990) and is representative of plutonium released in the early years of operation at Sellafield, then Windscale, when

Figure 3.17: ^{137}Cs:^{134}Cs Ratio across Lady Wood for soil and leaf litter samples.



* Indicates where activities below the limit of detection have been used during calculations (see text).

Figure 3.18: ^{238}Pu:$^{239+240}Pu$ Ratio across Lady Wood for soil and leaf litter samples.

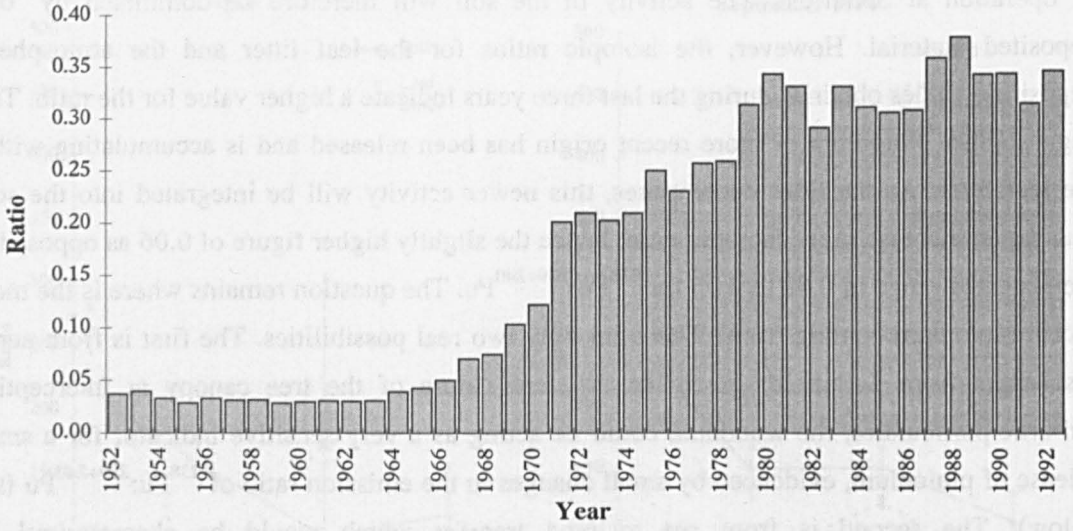


uranium of low irradiation was being processed. However, it is possible that the ratio is starting to increase. The soil will have been integrating the plutonium released over the period of operation at Sellafield. The activity in the soil will therefore be dominated by 'old' deposited material. However, the isotopic ratios for the leaf litter and the atmospheric deposition values obtained during the last three years indicate a higher value for the ratio. This suggests that plutonium of more recent origin has been released and is accumulating within the leaf litter. As the litter decomposes, this newer activity will be integrated into the soil, causing an increase in the isotopic ratio; hence the slightly higher figure of 0.06 as opposed to the expected range of 0.02 to 0.05 for ^{238}Pu:$^{239+240}Pu$. The question remains where is the more recent plutonium coming from? There are only two real possibilities. The first is from aerial discharges from Sellafield; given the efficient nature of the tree canopy at intercepting airborne particulates, the woodland could be acting as a very sensitive indicator for a small release of plutonium, evidenced by small changes in the emission ratio of ^{238}Pu:$^{239+240}Pu$ (see below). The second is from sea to land transfer which would be characterised by ^{238}Pu:$^{239+240}Pu$ ratio of about 0.2, reflecting the ratio currently observed in intertidal and nearshore marine sediments near Sellafield (BNFL, 1995).

Measurements of plutonium released to the atmosphere from Sellafield have shown a dramatic decline in activity over the last 40 years (section 1.3.1). Unfortunately, the stack emission measurements only monitor total plutonium alpha releases. However, measurements on the marine discharge do show changes in the plutonium activity and composition over time (section 1.3.1). Figure 3.19 presents the change in the ^{238}Pu:$^{239+240}Pu$ ratio over the last 40 years, using data from BNFL (1972-1995; Gray *et al.*, 1995). The ratio has increased from 0.03 to around 0.35. Although the ^{238}Pu:$^{239+240}Pu$ ratio in atmospheric discharges is not routinely measured it is likely to follow a similar trend to liquid emissions, i.e. be greater than 0.1. Consequently it is suggested that the 'current' plutonium deposition to the canopy originates from the aerial releases from Sellafield, especially since the ^{137}Cs:^{134}Cs ratio (Table 3.13) also indicates that material of a different nuclide composition is currently being deposited to the canopy/leaf litter when compared to that recorded for the forest soil.

The other question which remains is why the leaf litter and soil activities are of a similar order of magnitude for each radionuclide? There is still the possibility that a small amount of soil contamination is present on the leaf litter, although this is very remote. Measurements of the activity on leaf canopy samples would have completed the story and may have shed light on this problem. It is suggested that the canopy acts as a secondary sink for the radionuclides, similar in terms of its inventory to that of the soil, and that these two systems are possibly in a state of equilibrium. Unfortunately no canopy or tree samples were taken during the course of the study which would provide evidence of any radionuclide recycling through translocation from the soil to the canopy. It is hoped to rectify this in the future work.

Figure 3.19: The change in ^{238}Pu:$^{239+240}Pu$ ratio in the marine discharges from Sellafield over time (BNFL, 1972-1995; Gray *et al.*, 1995).



3.5.3.2 ANOVA Results

A balanced two factor Analysis of Variance was used to examine the activities of each of the radionuclides for statistically significant differences in temporal and spatial variation. The ANOVA results are presented in Table 3.14 for ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am respectively. For significant results a 5% LSD was calculated using Bonferroni's method and the results are also reported in Table 3.14.

Analysis of variance tests on the leaf litter data confirms that a similar spatial distribution for ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am to that observed in the soil with the greatest concentrations present along the front edge of the woodland (relative to Sellafield). With the exception of ^{134}Cs, this decline is significant for the transects as follows: E > F > G and H ($p<0.001$). ^{134}Cs results do not exhibit a significant difference between E and F ($p>0.05$). It was expected that for all the radionuclides, except ^{134}Cs, that the spatial distribution pattern would be similar to that of the soils. The results are therefore in agreement.

It was anticipated that the majority of ^{134}Cs present in the soil, was deposited following the Chernobyl accident and is consequently of different origin to the ^{134}Cs detected in the leaf litter. Furthermore, almost ten years have passed since the accident. Consequently, the leaves in the canopy will have been completely replaced at least twice, and the material deposited with the leaf litter will have undergone considerable weathering. The ^{134}Cs present in the canopy and measured on the leaf litter during this study will have been released from Sellafield, either to the atmosphere or to the marine environment. Consequently, a distribution pattern similar to the other four radionuclides was anticipated, because of the effects of dry

Table 3.14: Results from a balanced two factor Analysis of Variance. Test for spatial and temporal changes in leaf litter ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities respectively.

^{134}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Month	16	173.518	10.845	3.27	0.000***	2.64
Transect	3	141.487	47.162	14.24	0.000***	1.13
Month*Transect	48	221.405	4.613	1.39	0.104	
Error	68	225.192	3.312			
Total	135	76.601				

^{137}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Month	16	1013544	63346	4.65	0.000***	169.24
Transect	3	4279481	1426494	104.81	0.000***	72.59
Month*Transect	48	1056850	22018	1.62	0.034*	299.30
Error	68	925507	13610			
Total	135	7275381				

^{238}Pu:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Month	16	110.335	9.195	1.39	0.200	
Transect	3	668.308	222.769	33.72	0.000***	1.91
Month*Transect	48	234.357	6.510	0.99	0.512	
Error	68	343.518	6.606			
Total	135	1356.518				

$^{239+240}Pu$:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Month	16	5337.1	444.8	1.69	0.097	
Transect	3	31211.0	10403.7	39.44	0.000***	12.09
Month*Transect	48	12428.5	345.2	1.31	0.185	
Error	68	13717.1	263.8			
Total	135	62693.8				

^{241}Am:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Month	16	4850.2	303.1	1.23	0.272	
Transect	3	70652.5	23550.8	95.22	0.000***	9.79
Month*Transect	48	9123.9	190.1	0.77	0.831	
Error	68	16818.4	247.3			
Total	135	101444.9				

deposition from the point source of Sellafield. This is seen in the ^{134}Cs results because the activity measured along transects E and F is greater than that of G and H.

3.5.3.3 Radionuclide movement

Detailed sampling and analysis, of the deposition and relocation mechanisms was not considered appropriate for this study. However several studies have investigated the relocation of radionuclides from the canopy to soil and have shown leaching, stemflow, and leaf fall to be the main pathways for radionuclides to be transferred to the forest floor. There are a number of factors which will affect radionuclide deposition, not least the age, spatial distribution, canopy structure, edge effects (Draaijers *et al.*, 1992), and the uptake/recycling in trees of radionuclides deposited to the forest floor (Murphy and Tuckfield, 1994). As a consequence, it has been suggested that radionuclides reaching the forest floor might not be uniform (Beier *et al.*, 1993; Franklin *et al.*, 1967). Primarily, this is a result of non-uniform dry deposition arising from the heterogeneity of the forest canopy (Guillitte *et al.*, 1990).

During rainfall, radionuclides may be deposited directly to the forest floor, a process known as throughfall. The quantity will be dependent upon the canopy structure. Measurements made of throughfall under a forest canopy will however contain a proportion of material which has been leached from the leaves, particularly if they are present in a water soluble form, during the rainfall (Hansen *et al.*, 1994). Leaching can occur in two ways, firstly the direct wash off of the particulate material to lower sections of the canopy and ultimately the forest floor, and secondly, water may accumulate and then run across the leaves, on to the twigs, branches and trunk of the tree, leading to a washing out effect of the radionuclide deposition around the bases of the tree stems, so-called stemflow. This generally leads to a pattern of distribution whereby concentrations of radionuclides around the base of the trunk are greater than those measured under the peripheral canopy (Gersper, 1970; Franklin *et al.*, 1967).

In addition to leaching and stemflow, leaf litter fall is an important transfer route for the movement of radionuclides from the canopy to the forest floor (Bonnett and Anderson, 1993). Moreover, studies have shown an enhanced radionuclide uptake into tree and vegetation species within a woodland because of the characteristically high organic matter content within forest soils. This is related to the high availability of the radionuclides which are adsorbed to organic matter either directly to plants through root uptake (Barber, 1964; Cremers *et al.*, 1990; Russell, 1966; Valcke and Cremers, 1994), or facilitated through the action of micro-organisms present within the soil (Clint *et al.*, 1990). Murphy and Tuckfield (1994) suggested that this uptake and the subsequent translocation of radionuclides from soil to leaves may form a significant route by which radionuclides are returned from the soil to the forest floor.

For the current study site, however, the canopy structure is uniform and dense with very few openings in the canopy. Most of the trees are no more than 2 m apart and the canopies are heavily intertwined. Therefore, it is suggested that direct throughfall to the forest floor is rare

and that, comparatively, leaching and stemflow will be more important. Moreover, because of the density of the stand, it is further suggested that the effects of stemflow on the radionuclide distribution in the soil will be moderated.

The leaf litter activity for the actinides remained reasonably consistent (insignificant at $p<0.05$) throughout the sampling period, although the caesium isotopes did exhibit significant differences in activity over the two year period. Nevertheless, Witkamp and Frank (1967) showed that, after three months of simulated rainfall, more than 80% of the ^{137}Cs remained adsorbed to the leaf litter fraction. Further investigation of the changes in caesium activities was attempted to explore possible reasons for the variation observed. However for ^{137}Cs, plotting the total activity across each transect (Figure 3.13) showed that only three sampling occasions (June 1993, November 1994 and February 1995) showed a significant difference from the general pattern when compared to the 5% LSD value. Climatic data were mapped, particularly monthly rainfall, to investigate these changes. It was hoped to assess whether changes in the monthly rainfall, or periods of dry conditions prior to rainfall, had any impact upon the litter values, either directly or indirectly through changes in leaf fall rates or increased leaching from the canopy. It was believed that during the summer period, greater amounts of dry deposition would occur to the leaf canopy. Subsequent heavy rainfall would then relocate the radionuclides to the forest floor and a greater proportion of the radionuclides would be leached from the canopy as a consequence of previously high periods of deposition to the canopy. Thus greater amounts of radioactivity would be seen during those months with higher rainfall, particularly following previously dry months. However, a poor fit to the data was obtained using rainfall as a measure of climatic conditions (using monthly summaries of the climate in Cumbria). It is therefore suggested that for ^{137}Cs the changes in activity are random, as with the actinide data, especially since climatic conditions would have affected all the radionuclides more or less equally.

3.5.3.4 Temporal variation of ^{134}Cs

The ANOVA results for ^{134}Cs also indicate that there is a strong difference in the temporal data for leaf litter. Again, after examination of the 5% LSD, it appears that only one month, June 1993, is significantly ($p<0.001$) different from the other months. As with the ^{137}Cs data, this suggests that there is little evidence for temporal variation during the study period. As mentioned earlier, it was expected that temporal variation would be seen in the data. However, the results indicate that this is not the case. It is probable that this is due to the lifespan of the needles within the canopy which encompasses several climatic seasons. Most of the needles on coniferous tree species survive for about two years and for *P. sitchensis* it can vary from 1 to 4 years, depending upon climatic and biotic conditions. For example, Bonnett and Anderson (1993) described the effect of aphid infestation upon premature leaf fall with regard

to the storage of ^{137}Cs in the canopy. Assuming that the leaves survive for more than a year, they will be accumulating airborne particulates over a number of seasons and they will be subject to a range of climatic conditions. Furthermore, pine needles fall continuously throughout the year. At any given point in time, the leaf litter will contain leaves of different ages and therefore different amounts of radionuclides adhered to the leaf surface. Moreover, the litter will vary in the degree of decomposition which also may be influenced by climatic conditions. Thus over the sampling period for this study, radionuclides deposited over a period of up to four years could be being relocated from the canopy and mixed with a leaf litter layer of a similar or older age. As a result, this will have a moderating impact on the activity of ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am within the leaf litter and prevent any trends in short term temporal variation from being seen. In order to assess short term (over one or two years) a more detailed sampling strategy would be required, one that samples the inventory within the canopy and in recent leaf fall.

3.5.4 Comparison of Leaf Litter Samples Collected in Cheshire to those from Lady Wood

Table 3.15 compares the activity of leaf litter samples collected in Lady Wood and Cheshire. The data clearly show, just as with the soils, elevated levels in Lady Wood compared to the Cheshire site. Results of a One-way Analysis of Variance and the calculated 5% LSD value show that the differences are statistically significant ($p<0.05$). The ANOVA output is shown in Table 3.16. This is in accordance with the anticipated results.

In general, the data obtained for Lady Wood are greater by two orders of magnitude except for ^{134}Cs which is only one. For example, the ^{137}Cs results are between 50 to 160 times higher at Lady Wood, depending upon which transect is used.

Table 3.15: A comparison of the ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am mean results from Lady Wood and Cheshire (data taken from samples collected during August 1993).

Transect / Site	^{134}Cs	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
E	4.9 ± 1.7	490 ± 180	8.8 ± 3.3	60 ± 21	70 ± 25
F	5.8 ± 5.8	320 ± 28	5.0 ± 1.0	33 ± 7	43 ± 5
G	5.2 ± 0.7	280 ± 7	3.0 ± 0.2	24 ± 2.4	33 ± 4.3
H	4.4 ± 0.7	160 ± 25	2.2 ± 0.2	14 ± 2.2	18 ± 2.7
Cheshire	0.22 ± 0.02	3.1 ± 1.5	0.02 ± 0.01	0.06 ± 0.01	0.23 ± 0.04

Results expressed as Bq kg^{-1} (dry weight) (± standard error, n=3) and reported to two significant figures.

These data demonstrate clear similarities to the soils (section 3.4.3) and provide strong evidence of Sellafield's role in the marked increase in the activity of ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am compared to the background levels produced as a consequence of the global release of anthropogenic radionuclides.

Table 3.16: Results for ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am from a One-way Analysis of Variance on Lady Wood and Cheshire leaf litter samples.

^{134}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	4	59.47	14.87	6.41	0.008**	1.76
Error	10	23.19	2.32			
Total	14	82.66				

^{137}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	4	395979	98995	5.12	0.01**	160.92
Error	10	193448	19345			
Total	14	589427				

^{238}Pu:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	4	132.83	33.21	4.74	0.021*	3.06
Error	10	70.08	7.01			
Total	14	202.91				

$^{239+240}Pu$:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	4	6158	1540	5.01	0.018*	5.11
Error	10	3074	307			
Total	14	9232				

^{241}Am:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	4	8142	2035	5.22	0.016*	22.85
Error	10	3897	390			
Total	14	12039				

3.5.5 Summary of Leaf Litter Results

Pine needle litter in Lady Wood shows elevated levels of all radionuclides measured, ranging from 2 to 11 Bq kg^{-1} (^{134}Cs), 130 to 1,100 Bq kg^{-1} (^{137}Cs), 2 to 15 Bq kg^{-1} (^{238}Pu), 15 to 110 Bq kg^{-1} ($^{239+240}Pu$), and 18 to 105 Bq kg^{-1} (^{241}Am). Given the hypothesis that much of the deposition to soils is historic, concentrations are surprisingly high in relation to underlying soils, the ratio between leaf litter and soil concentrations typically lying between 0.3 and 1.0.

The elevated concentrations in needle litter cannot be explained by adventitious soil contamination, and there are significant differences between isotopic ratios of ^{134}Cs:^{137}Cs and ^{238}Pu:$^{239+240}Pu$ which imply that contamination on the needle litter is of more recent origin than soils.

The relative levels of contamination on needle litter and soil cannot fully be explained, although some researchers have suggested that mechanisms involving fungal mycelia can lead to re-concentration of material into the litter layer.

Concentrations in needle litter show the same spatial variation as concentration in soils, with a fall in concentrations from the 'front' to 'back' edge of the wood, confirming that interception of aerosols by the woodland canopy is a dominant mechanism in deposition processes at the site.

3.6 VEGETATION RESULTS

The mix of grass and herbaceous species found within Lady Wood has already been described (section 3.2) and is limited in diversity and biomass. Nevertheless, it was believed to be important to assess radioactivity present within/on the vegetation growing in the woodland as this will form part of the wood mouse (*Apodemus sylvaticus*) diet during the year (section 1.4.4.5). Because of the limited biomass, samples were only taken from transects E and H on three occasions during the study period when sufficient material could be collected. Samples were taken during August 1993, June and September 1994. Additional samples were obtained during May 1994 from just outside the wood (but not from the neighbouring pasture land). Table 3.17 presents the data and compares it to vegetation samples collected from a clean reference site in Cheshire in August 1993 and July 1994.

The data are limited in scope, especially because the indigenous species present were combined into a composite sample. Care is required in the interpretation of these data. Table 3.17 shows, for the most part, that vegetation sampled in Lady Wood is considerably higher in activity of each of the radionuclides, ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am, than is the case for the Cheshire site. However, the raw data are variable and a One-way Analysis of Variance (ANOVA) was undertaken to determine any significant differences between the two field sites using the data from August 1993. The results from the ANOVA and a 5% LSD calculation are presented in Table 3.18.

Table 3.17: Mean activities in mixed vegetation samples collected from Lady Wood and compared to the Cheshire site.

Site/Transect	^{134}Cs	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
August 1993					
Transect E	4.7 ± 1.4	280 ± 7.0	1.4 ± 0.1	8.3 ± 0.3	16 ± 1.8
Transect H	1.6 ± 0.5	66 ± 1.4	0.3 ± 0.1	1.4 ± 0.1	2.1 ± 0.1
June 1994					
Transect E	<1.8	182 ± 50	0.37 ± 0.08	2.0 ± 1.0	5.8 ± 3.7
Transect H	<0.9	122 ± 66	0.16 ± 0.09	0.75 ± 0.3	0.6 ± 0.4
September 1994					
Transect E	1.1 ± 0.3	133 ± 15	n/a	n/a	n/a
Transect H	1.3 ± 0.4	117 ± 17	n/a	n/a	n/a
June 1994					
By stream (1)	<0.55	9.8 ± 0.5	0.13 ± 0.07	0.6 ± 0.3	0.9 ± 0.9
By stream (2)	<0.51	120 ± 64	0.09 ± 0.04	3.3 ± 3.1	0.56 ± 0.47
Cheshire:					
August 1993	<0.25	<0.97	0.09 ± 0.05	0.04 ± 0.00	0.10 ± 0.08
July 1994	<0.12	<0.38	0.05 ± 0.01	0.07 ± 0.02	<0.38

n/a = sample not analysed.
< Limit of detection reported.
Results expressed as Bq kg^{-1} (dry weight), ± standard deviation, n=2.
By stream (1) and (2) collected from just in front of the western entrance (indicated as main entrance in Figure 3.2). There is no tree cover at either of these two locations, although stream sample number 2 is close to some larch, *L. decidua*, trees.

3.6.1 ANOVA Results

The ANOVA indicates that there are statistically significant differences within the data set. Further examination using the calculated 5% LSD values shows that this is the case for each radionuclide, with the exception of $^{239+240}Pu$ and ^{241}Am along transect H, between Lady Wood and the Cheshire site. In addition, the radionuclide activity along transect E is significantly ($p<0.01$ and $p<0.001$) greater than along transect H. For example, ^{137}Cs values from August 1993 are 280 Bq kg^{-1} and 66 Bq kg^{-1} along transects E and H respectively. Given the data on soil and leaf litter activities described previously, this spatial distribution of activity is not surprising. Interestingly, the data from the samples collected near the stream outside of Lady Wood also exhibit comparatively low activities to those samples collected along transect E, particularly for the sample taken furthest from the tree line (Table 3.17). For this sample especially, the activities measured are not significantly different ($p>0.05$) from the reference samples in Cheshire. This was unexpected but the large standard deviation recorded for the control and stream samples may be responsible. Also, vegetation collected along transect H exhibited low activities which, in the case of $^{239+240}Pu$, are comparable to the Cheshire site.

The similarities between the Cheshire site, transect H and the open canopy area near the stream (marked on Figure 3.1) may be a consequence of forming composite samples from the different species present. Alternatively, it could be due to differences in the direct foliar deposition on to the vegetation. The similarities in the data between transect H, the open canopy area and the Cheshire site, may reflect root uptake of radionuclides. Moreover, given the soil and leaf litter activity described earlier, it is likely that vegetation along transect E is particularly subject to aerial deposition. Consequently, the observed differences in vegetation activity could be due to the presence of radionuclides bound to the external surfaces of the vegetation (section 1.4.2.2).

Table 3.18: ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in a One-way Analysis of Variance on Lady Wood and Cheshire vegetation samples collected in August 1993.

^{134}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	9	25.838	2.871	6.42	0.004**	0.76
Error	10	4.474	0.447			
Total	19	30.312				

^{137}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	9	132589	14732	12.23	0.000***	39.30
Error	10	12044	1204			
Total	19	144632				

^{238}Pu:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	7	2.7170	0.38814	132.13	0.000***	0.06
Error	8	0.0235	0.00294			
Total	15	2.7405				

$^{239+240}Pu$:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	7	101.80	14.54	7.38	0.006**	1.59
Error	8	15.77	1.97			
Total	15	117.56				

^{241}Am:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Site	9	393.20	43.69	16.37	0.000***	1.85
Error	10	26.69	2.67			
Total	19	419.89				

Site refers to transects E and H, two locations close to Lady Wood near a stream which were under no tree canopy, and also the Cheshire reference site.

There appears to be a greater amount of activity for each radionuclide along transect E in August 1993. This declined during later sampling periods. This could be related to the lower activity along transect E for the soils (section 3.4.1) or may be a function of the following factors known to influence plant uptake of radionuclides: weather conditions preceding, and at the time of, sampling; variation in species composition forming the composite sample according to season; and changes in the form and structure of the species sampled. Sections 1.4.2, 4.6 and 5.6 describe in greater detail the factors that influence plant uptake of radionuclides. Sections 4.6 and 5.6 discuss them further as relevant in regard to the more complete vegetation sampling programme at the two other field sites.

The plant species present during the three sampling occasions were, not surprisingly, different. For example, bluebells, *E. scriptus,* dominated along the leading edge of the woodland during the spring months while later in the year, the vegetation cover changed to bracken, *P. aquilinum*, and grasses such as *A. capillaris, D. cespitosa,* and *F. rubra*. This caused not only changes in the growth habit and form of the cover, which affects deposition and retention of radionuclides, but different plant species also vary in their uptake of radionuclides under similar circumstances (Demirel *et al.*, 1994; Horrill *et al.*, 1990; Romney *et al.*, 1957; Sawidis, 1988; Szabo, 1979). With the data set obtained it is impossible to determine whether the species composition at the time of sampling was important within Lady Wood. Further sampling would have been required but it should be noted that the vegetation mirrors that available during the year, and this will influence the diet of those species representing the higher trophic levels, in this case, *A. sylvaticus*.

3.6.2 Concentration Ratios

In addition to the changes in vegetation, climatic conditions varied over the sampling period. Although the external and internal contribution of radionuclides to the total burden of plants was not determined in this study, direct deposition of radionuclides to the vegetation species within the understorey would surely have occurred. Therefore, species growing at different times of the year will be (a) subject to different seasonal conditions of climate and, (b) of different form and structure which can influence the deposition and retention of radionuclides on foliage (section 1.4.2.1). It is notable that changes in activity over the three sampling periods are reflected in the results obtained for both transects. This suggests that activity changes arise from a common cause. To assess whether this is related to soil, concentration ratios were calculated for the two sampling periods (Table 3.19).

Table 3.19: Concentration ratios calculated for vegetation collected from Lady Wood in August 1993 and June 1994.

Transect	*Date*	^{134}Cs	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
E	August 1993	0.87	0.28	0.17	0.08	0.32
H	August 1993	0.28	0.17	0.13	0.03	0.14
E	June 1994	<0.72*	0.28	0.06	0.02	0.13
H	June 1994	0.27	0.28	0.04	0.01	0.02

Concentration ratios calculated using mean soil core activities (Table 3.3) to a depth of 9 cm.
Soil data from June 1993 were used in the calculation for August 1993 vegetation, June 1994 data for the vegetation were compared against September 1994 soil data.
* Both the soil and vegetation data used in the calculation are < limit of detection.

For this study, concentration ratios are defined as:

$$\text{Concentration Ratio (C.R.)} = \frac{\text{Activity in vegetation (Bq kg}^{-1}\text{ dry weight)}}{\text{Activity in soil (Bq kg}^{-1}\text{ dry weight)}}$$

The activity in soil was taken from the whole core data, i.e. to a depth of 9 cm. The total activity present in the vegetation samples consists of two components, the external and internal contamination and will incorporate radionuclides transferred to the foliage via rain splash and direct aerial deposition. With the present data set, these components cannot be sub-divided.

The concentration ratios presented in Table 3.19 are higher than the ranges of 0.013 to 0.1 for ^{137}Cs; 0.00026 to 0.02 for $^{239+240}Pu$; and 0.000086 to 0.0048 for ^{241}Am, for cereal crops grown in soil contaminated only by global fallout from weapons testing (Bunzl and Kracke, 1987). It is notable that these latter transfer factors were determined where root uptake was the dominant mechanism, although some superficially bound radionuclides could be expected from rain splash. The values obtained in the present study are higher, suggesting some additional feature to the plant burden in Lady Wood. For example, there may be a higher contribution of radionuclides deposited to the plant foliage or there may be a higher root uptake of the radionuclides as a consequence of factors which influence bioavailability within the forest soil (section 1.4.2). Further research is required to confirm this.

The results indicate that there is ongoing aerial deposition across the Lady Wood site. The importance of aerial deposition in determining plant concentration ratios is argued by Rudge (1989) who studied the after-effects of Chernobyl fallout. Concentration ratios in the Sellafield area were recorded for vegetation sampled during 1987. These were higher than expected and Rudge (1989) attributed this to foliar uptake or foliar retention of radionuclides

from Chernobyl. Moreover, Jackson *et al.* (1987) reported soil to grass concentration ratios of 0.23 and 0.41 for ^{137}Cs following deposition of Chernobyl fallout, similar to the ratios reported in Table 3.19. This implies that foliar deposition of radionuclides across Lady Wood is important. Concentration ratios in Lady Wood are double those reported by Bunzl and Kracke (1987) and from several other studies (Adriano *et al.*, 1981; Coughtrey *et al.*, 1983; Romney *et al.*, 1981; Schulz and Ruggieri, 1981). It is hypothesised that foliar absorption/retention is particularly important for the actinides since, unlike ^{137}Cs, there is no active mechanism which can influence plant uptake from the soil. However, in determining the transfer of radionuclides through the food chain, the contributions of external and internal contamination are not necessarily important since both will be ingested and therefore potentially bioavailable.

It was anticipated that the concentration ratios would be similar along both transects at each sampling period if soil parameters that affect plant uptake of radionuclides were dominant in controlling the radioactivity burden. As it is, only ^{137}Cs shows any similarity between the two transects and across the two reported sampling sessions. However, there is also a strong similarity between the ^{137}Cs and ^{134}Cs results. The exception is the ^{134}Cs data from June 1994 (transect E) which is a conservative estimate of the true concentration ratios, as a consequence of using limit of detection values in the calculation. This may be related to the biological availability/uptake of radiocaesium. Because caesium is analogous to potassium there are active transport mechanisms which can encourage plant uptake (section 1.4.2.2). Furthermore, the presence of large quantities of organic matter means that there is a pool of exchangeable caesium to support root uptake. It is very probable that the uptake of caesium is dominated by root uptake and that the plants are in equilibrium with the soil. The actinides, in contrast, are subject to stable complexation with organic matter (section 1.4.2.2) and therefore less available for root uptake, in which case foliar deposition is more significant.

3.6.3 Summary of vegetation results

Only a sparse data set was obtained for the vegetation of Lady Wood, which is limited in both biodiversity and biomass. Radionuclide concentrations ranged from 1 to 5 Bq kg^{-1} (^{134}Cs), 65 to 280 Bq kg^{-1} (^{137}Cs), 0.3 to 1.5 Bq kg^{-1} (^{238}Pu), 0.8 to 8 Bq kg^{-1} ($^{239+240}Pu$) and 0.6 to 16 Bq kg^{-1} (^{241}Am). However, these broad ranges also reflect the statistically significant ($p<0.01$) spatial variation within Lady Wood as regards the soil and leaf litter data .

Radionuclide concentration ratios were determined for composite vegetation samples, since the vegetation represents the diet available during the year for herbivorous woodland fauna. The concentration ratios were higher than could be explained by root uptake alone and therefore indicate the importance of contemporary atmospheric deposition.

3.7 INVERTEBRATE RESULTS

Table 3.20 lists the major invertebrate orders and principal species caught in pitfall traps in Lady Wood. Where possible, invertebrates were separated and analysed as individual species. This is indicated in Tables 3.20, 3.21 and in the text.

Table 3.21 provides information on the seasonal abundance of invertebrates in the groups used for analysis, and indicates the biomass (as dry weight) collected. The seasonal abundance should be reflected in the diet of *A. sylvaticus* (section 1.4.4.5) so consideration of seasonal radionuclide activities is required. The contents of the pitfall traps were combined over a period of approximately four months (section 3.3). These were taken to indicate seasonal diet opportunities of *A. sylvaticus*. The sample groupings were: April 1993 to August 1993; September 1993 to February 1994; March 1994 to July 1994; and August 1994 to January 1995. The last period was extended because of very low capture rates during November and

Table 3.20: The major invertebrate orders and species caught in pitfall traps in Lady Wood.

CLASS/ SUBCLASS	*ORDER*	*FAMILY*	*GENUS/SPECIES*
Arachnida	**Araneida**		
	Opiliones		
Crustacea	Isopoda		***Oniscus asellus***
Diplopoda*			
Chilopoda*			
Insecta	Collembola*		
	Coleoptera	**Carabidae**	***Amara aulica***
			Carabus nemoralis
			Carabus violaceous
			Feronia nigrita
			Leistus spinibarbis
		Cryptophagidae	
		Curculionidae	***Caenopsis fissirostris***
			Otiorhynchus singularis
		Elateridae	***Athous haemorrhoidalis***
		Scarabaeidae	***Strophosomus melanogrammus***
		Staphylinidae	
	Diptera		
	Dermaptera		***Forficula auricularia***
	Hemiptera*		
	Hymenoptera		***Bombus spp.***
	Lepidoptera*		
Oligochaeta			***Lumbricus spp.***

Those names highlighted in bold represent groupings used for analysis; where no species are listed or are not highlighted, then the species were bulked into the higher Family or Order.
Adult and larvae forms of Coleoptera were analysed separately.
* Very small masses (<0.5 g wet weight) were collected and not analysed.

Table 3.21: Seasonal abundance and biomass (g dry weight) of the invertebrates analysed from Lady Wood (all transects).

Sample Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August '94 to January '95*
Araneida	0.42	0.51	0.25	-----
Opiliones	0.41	2.06	0.57	1.50
Oniscus asellus	4.71	3.94	1.27	1.64
Coleoptera (adults)	7.61	6.26	2.31	27.51
Coleoptera (larvae)	1.13	0.54	0.95	2.52
Carabidae	10.78	11.96	1.06	-----
Carabus violaceous	37.42	72.19	-----	36.35
Feronia nigrita	23.99	29.88	-----	20.82
Staphylinidae	1.94	0.68	0.85	2.51
Diptera	0.50	0.98	0.29	1.30
Forficula auricularia	0.18	0.84	2.00	2.21
Hymenoptera	3.04	0.56	-----	-----
Oligochaeta	0.27	0.33	0.86	0.74

----- Sample not obtained.

December 1995. Low capture rates were not the only problem at this site. The activities of badgers from a set near to Lady Wood frequently disturbed the pitfalls. The lids were removed allowing rain and leaf litter in, or the containers were completely dislodged. On average, four of the six pitfalls laid along each transect were usable each month. On two occasions every pitfall on the site was disturbed.

The species composition and numbers caught in the pitfall traps provide further evidence that the woodland soil and leaf litter is generally inhospitable for most invertebrates, due to the low pH. Most of the species are either detritivores or predatory. The lack of herbivorous insects is not surprising, given the scarcity of vegetation in the understorey, although a small number of herbivorous weevils were caught (Curculionidae) and are believed to have been feeding on the *P. sitchensis*. Because only small numbers were caught, these were combined into the group 'Coleoptera' for analysis.

The numbers of Oligochaeta remained consistent throughout the study. Numbers were low but this was expected due to the acid soil. Of the other detritivores, only woodlice, *Oniscus asellus*, made a major contribution to the samples. Millipedes (Diplopoda) and springtails (Collembola) were present but only in very small numbers. The resulting dry weight was so small that the germanium detectors were unable to quantify the radionuclides within a reasonable counting time.

Harvestmen (Opiliones) and spiders (Araneida) were also present throughout the study period, albeit in low numbers. There was an four-fold increase in harvestmen in the September 1993

to February 1994 period. Across the Lady Wood site though, it was the predatory ground beetles, Carabidae, that showed the greatest fluctuation in numbers. Highest numbers were seen in the late summer and autumn months. Samples collected during the spring and early summer contained lower numbers of the two main species, *Carabus violaceous* and *Feronia nigrita*, and only a very small quantity of other Carabidae, particularly in year two. There was no coincidental increase in the number of Coleopteran larvae caught, although Carabid beetles are reported to exhibit two peaks in adult activity during the spring/early summer and late summer/autumn, with more larvae present during the winter and mid-summer (Wallwork, 1970).

Adult Diptera and Hymenoptera were caught in low numbers throughout the year. This low frequency is not surprising, since few flying insects were observed under the tree canopy. Earwigs, *Forficula auricularia*, showed an increase in number over the two years, but this could simply be related to variation in sampling.

3.7.1 ^{137}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am Activity in Invertebrates

Radionuclide activities recorded for each invertebrate species are measurements of the whole body burden. No attempt was made to remove the gut of the invertebrates. They were rinsed in clean water after storage in 2% formalin solution (section 3.3) but this would not remove all surface contamination. Reported activities are therefore probably overestimates of the absorbed body burden. This is not necessarily important for two reasons; firstly, visual inspection of invertebrate body surfaces showed negligible signs of contamination with soil particles; and secondly, any material present within, or on, the invertebrate prey will be ingested by predatory small mammals.

Tables 3.22 to 3.25 present data for ^{137}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am for each invertebrate species over the four sampling periods. In the counting time allocated for each sample, it was not possible to quantify ^{134}Cs. Also, most of the ^{241}Am results were generated on the germanium detectors and have higher than ideal counting uncertainties. The pitfall contents were combined from two transects (E and F; G and H) to generate two sets of results in the tables.

There is considerable variation between the different species and families/orders caught. For example, the ^{137}Cs values ranged from 8 Bq kg^{-1} in *Feronia nigrita* to 480 Bq kg^{-1} for Oligochaeta. As expected, the ^{137}Cs results are much higher than those for the actinides. Given previous discussion about the behaviour and bioavailability of the actinides this is not surprising.

Table 3.22: ^{137}Cs Activities (Bq kg^{-1} ± 2σ counting error) in invertebrate samples from Lady Wood.

Sample Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August '94 to January '95*
		Transects E and F		
Araneida	34 ± 20	146 ± 12	-----	-----
Opiliones	113 ± 9	75 ± 16	226 ± 60	572 ± 37
Oniscus asellus	76 ± 15	201 ± 21	174 ± 9	429 ± 32
Coleoptera (adults)	9 ± 5	47 ± 5	63 ± 19	148 ± 5
Coleoptera (larvae)	-----	223 ± 84	104 ± 47	120 ± 26
Carabidae	29 ± 10	10 ± 2	-----	-----
Carabus violaceous	9 ± 2	52 ± 1	-----	23 ± 2
Feronia nigrita	24 ± 3	8 ± 1	-----	43 ± 2
Staphylinidae	41 ± 17	50 ± 4	94 ± 39	59 ± 16
Diptera	72 ± 38	82 ± 32	45 ± 6	267 ± 28
Forficula auricularia	80 ± 9	46 ± 28	59 ± 17	231 ± 19
Hymenoptera	59 ± 16	91 ± 19	-----	1013 ± 302
Oligochaeta	-----	478 ± 129	620 ± 59	-----
		Transects G and H		
Araneida	44 ± 12	26 ± 20	115 ± 39	-----
Opiliones	-----	-----	-----	-----
Oniscus asellus	157 ± 23	72 ± 14	43 ± 8	-----
Coleoptera (adults)	14 ± 4	23 ± 11	76 ± 28	9 ± 2
Coleoptera (larvae)	34 ± 8	-----	-----	233 ± 21
Carabidae	65 ± 9	-----	46 ± 11	-----
Carabus violaceous	11 ± 2	17 ± 3	-----	28 ± 3
Feronia nigrita	13 ± 2	6 ± 1	-----	-----
Staphylinidae	<2.2	71 ± 48	-----	53 ± 20
Diptera	-----	-----	-----	176 ± 82
Forficula auricularia	-----	125 ± 50	-----	-----
Hymenoptera	-----	-----	-----	-----
Oligochaeta	868 ± 135	-----	-----	1800 ± 91

----- Sample not obtained.
n/a Sample not analysed.

Of the species analysed, the detritivorous fauna were represented by two major groups: Oligochaeta and Isopoda. In addition, the diet of the earwig, *Forficula auricularia*, includes some detritus. Species representing these detritivorous groups generally contain more activity than, for example, in the predatory group. As an illustration, the activities of *O. asellus* range from 76 to 429 Bq kg^{-1}, 1 to 9.4 Bq kg^{-1},14 to 74 Bq kg^{-1}, and 21 to 36 Bq kg^{-1} for ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am respectively. The samples of Oligochaeta contain levels of ^{137}Cs ranging from 480 to 1800 Bq kg^{-1} and ^{241}Am levels of 17 to 50 Bq kg^{-1}. Unfortunately, because there were only a few samples collected, no radiochemical analysis was undertaken on the Oligochaeta. The high levels of ^{137}Cs and ^{241}Am in the Oligochaeta are accounted for by the presence of soil in their gut. Rudge (1989) quotes values of 22 to 55% for the soil in the gut contribution to earthworm dry weight. Levels of ^{137}Cs in earthworm tissues were far

Table 3.23: ^{238}Pu Activities (Bq kg^{-1} ± 2σ counting error) in invertebrate samples from Lady Wood.

Sample Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August '94 to January '95*
		Transects E and F		
Araneida	<1.3	n/a	-----	-----
Opiliones	16 ± 2	1.4 ± 0.3	n/a	2.6 ± 0.6
Oniscus asellus	1 ± 0.2	6.8 ± 1.1	9.4 ± 1.7	7.7 ± 1.4
Coleoptera (adults)	n/a	0.9 ± 0.2	2.9 ± 0.5	18 ± 0.9
Coleoptera (larvae)	-----	n/a	n/a	4.6 ± 1.0
Carabidae	0.6 ± 0.1	<0.05	-----	-----
Carabus violaceous	0.03 ± 0.01	<0.46	-----	1.0 ± 0.1
Feronia nigrita	<0.05	0.10 ± 0.03	-----	1.4 ± 0.1
Staphylinidae	n/a	15 ± 1.5	4.1 ± 0.8	2.0 ± 0.4
Diptera	3.6 ± 0.7	n/a	23.3 ± 2.6	6.4 ± 0.9
Forficula auricularia	-----	5.4 ± 1.1	23.4 ± 2.9	1.0 ± 0.3
Hymenoptera	2.1 ± 0.4	9.0 ± 1.3	-----	n/a
Oligochaeta	-----	n/a	n/a	-----
		Transects G and H		
Araneida	n/a	10 ± 1.7	14 ± 2	-----
Opiliones	-----	-----	-----	-----
Oniscus asellus	14.5 ± 1.8	1.9 ± 0.3	4.9 ± 0.7	-----
Coleoptera (adults)	0.8 ± 0.2	1.9 ± 0.4	148 ± 4.6	0.34 ± 0.05
Coleoptera (larvae)	n/a	-----	-----	3.9 ± 0.8
Carabidae	n/a	-----	52 ± 1.8	-----
Carabus violaceous	0.18 ± 0.03	0.6 ± 0.1	-----	0.9 ± 0.1
Feronia nigrita	0.14 ± 0.02	0.33 ± 0.09	-----	-----
Staphylinidae	2.3 ± 0.3	33 ± 2.4	-----	9.4 ± 1.1
Diptera	-----	-----	-----	8.8 ± 1.1
Forficula auricularia	-----	n/a	-----	-----
Hymenoptera	-----	-----	-----	-----
Oligochaeta	n/a	-----	-----	n/a

----- Sample not obtained.
n/a Sample not analysed.

below those of the gut contents. Had sufficient biomass been available, similar differences would probably have been observed in the case of Lady Wood. Earthworms are known to preferentially ingest organic debris and small particles such as clay and silt (Rudge, 1989), and it has been shown that fixation of radionuclides in soil is influenced by particle size and organic matter content (section 1.4.1). Consequently, the high activities measured are probably related to soil in the Oligochaete gut.

Unlike the Oligochaeta *O. asellus* was caught throughout the study period. *O. asellus* feeds mainly on decaying vegetable matter, but also on algae and fungi growing on tree bark of, or decaying, wood (Wallwork, 1970). With the exception of ^{238}Pu, each radionuclide exhibited a clear pattern whereby activity declined during the spring and summer compared to the winter

Table 3.24: $^{239+240}$Pu Activities (Bq kg^{-1} ± 2σ counting error) in invertebrate samples from Lady Wood.

Sample Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August '94 to January '95*
	Transects E and F			
Araneida	10.5 ± 1.0	n/a	-----	-----
Opiliones	11.3 ± 2.0	8.4 ± 0.7	n/a	15.6 ± 1.4
Oniscus asellus	14.1 ± 0.9	59.4 ± 3.3	21.1 ± 2.6	74 ± 4.3
Coleoptera (adults)	n/a	4.9 ± 0.4	7.9 ± 0.8	8.4 ± 0.6
Coleoptera (larvae)	-----	n/a	n/a	8.7 ± 1.4
Carabidae	4.6 ± 0.4	0.10 ± 0.02	-----	-----
Carabus violaceous	0.52 ± 0.05	0.21 ± 0.02	-----	4.2 ± 0.3
Feronia nigrita	0.16 ± 0.02	0.43 ± 0.05	-----	5.7 ± 0.3
Staphylinidae	n/a	2.2 ± 0.6	3.8 ± 0.8	2.3 ± 0.4
Diptera	6.8 ± 0.9	n/a	8.5 ± 1.6	9.0 ± 1.1
Forficula auricularia	n/a	5.4 ± 1.1	15.9 ± 2.4	4.6 ± 0.5
Hymenoptera	4.7 ± 0.6	6.0 ± 1.0	-----	n/a
Oligochaeta	-----	n/a	n/a	-----
	Transects G and H			
Araneida	n/a	12.1 ± 1.9	8.8 ± 1.6	-----
Opiliones	-----	-----	-----	-----
Oniscus asellus	15.5 ± 1.9	10.5 ± 0.6	6.5 ± 0.8	-----
Coleoptera (adults)	1.2 ± 0.2	4.1 ± 0.6	4.8 ± 0.8	1.4 ± 0.1
Coleoptera (larvae)	n/a	-----	-----	10.5 ± 1.3
Carabidae	n/a	-----	1.9 ± 0.3	-----
Carabus violaceous	0.5 ± 0.05	0.9 ± 0.1	-----	1.8 ± 0.1
Feronia nigrita	0.30 ± 0.04	0.7 ± 0.1	-----	-----
Staphylinidae	1.3 ± 0.2	3.2 ± 0.8	-----	3.4 ± 0.6
Diptera	-----	-----	-----	6.1 ± 0.9
Forficula auricularia	-----	n/a	-----	-----
Hymenoptera	-----	-----	-----	-----
Oligochaeta	n/a	-----	-----	n/a

----- Sample not obtained.
n/a Sample not analysed.

(Figures 3.20 and 3.21). This may relate to changes in feeding patterns; for example, an increased consumption of decaying fungi by *O. asellus* during the winter period. Recent data on the activity in fungi suggests that this may be a significant pathway for radionuclide transfer (Toal, pers. comm.). Furthermore, there is evidence that the hepatopancreas of *O. asellus* concentrates heavy metals from the environment. In particular, this has been recorded for copper and appears to be a physiological response to detoxify the body of excess or non-metabolically important metals (Nriagu, 1979). This mechanism may also influence the radionuclide content of *O. asellus*.

The results obtained for *F. auricularia* show considerable variation between radionuclides. Within the woodland, these omnivorous insects feed mainly on decaying plant and animal

Table 3.25: ^{241}Am Activities (Bq kg^{-1} ± 2σ counting error) in invertebrate samples from Lady Wood.

Sample Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August '94 to January '95*
		Transects E and F		
Araneida	<0.3	<1.5	-----	-----
Opiliones	5.8 ± 1.8	4.3 ± 2.0	25.5 ± 12.9	11.8 ± 2.5
Oniscus asellus	21.1 ± 7.9	39.1 ± 18.8	21.4 ± 2.9	36 ± 14
Coleoptera (adults)	<0.44	<1.45	11.7 ± 3.1	3.0 ± 0.8
Coleoptera (larvae)	-----	14.9 ± 3.3	7.8 ± 2.7	<0.14
Carabidae	<0.21	0.7 ± 0.4	-----	-----
Carabus violaceous	0.5 ± 0.3	0.5 ± 0.1	-----	1.1 ± 0.4
Feronia nigrita	1.5 ± 0.9	<0.09	-----	<1.49
Staphylinidae	<2.0	<2.1	8.0 ± 6.9	<1.49
Diptera	3.8 ± 2.0	11.6 ± 6.6	5.7 ± 2.4	3.5 ± 1.2
Forficula auricularia	<1.5	<0.56	3.0 ± 2.1	7.1 ± 3.3
Hymenoptera	<2.89	18.5 ± 10.4	-----	400 ± 148
Oligochaeta	-----	35.4 ± 16.9	17.3 ± 7.4	-----
		Transects G and H		
Araneida	<2.9	5.9 ± 1.7	6.4 ± 3.4	-----
Opiliones	-----	-----	-----	-----
Oniscus asellus	8.1 ± 4.4	10.6 ± 7.0	6.0 ± 4.5	-----
Coleoptera (adults)	<0.65	10.9 ± 7.9	28 ± 15	<0.31
Coleoptera (larvae)	<0.23	-----	-----	6.6 ± 4.3
Carabidae	1.2 ± 0.4	-----	2.6 ± 1.8	-----
Carabus violaceous	<0.16	<0.23	-----	<0.2
Feronia nigrita	0.24 ± 0.09	0.8 ± 0.3	-----	-----
Staphylinidae	13.5 ± 1.7	<0.41	-----	4.9 ± 2.4
Diptera	-----	-----	-----	23.6 ± 18.0
Forficula auricularia	-----	145 ± 92	-----	-----
Hymenoptera	-----	-----	-----	-----
Oligochaeta	27.0 ± 7.9	-----	-----	50 ± 25

----- Sample not obtained.

material (Chinery, 1993). In addition, the capture rates in pitfalls located along different transects suggest that earwigs are more prevalent along transect E and therefore may also be feeding upon the vegetation growing on site which would be an additional source of radionuclides.

Of the predatory invertebrates, those exhibiting the highest radionuclide activities were Arachnids, present as spiders (Araneida) and harvestmen (Opiliones). From the data, the harvestmen demonstrated the highest activity levels for each radionuclide, being between 1 and 4 times higher than the spiders, although the differences were most notable for ^{137}Cs. Actinide levels remained mostly consistent throughout the study period, although levels of ^{137}Cs showed a marked increase in activity over the two years (Figures 3.22 and 3.23). There

Figure 3.20: Temporal changes in the ^{137}Cs activity in *Oniscus asellus*.

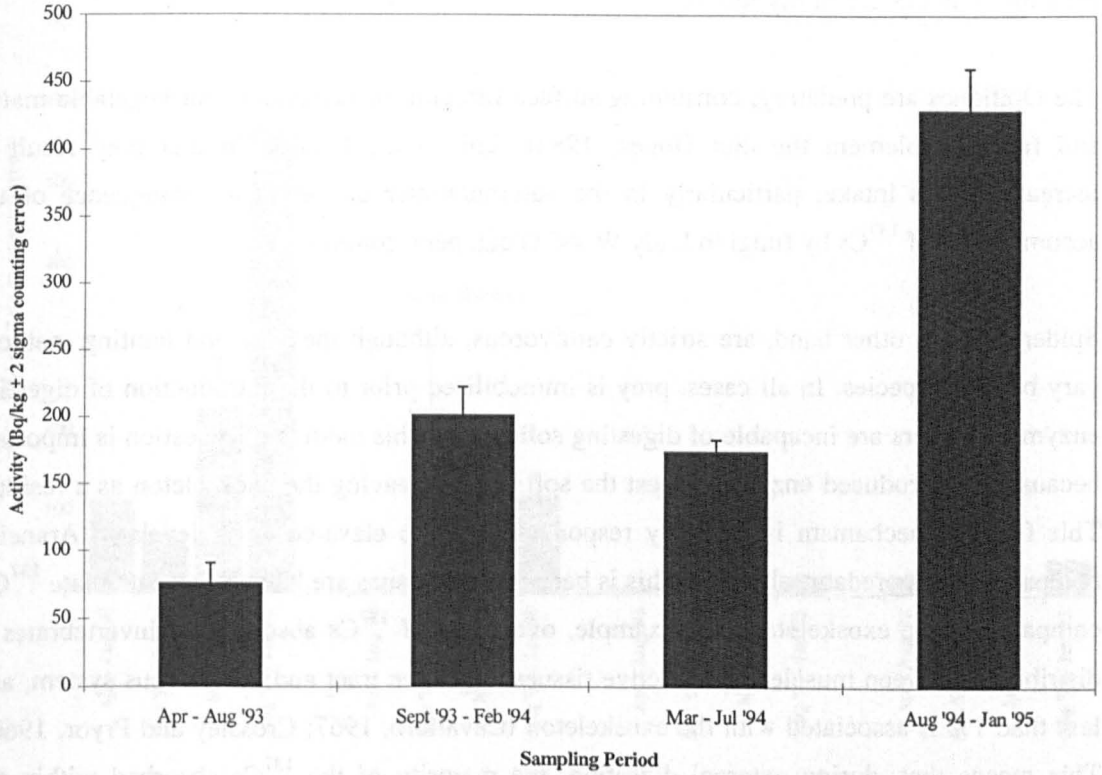
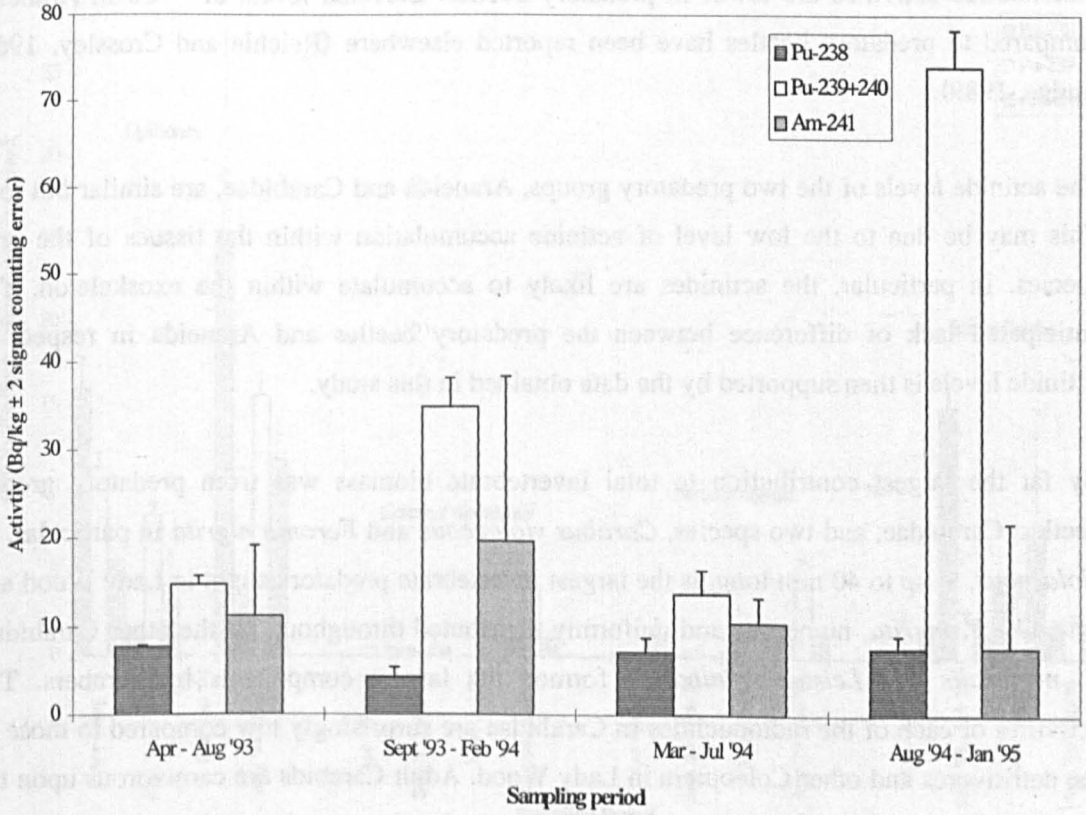


Figure 3.21: Temporal changes in the activity of ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in *Oniscus asellus*.



are no obvious reasons for this increase in activity given that the other radionuclides appear reasonably stable in activity levels.

The Opiliones are predatory, consuming surface foraging invertebrates; but vegetable matter and fungi supplement the diet (Jones, 1983). This diversification in diet may result in increased ^{137}Cs intake, particularly in the autumn/winter period as a consequence of the accumulation of ^{137}Cs by fungi in Lady Wood (Toal, pers. comm.).

Spiders, on the other hand, are strictly carnivorous, although the prey and hunting methods vary between species. In all cases, prey is immobilised prior to the introduction of digestive enzymes. Spiders are incapable of digesting solid food. This method of digestion is important because the introduced enzymes digest the soft tissues, leaving the exoskeleton as a residue. This feeding mechanism is probably responsible for the elevated ^{137}Cs levels in Araneida compared to the predatory beetles. This is because soft tissues are known to accumulate ^{137}Cs, compared to the exoskeleton. For example, over 90% of ^{137}Cs absorbed by invertebrates is distributed between muscle, reproductive tissues, digestive tract and the nervous system, and less than 1% is associated with the exoskeleton (Cavalloro, 1967; Crossley and Pryor, 1960). This means that, during external digestion, the majority of the ^{137}Cs absorbed within the invertebrates' body will be transferable to the predator. Furthermore, the presence of a large and heavy exoskeleton will lead to an underestimation of the significance of contamination in the soft tissues (on a weight for weight (w/w) basis) and may be an additional reason why radionuclide activities are lower in predatory beetles. Elevated levels of ^{137}Cs in Araneida compared to predatory beetles have been reported elsewhere (Reichle and Crossley, 1969; Rudge, 1989).

The actinide levels of the two predatory groups, Araneida and Carabidae, are similar but low. This may be due to the low level of actinide accumulation within the tissues of the prey species. In particular, the actinides are likely to accumulate within the exoskeleton. The anticipated lack of difference between the predatory beetles and Araneida in respect of actinide levels is then supported by the data obtained in this study.

By far the largest contribution to total invertebrate biomass was from predatory ground beetles, Carabidae, and two species, *Carabus violaceous* and *Feronia nigrita* in particular. *C. violaceous*, at up to 40 mm long, is the largest invertebrate predator caught in Lady Wood and was, like *F. nigrita*, numerous and uniformly distributed throughout. Of the other Carabidae, *C. nemoralis* and *Leistus spinibarbis* formed the largest components by numbers. The activities of each of the radionuclides in Carabidae are surprisingly low compared to those of the detritivores and other Coleoptera in Lady Wood. Adult Carabids are carnivorous upon the more active surface dwelling animals, such as smaller beetles, earwigs, and some detritivores

Figure 3.22: Temporal changes in ^{137}Cs activity in predatory invertebrates.

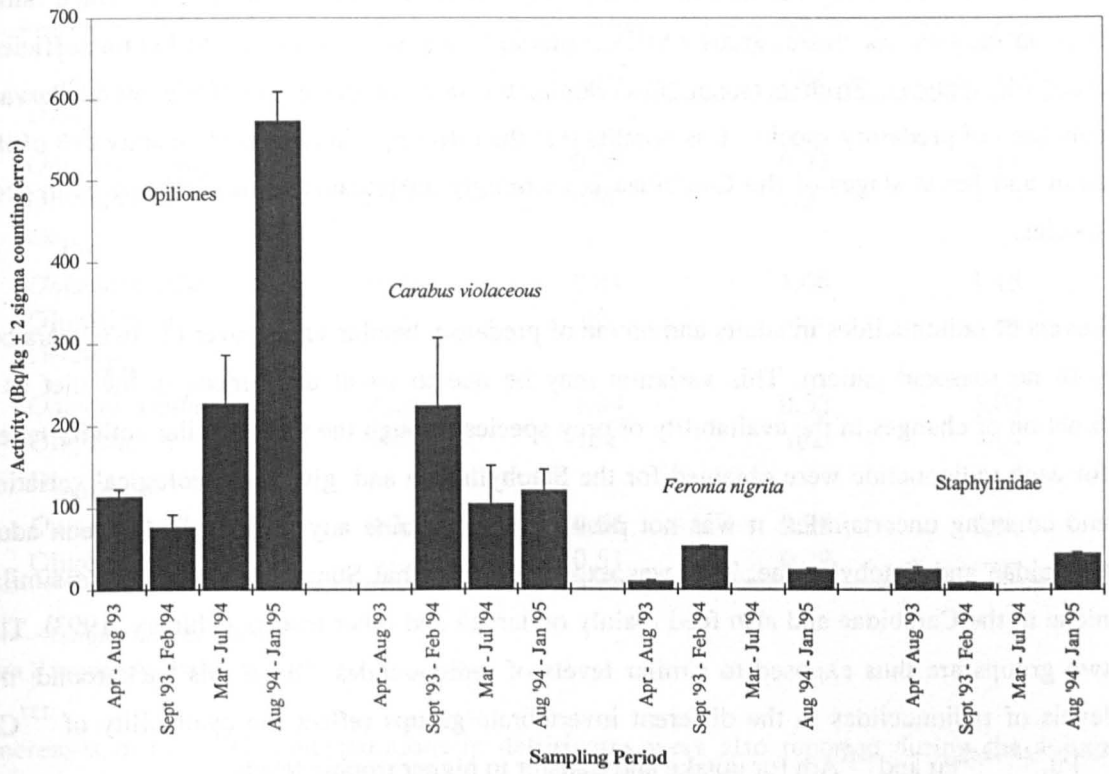
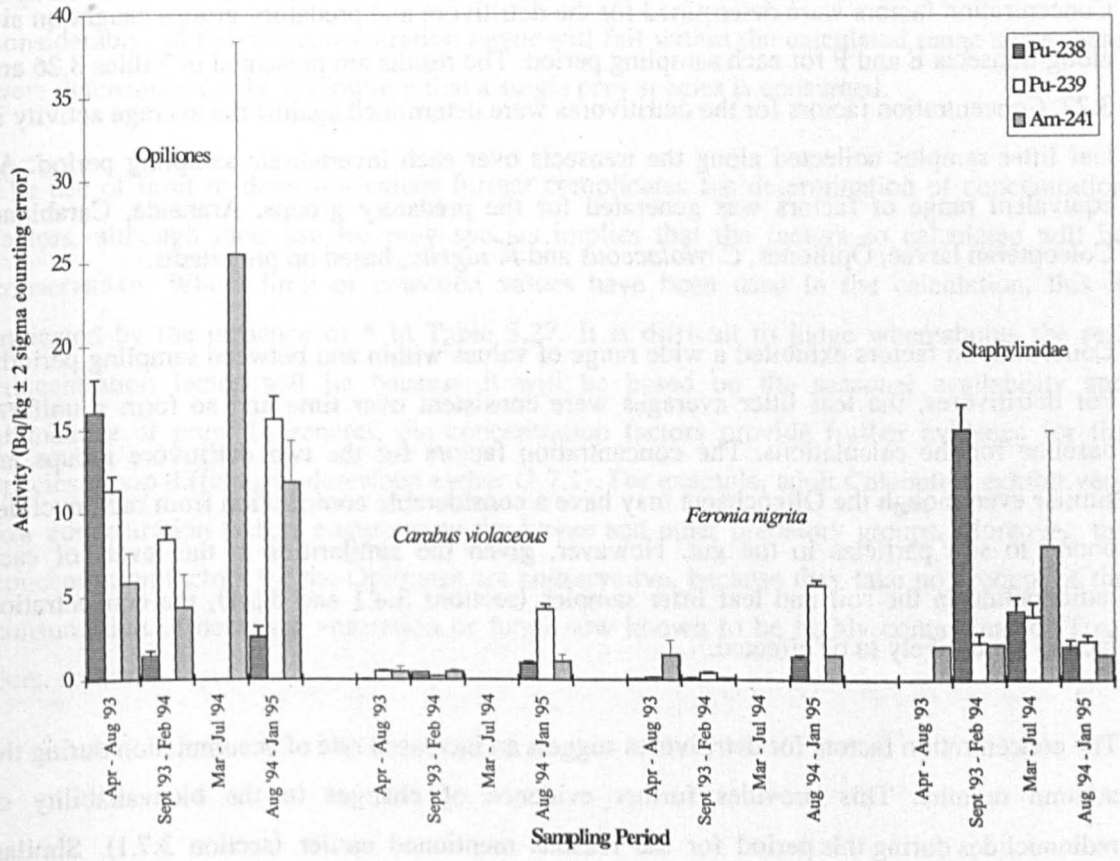


Figure 3.23: Temporal changes in the ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities in predatory invertebrates.



(Chinery, 1993). The larval stages consume large numbers of detritivores such as *O. asellus* and Collembola, mainly because the larvae spend more time in the soil-litter interface rather than on the surface. In this study all Coleopteran larvae were combined to obtain sufficient mass for analysis. From personal observation, the bulk of the group 'Coleopteran larvae' consisted of predatory species. It is notable that the difference in radionuclide activities of the adult and larval stages of the Carabidae is seemingly attributable to the differences in prey species.

Levels of radionuclides in adults and larvae of predatory beetles varied over the two years but with no seasonal pattern. This variation may be due to small differences in the diet as a function of changes in the availability of prey species through the year. Similar activity levels for each radionuclide were obtained for the Staphylinidae and, given the biological variation and counting uncertainties, it was not possible to determine any differences between adult Carabidae and Staphylinidae. This was expected given that Staphylinidae occupy a similar niche to the Carabidae and also feed mainly on larvae and other insects (Chinery, 1993). The two groups are thus exposed to similar levels of radionuclides. Given this background, the levels of radionuclides in the different invertebrate groups reflect the availability of ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am for uptake and transfer to higher trophic levels.

3.7.2 Radionuclide Concentration Factors in Invertebrates

Concentration factors were determined for the detritivore and predatory groups caught on site along transects E and F for each sampling period. The results are presented in Tables 3.26 and 3.27. Concentration factors for the detritivores were determined against the average activity in leaf litter samples collected along the transects over each invertebrate sampling period. An equivalent range of factors was generated for the predatory groups, Araneida, Carabidae, Coleopteran larvae, Opiliones, *C. violaceous* and *F. nigrita*, based on prey items.

Concentration factors exhibited a wide range of values within and between sampling periods. For detritivores, the leaf litter averages were consistent over time and so form a uniform baseline for the calculations. The concentration factors for the two detritivore groups are similar even though the Oligochaeta may have a considerable contribution from radionuclides bound to soil particles in the gut. However, given the similarities in the levels of each radionuclide in the soil and leaf litter samples (sections 3.4.1 and 3.5.1), the concentration factors are unlikely to be affected.

The concentration factors for detritivores suggest an increased rate of accumulation during the autumn months. This provides further evidence of changes in the bioavailability of radionuclides during this period for the reasons mentioned earlier (section 3.7.1). Similar

Table 3.26: Detritivore dietary concentration factors determined against average radionuclide activities in leaf litter samples.

Radionuclide and Species or Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August '94 to January '95*
^{137}Cs				
Oniscus asellus	0.13	0.38	0.37	0.73
Oligochaeta	-----	0.90	1.33	-----
^{238}Pu				
Oniscus asellus	0.13	0.81	1.68	1.15
Oligochaeta	-----	n/a	n/a	-----
$^{239+240}$Pu				
Oniscus asellus	0.26	1.04	0.55	1.80
Oligochaeta	-----	n/a	n/a	-----
^{241}Am				
Oniscus asellus	0.33	0.56	0.35	0.56
Oligochaeta	-----	0.51	0.29	-----

----- Sample not obtained.
n/a Sample not analysed.

increases in the ^{137}Cs concentrations in detritivores were also reported during the autumn period by Rudge (1989).

Concentration factors for the predators were determined against those prey species with the highest and lowest radionuclide activities. However, the diet of the predators vary considerably, so the real concentration factor will fall within the calculated range since these were determined on the assumption that a single prey species is consumed.

The use of limit of detection values further complicates the determination of concentration factors, although their use for prey species implies that the factors so calculated will be conservative. Where limit of detection values have been used in the calculation, this is indicated by the presence of * in Table 3.27. It is difficult to judge whereabouts the real concentration factor will lie because it will be based on the seasonal availability and abundance of prey. In general, the concentration factors provide further evidence for the species/group differences described earlier (3.7.1). For example, adult Coleoptera exhibit very low concentration factors compared to the larvae and other predatory groups. Moreover, the concentration factors for the Opiliones are conservative, because they take no account of the consumption of decaying vegetation or fungi now known to be highly contaminated (Toal, pers. comm.).

Table 3.27: Dietary concentration factors for the predators (against a range of prey species) from Lady Wood.

Radionuclide and Species or Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August '94 to January '95*
^{137}Cs				
Araneida	0.43 - 0.47	0.73 - 3.17	-----	-----
Carabidae	0.26 - 3.22	0.04 - 1.25	-----	-----
Coleopteran larvae[1]	-----	1.11	0.60	0.28
Opiliones	1.41 - 1.57	0.37 - 1.63	1.30 - 5.02	1.33 - 2.48
Carabus violaceous	0.08 - 1.00	0.23 - 6.50	-----	0.04 - 1.00
Feronia nigrita	0.21 - 2.67	0.04 - 1.00	-----	0.08 - 1.87
^{238}Pu				
Araneida	0.36* - 2.17*	n/a	-----	-----
Carabidae	0.04 - 0.60	0.003* - 0.50*	-----	-----
Coleopteran larvae[1]	-----	n/a	n/a	0.26
Opiliones	4.44 - 26.7	0.21 - 1.56	n/a	0.14 - 2.60
Carabus violaceous	0.002 - 1.00	0.03* - 4.60*	-----	0.06 - 1.67
Feronia nigrita	0.003* - 1.67*	0.01 - 1.00	-----	0.08 - 1.40
$^{239+240}$Pu				
Araneida	0.74 - 2.28	n/a	-----	-----
Carabidae	0.33 - 0.98	0.002 -- 0.48	-----	-----
Coleopteran larvae[1]	-----	n/a	n/a	
Opiliones	0.80 - 2.46	0.14 - 21.0	n/a	
Carabus violaceous	0.04 - 0.11	0.004 - 0.49	-----	
Feronia nigrita	0.01 - 0.03	0.007 - 0.09	-----	
^{241}Am				
Araneida	0.01* - 0.70*	0.04* - 1.03*	-----	-----
Carabidae	0.01* - 0.42*	0.02 - 1.25-	-----	-----
Coleopteran larvae[1]	-----	0.38	0.36	0.00*
Opiliones	0.27 - 3.87*	0.11 - 2.76*	1.19 - 8.50	0.33 - 1.66
Carabus violaceous	0.02 - 1.14	0.01 - 0.71	-----	0.03 - 0.74
Feronia nigrita	0.07 - 3.40*	0.002* - 0.16*	-----	0.04* - 0.50*

[1] Radionuclide activities were only determined in one prey item, *O. asellus*, which was used to calculate the concentration factor.
* Limit of detection values used during the calculation of the concentration factor.

A number of concentration factors of unity or greater are reported in Table 3.27. These indicate either that the predators are consuming other smaller individuals from their own group/species, or, alternatively, it reflects the consumption of large numbers of prey with elevated levels of radionuclides; for example, *O. asellus* (Tables 3.22 to 3.25). The latter is more important because it cannot be assumed that concentrations of the radionuclides will be as high in smaller individuals of a taxonomic group. Furthermore, the relationship between body burdens of radionuclides in invertebrates and their diet as investigated in detail by Reichle and van Hook (1970), indicates that higher trophic levels are generally exposed to lower radionuclide concentrations.

The ^{137}Cs concentration factors show reasonable agreement with those determined for the same predatory groups inhabiting a grassland area at the BNFL low level waste disposal site at Drigg (Rudge, 1989). Other authors have also quoted concentration factors for ^{137}Cs for predators in the range 0.3 to 0.92 (Crossley, 1969; Reichle and Crossley, 1969; Reichle and Van Hook, 1970). Not surprisingly, concentration factors for the actinides are generally lower than those for ^{137}Cs, particularly for the Carabidae and the two species of beetle, *C. violaceous* and *F. nigrita*. This was expected, given the behaviour and transfer of the actinides through food chains, although the contrast between the actinide and ^{137}Cs concentration factors further emphasises the absorption and transfer differences between the predatory beetles and the two groups of arachnids.

For each of the radionuclides, the Opiliones exhibit the highest range of concentration factors. This is due to their dietary habits, particularly the fact that they are not strictly carnivorous. By feeding on other materials available within the woodland, their radionuclide burden will be elevated.

It is known that the biological half-life of ^{137}Cs in invertebrates is short, typically less than a day (Crossley, 1963b; Digregorio *et al.*, 1978) however, it is speculated that the actinides will have much longer biological half-life especially if the actinides accumulate within the exoskeleton. This could act as a sink for the actinides in the same way that chitin is known to accumulate heavy metals. Consequently, it is projected that subtle seasonal differences might be obscured. For example, time lags between the transfer of more biologically mobile radionuclides, such as ^{137}Cs, have been reported between invertebrate and other trophic levels (Reichle and Crossley, 1969). The pooling undertaken in the present study, particularly across several months, will further mask these differences.

3.7.3 Isotopic and Nuclide Ratios

Table 3.28 shows calculated isotopic (^{238}Pu:$^{239+240}Pu$) and nuclide (^{137}Cs:$^{239+240}Pu$ and ^{137}Cs:^{241}Am) ratios for selected invertebrate groups across the four trapping campaigns. The lack of ^{134}Cs data prevented calculation of the ^{134}Cs:^{137}Cs ratio.

The ratios are very variable for the different taxonomic groups and across the trapping sessions; although the range of values for the ^{238}Pu:$^{239+240}Pu$ ratio for *O. asellus* is similar to that for leaf litter and therefore provides collateral evidence of its principal dietary component. The predatory invertebrates exhibit a range of values for the isotopic ratio, ^{238}Pu:$^{239+240}Pu$, which again reflects their more varied diet. It is difficult to distinguish any principal prey in each trapping period but in general it is suggested that plutonium is ingested

Table 3.28: **Isotopic and nuclide ratios for selected invertebrate groups from Lady Wood. Reported values calculated from Transects E and F.**

Radionuclide and Species or Group	April to August '93	September '93 to February '94	March to July '94	August '94 to January '95
Araneida				
^{238}Pu:$^{239+240}Pu$	>0.12	n/a	-----	-----
^{137}Cs:$^{239+240}Pu$	38.24	n/a	-----	-----
^{137}Cs:^{241}Am	>109.68	>97.33	-----	-----
Opiliones				
^{238}Pu:$^{239+240}Pu$	1.42	0.17	n/a	0.49
^{137}Cs:$^{239+240}Pu$	10.00	8.93	n/a	36.67
^{137}Cs:^{241}Am	19.48	17.44	8.86	48.47
Onsicus asellus				
^{238}Pu:$^{239+240}Pu$	0.07	0.11	0.45	0.10
^{137}Cs:$^{239+240}Pu$	5.39	3.38	8.25	5.80
^{137}Cs:^{241}Am	3.60	5.14	8.13	11.92
Coleopteran adults				
^{238}Pu:$^{239+240}Pu$	n/a	0.18	0.37	2.14
^{137}Cs:$^{239+240}Pu$	n/a	9.59	7.97	17.62
^{137}Cs:^{241}Am	>20.45	>32.41	5.38	49.33
Carabidae				
^{238}Pu:$^{239+240}Pu$	0.13	>0.50	-----	-----
^{137}Cs:$^{239+240}Pu$	6.30	100.00	-----	-----
^{137}Cs:^{241}Am	>138.10	14.29	-----	-----
Carabus violaceous				
^{238}Pu:$^{239+240}Pu$	0.06	>2.19	-----	0.24
^{137}Cs:$^{239+240}Pu$	17.31	247.62	-----	5.48
^{137}Cs:^{241}Am	18	104.00	-----	20.91
Staphylinidae				
^{238}Pu:$^{239+240}Pu$	n/a	6.82	1.08	0.87
^{137}Cs:$^{239+240}Pu$	n/a	22.73	24.74	56.70
^{137}Cs:^{241}Am	>10.25	>23.81	11.75	>39.60
Diptera				
^{238}Pu:$^{239+240}Pu$	0.53	n/a	2.74	0.71
^{137}Cs:$^{239+240}Pu$	10.59	n/a	5.29	29.67
^{137}Cs:^{241}Am	18.95	7.07	7.89	76.29

----- Sample not collected.
> LOD values used therefore the ratio will be greater than the value reported.
n/a Sample not analysed.

in a ratio similar to that in the environment. Having said that, there are a few values close to or greater than unity. This was unexpected and there is no clear reason for it, suggesting high biological variation within the data set which makes these comparisons difficult.

The nuclide ratios, ^{137}Cs:$^{239+240}Pu$ and ^{137}Cs:^{241}Am, are very similar to those reported for soil and leaf litter for *O. asellus*. From the known transfer coefficients for caesium and the actinides, it was anticipated that the invertebrate ratios would indicate an elevated concentration of ^{137}Cs. Several results do show an enhanced level of ^{137}Cs, particularly for

predators such as Araneida, Carabidae, *C. violaceous* and Staphylinidae. This is in agreement with the hypothesis of lower actinide transfer.

3.7.4 Comparison with Cheshire Invertebrates

Invertebrate data from Lady Wood were compared with a set of composite invertebrate samples caught in pitfalls and collected by vacuum sampling from the Cheshire site. The latter samples were collected on three occasions during the two year study period: October 1993, March 1994 and July 1994. In each case the pitfalls were exposed for approximately one month. Unfortunately, as with Lady Wood, the traps were disturbed regularly so only 25 to 50% were usable. Again it was necessary to generate pooled composite samples for the invertebrates. The results are presented in Table 3.29 and can be compared directly with the data in Tables 3.22 to 3.25.

Table 3.29: Radionuclide activity (Bq kg^{-1} ± 2σ counting error) in composite invertebrate samples collected from Cheshire.

Sample Dates	*Composite Invertebrate samples*			
	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
October 1993	<1.32	<0.19	<0.08	<0.38
March 1994	<0.57	<0.34	0.74 ± 0.09	<0.84
July 1994	2.51 ± 1.7	0.27 ± 0.04	0.23 ± 0.04	<0.73

< Values obtained were less than the limit of detection, the LOD value is reported.

Despite the limitations of sampling pooling, the results presented in Table 3.29 indicate differences between the Cheshire and Lady Wood sites. These are thought to be biologically significant, but cannot be subjected to statistical analysis. The Cheshire site values are mostly less than the limits of detection values dictated for ^{137}Cs by the Omnigam software and by manual techniques for the actinides (section 2.2.5).

3.7.5 Summary of Invertebrate Results

Whole body burdens of invertebrates from the woodland varied between species and pooled taxonomic groups, in the range 6 to 1,800 Bq kg^{-1} (^{137}Cs), 0.3 to 148 Bq kg^{-1} (^{238}Pu), 0.3 to 59 Bq kg^{-1} ($^{239+240}Pu$) and <0.3 to 400 Bq kg^{-1} (^{241}Am). Low levels of ^{134}Cs precluded its quantification.

The detritivorous fauna, arguably the most important in a coniferous woodland ecosystem, showed higher values than for other taxa, namely 43 to 1,800 Bq kg^{-1} (^{137}Cs), 1 to 15 Bq kg^{-1} (^{238}Pu), 7 to 74 Bq kg^{-1} ($^{239+240}Pu$) and <0.3 to 400 Bq kg^{-1} (^{241}Am). In the case of ^{137}Cs and

^{241}Am, the highest values were for Oligochaete earthworms wherein the gut contains residual quantities of contaminated soil ingested involuntarily along with fragments of food. Isopods also showed strongly elevated levels of all measurable nuclides, reflecting the level of contamination of their litter food source.

Seasonal patterns of radionuclide activity were such that, with the exception of ^{238}Pu levels in biota were lower in the spring and summer, compared to the autumn and winter. This is thought to be coincident with changes in feeding strategy, and the increased role of higher activity food items, such as fungi, which are favoured by detritivores in the over-wintering period. Of the predatory species, the Araneida and Opiliones showed the highest levels of ^{137}Cs. The high levels in spiders, compared to beetles at a similar trophic level, may reflect their exo-enzyme digestion system whereby the soft tissues of their prey are digested externally and then ingested as a partly processed foodstuff. ^{137}Cs is preferentially associated with soft, as opposed to skeletal, tissues.

Radionuclide concentration factors (predator:prey) were mostly variable across species and seasons. Those concentration factors reported for the detritivores showed some stability, and were consistently higher than unity. The latter observation was restricted to ^{137}Cs, with the actinides showing much lower concentration factors due to their lower absorption and biological transfer coefficients.

3.8 SMALL MAMMAL RESULTS

Table 3.30 presents the general details of animals caught at each trapping session. Table 3.31 and Figures 3.24 and 3.25 present the radionuclide data for small mammals. Four trapping campaigns were undertaken during the study period. For animals from the first three, whole body burdens of ^{137}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am were determined. From the fourth, the animals were dissected and pooled samples formed of body hair, gut, lungs, muscle, organs (comprising: heart, kidneys, liver and spleen) and skeleton. Animals collected for the whole body determinations were trapped along both the front and back edges of the woodland (with respect to the Sellafield complex as indicated in Figure 3.1), whereas the final trapping was undertaken along transects E and F only.

An attempt was made to collect a minimum of three animals of each sex. This was not possible, however, as indicated in the table. Wet weights are reported as a broad guide to age. No juvenile animals were taken. Statistical analysis using the Mann-Whitney U test indicated that there were no significant differences between the activity of ^{137}Cs in males and females ($p>0.05$) and therefore the whole body burden data were pooled to generate arithmetic means

Table 3.30: ***Apodemus sylvaticus*** **caught in Lady Wood.**

Trapping Session	*Number Caught*	*Sexes*	*Wet Weights* (g)
September 1993	25 (7)	4 male, 2 female	24.3, 25.2, 19.8, 27.9, 27.0, 20.7
March 1994	29 (6)	5 male, 1 female	22.8, 19.2, 25.5, 27.1, 24.2, 18.3
July 1994	12 (6)	4 male, 2 female	27.4, 29.4, 29.9, 25.5, 27.3, 25.2
May 1995	14 (10)	Full details are provided in section 3.8.2	

Number caught = the total number of animals caught, the value in brackets indicates the number killed. Note that large numbers of juvenile animals were released.

and standard errors. The output from the Mann-Whitney U test for sex differences within the mammal data set is presented in Table B1 (Appendix B).

3.8.1 Spatial variation

With the exception of the fourth and final trapping session in May 1995, traps were set along the two edges of the woodland on the hypothesis that, given the spatial distribution of the radionuclides in the soil and leaf litter, differences might be observed in the body burdens of the small mammals inhabiting the woodland's different fringes. However, statistical analysis of the ^{137}Cs and actinide data for samples collected in September 1993, using the Mann-Whitney U test, indicated no significant differences between the body burdens of animals caught in the two areas ($p>0.05$). The Mann-Whitney U test output is given in Table B2 (Appendix B).

Following a pilot radiochemical separation using the September 1993 catch from Lady Wood, mammal samples were combined to increase the mass of sample. This led to a reduction in counting times on the alpha spectroscopy PIPS detectors. Furthermore, the increased mass of sample reduced the impact of any loss during the separation procedure, a problem observed in the pilot study. This reduced the number of replicates but the improvements in preparation chemistry and sample counting justified this decision. ^{137}Cs levels were determined by gamma spectrometry and the samples were then paired for radiochemistry. Samples were paired using random number tables. The absence of statistical differences between the sexes and trapping locations within Lady Wood allowed all the mammals to be pooled. This protocol was then employed for all subsequent measurements of actinides in small mammals collected from the field.

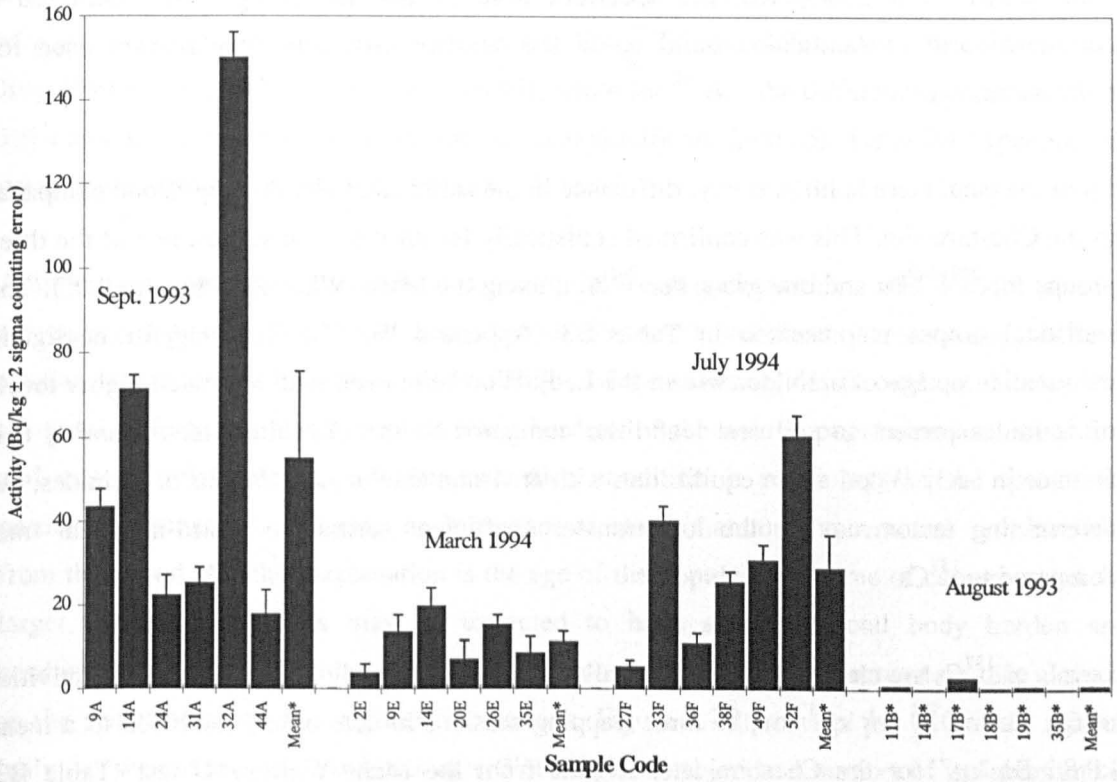
The data presented in Table 3.31 and Figures 3.24 and 3.25 show markedly higher concentrations of the more biologically mobile ^{137}Cs than of the actinides, ^{238}Pu, $^{239+240}Pu$, and ^{241}Am. This is not surprising, although the actinide data from Lady Wood are very low and not much greater than for the Cheshire reference site, where the only actinides present derive from global fallout from weapons testing and the Chernobyl accident. This was surprising, given the difference between Lady Wood and Cheshire in the actinide levels for soil, vegetation and leaf litter (sections 3.4.3, 3.5.2 and 3.6). For example, the mean activity data for Lady Wood mammals ranged from 0.07 to 0.25 Bq kg^{-1} for ^{238}Pu, 0.14 to 0.56 Bq kg^{-1} for $^{239+240}Pu$, and 0.16 to 0.41 Bq kg^{-1} for ^{241}Am over the three trapping campaigns, compared to means of 0.42, 0.10 and 0.05 Bq kg^{-1} for these radionuclides respectively for *A. sylvaticus* caught in Cheshire. The high mean for ^{238}Pu is strongly influenced by one high,

Table 3.31: Radionuclide activities in each animal taken from Lady Wood. All values in Bq kg^{-1} (± 2 σ counting error) except means which are ± standard error.

Sample Code	*^{137}Cs*	*Combined with*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*
9A	43.5 ± 4.2	-----	0.11 ± 0.02	0.48 ± 0.04	0.24 ± 0.05
14A	71.3 ± 3.5	-----	0.11 ± 0.02	0.29 ± 0.03	0.56 ± 0.11
24A	22.1 ± 5.0	-----	0.18 ± 0.03	0.28 ± 0.03	0.35 ± 0.07
31A	25.2 ± 3.7	-----	0.12 ± 0.02	0.45 ± 0.03	0.38 ± 0.07
32A	150.0 ± 5.9	-----	0.07 ± 0.01	0.12 ± 0.02	0.31 ± 0.06
44A	17.8 ± 5.8	-----	0.32 ± 0.04	1.74 ± 0.08	0.61 ± 0.12
Mean (A)	55.0 ± 20.6	-----	0.15 ± 0.003	0.56 ± 0.01	0.41 ± 0.01
2E	3.7 ± 2.4	14E	0.58 ± 0.05	0.14 ± 0.03	0.13 ± 0.03
9E	13.4 ± 4.3	35E	0.08 ± 0.02	0.13 ± 0.02	0.34 ± 0.07
14E	19.6 ± 4.5	-----	-----	-----	-----
20E	7.1 ± 4.5	26E	0.09 ± 0.02	0.16 ± 0.02	0.17 ± 0.04
26E	15.2 ± 2.5	-----	-----	-----	-----
35E	8.6 ± 3.9	-----	-----	-----	-----
Mean (E)	11.3 ± 2.4	-----	0.25 ± 0.01	0.14 ± 0.001	0.21 ± 0.01
27F	5.4 ± 1.6	33F	0.10 ± 0.02	0.08 ± 0.01	0.03 ± 0.01
33F	40.1 ± 3.7	-----	-----	-----	-----
36F	10.9 ± 2.4	49F	0.07 ± 0.02	0.24 ± 0.03	0.14 ± 0.03
38F	25.4 ± 2.6	52F	0.05 ± 0.01	0.19 ± 0.03	0.31 ± 0.05
49F	30.7 ± 3.8	-----	-----	-----	-----
52F	60.4 ± 5.1	-----	-----	-----	-----
Mean (F)	28.8 ± 8.2	-----	0.07 ± 0.001	0.17 ± 0.01	0.16 ± 0.01
11B	<0.86	17B	1.03 ± 0.07	0.15 ± 0.03	0.06 ± 0.02
14B	<0.12	35B	0.17 ± 0.03	0.07 ± 0.02	0.04 ± 0.00
17B	<2.90	-----	-----	-----	-----
18B	<0.46	19B	0.07 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
19B	<0.74	-----	-----	-----	-----
35B	<0.08	-----	-----	-----	-----
Mean (B)	<0.86	-----	0.42 ± 0.02	0.10 ± 0.003	0.05 ± 0.01

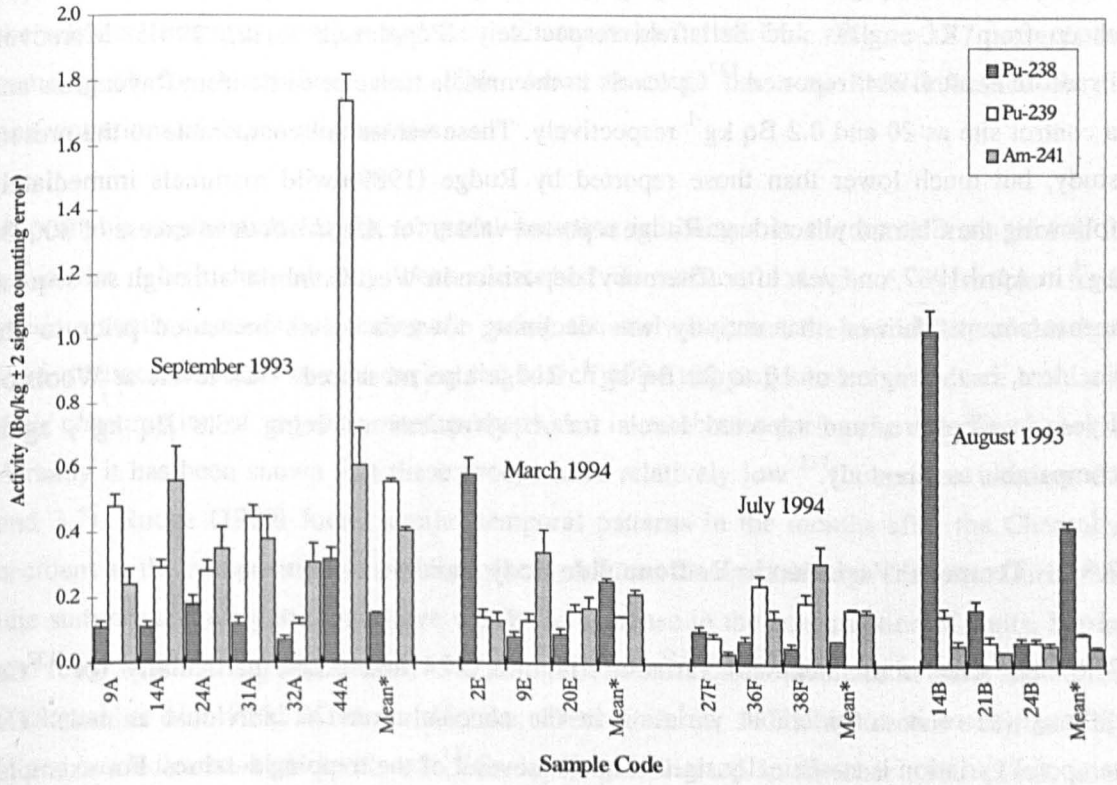
A = September 1993, E = March 1994, and F = July 1994. B = August 1993 trap in Cheshire.

Figure 3.24: ^{137}Cs in the field mouse, *Apodemus sylvaticus*, caught in Lady Wood.



* Error reported is standard error of the mean. All the control values are < LOD.

Figure 3.25: ^{238}Pu, $^{239+240}$Pu and ^{241}Am in the field mouse, *Apodemus sylvaticus*, caught in Lady Wood.



* Error reported is standard error of the mean. All the control values are < LOD.

anomalous, value which, although the data calculations were confirmed, may be a consequence of a poorly resolved spectrum peak or due to 'background' counts as a consequence of contamination build up in the detector chambers or glassware used for radiochemistry.

From the data, there is little, if any, difference in the actinide uptake in Lady Wood compared to the Cheshire site. This was confirmed statistically for all the ^{238}Pu values, two of the three groups for $^{239+240}$Pu and one group for ^{241}Am, using the Mann-Whitney U test ($p>0.05$). The statistical output is presented in Table B3 (Appendix B). The data suggest negligible mammalian uptake of actinides within the Lady Wood site even with the much higher levels of actinides present in soil and leaf litter compared to the Cheshire site. Assuming that animals in Lady Wood are in equilibrium with environmental concentrations of actinides, the determining factor may be the low transfer coefficient across the gastro-intestinal tract compared to ^{137}Cs.

Levels of ^{137}Cs are clearly elevated in small mammals from Lady Wood, with mean activities of 55, 11 and 29 Bq kg^{-1} for the three trapping sessions in date order, compared to a mean <0.86 Bq kg^{-1} for the Cheshire site. Results from the Mann-Whitney U test (Table B3, Appendix B) show very clear, significant differences ($p<0.01$) between the Lady Wood and Cheshire sites. The uptake and transfer of ^{137}Cs in animals has been reported by several authors and is known to be greater than for the actinides. ^{137}Cs activities in sheep and cattle of 7.8 Bq kg^{-1} (wet weight) and 13.8 Bq kg^{-1} (wet weight) have been reported in the muscle of sheep from Ravenglass and Sellafield respectively (Popplewell *et al.*, 1981). Moreover, Bradford *et al.* (1984) reported ^{137}Cs levels in the muscle tissue of cattle from Ravenglass and a control site as 20 and 0.2 Bq kg^{-1} respectively. These values are comparable to the present study, but much lower than those reported by Rudge (1989) in wild mammals immediately following the Chernobyl accident. Rudge reported values for *A. sylvaticus* in excess of 300 Bq kg^{-1} in April 1987, one year after Chernobyl deposition in West Cumbria, although subsequent measurements showed that activity was declining towards levels measured prior to the accident, in the region of 10 to 20 Bq kg^{-1}. Rudge also measured ^{137}Cs levels at Woolston Eyes, in Cheshire, and reported levels for *A. sylvaticus* as being <3.8 Bq kg^{-1}, again comparable to this study.

3.8.2 Temporal Variation in Radionuclide Body Burdens

The data show some temporal variation (Figures 3.24 and 3.25), particularly for ^{137}Cs, although there is considerable variation in the concentrations in individual animals. The temporal variation is statistically significant for several of the trapping sessions. For example, ^{137}Cs exhibits a temporal difference between September 1993 and March 1994 ($p<0.01$).

Differences between the March and July 1994 data were marginally non-significant ($p>0.05$). Similar differences were noted for ^{238}Pu and ^{241}Am, although uptake of the actinides is limited at Lady Wood compared to the Cheshire site. For ^{238}Pu, the difference between March and July 1994 is statistically significant ($p<0.05$), while for ^{241}Am the differences between March 1994 and the next two trapping sessions are also significant ($p<0.05$). Table B4 (Appendix B) presents the output from the Mann-Whitney U tests.

Animals are exposed to a continuous active influx of radionuclides and also to activity previously deposited throughout Lady Wood. Since the woodland is reasonably uniform, similar parameters will influence bioavailability of the radionuclides throughout the site. The temporal differences in radionuclide body burdens of the animal population are interesting and may be related to the age-structure, diet, condition or physiology of the species. In particular, changes in diet may be important as small mammals accumulate radionuclides from their food. Another explanation is the age of the population at time of capture, whereby larger, mature individuals may be expected to have a greater total body burden and concentration of radionuclides. Within this study, most of the effort focused on diet changes as the principal cause of temporal fluctuations. Figure 3.26 presents the ^{137}Cs data against body wet weight as a crude index of age. Wet weight is subject to a number of variables and is not regarded as the best index for ageing small mammals. It is, however, sufficient to show that age might be a contributory factor to variation in ^{137}Cs levels. The trapping sessions between September 1993 and July 1994 show a general trend whereby ^{137}Cs activity in mammals increases with wet weight. It is not possible to be absolutely definitive because there is insufficient replication and the data are somewhat variable. Figure 3.27 presents the same graph for the actinide data. This shows that age has very little influence on the concentrations of these radionuclides.

A probable explanation for the temporal variation is changes in the omnivorous diet of *A. sylvaticus* which has already been described in section 1.4.4.5. *A. sylvaticus* feeds predominantly on seeds but, during the spring and early summer, the availability of this food item declines. Animals caught during the March 1994 trapping session may have increased their consumption of vegetation and perhaps also invertebrates during the antecedent period. Already it has been shown that these groups have relatively low ^{137}Cs burdens (sections 3.6 and 3.7). Rudge (1989) found similar temporal patterns in the months after the Chernobyl accident as the composition of the diet of *A. sylvaticus* changed through the year. During the late summer and early autumn, there will be an increase in the consumption of fruits, berries and fungi (Churchfield and Brown, 1987; Watts, 1968). ^{137}Cs accumulates in fruits and berries (Bukovak *et al.*, 1965; Bunzl and Kracke, 1986; Gamble, 1971) and recent work in Lady Wood has indicated high levels of ^{137}Cs in fungi collected in October 1995 (Toal, pers.

Figure 3.26: **^{137}Cs data presented in ascending order of wet weight (reported) as an index of age for each trapping session.**

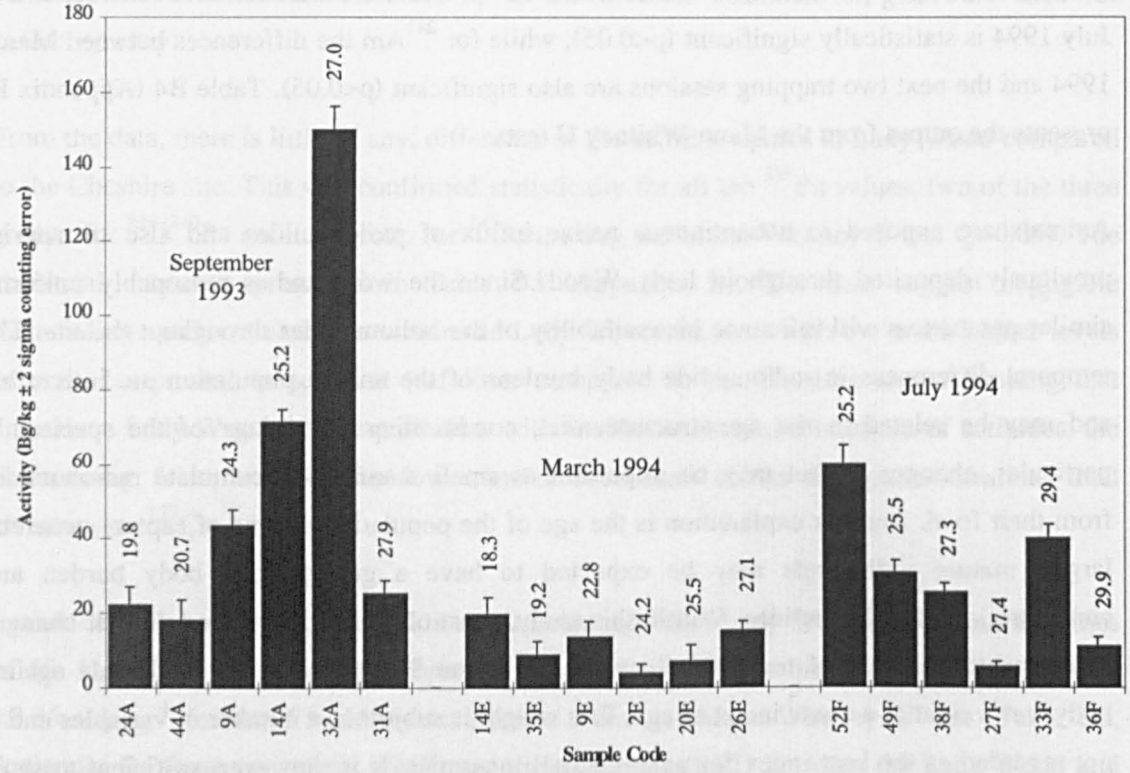
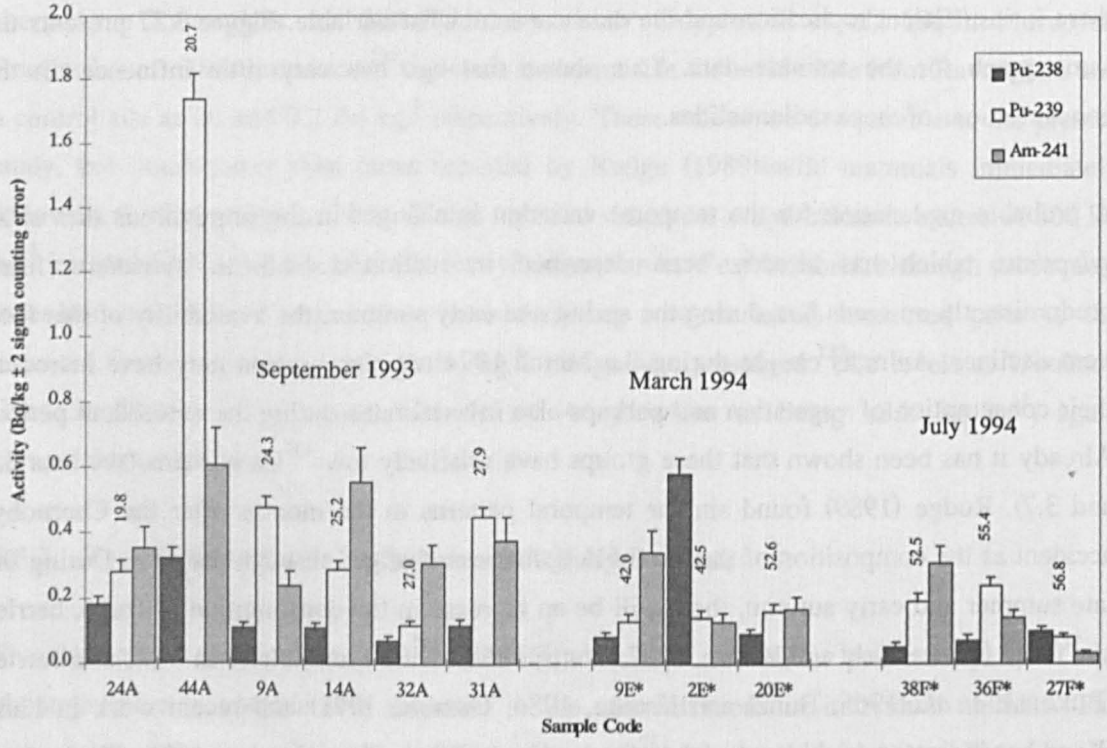


Figure 3.27: **Actinide data presented in ascending order of wet weight (reported) as an index of age for each trapping session.**



* Combined mammal weights are reported.

comm.). Animals caught in September 1993 may well have selected these higher activity foodstuffs.

The actinide data show little fluctuation during the study period, with the exception of September 1993 compared to other sessions. Activity data for plutonium and americium were significantly higher during September 1993 than later in the year, perhaps again reflecting changes in dietary composition, particularly with fruits, berries and seeds. During this study, extensive study of fruits, berries and seeds was excluded from the sampling strategy so there is currently no data available for these components from Lady Wood. It is postulated from other work that there will be a seasonal increase in the concentrations of ^{238}Pu, $^{239+240}Pu$ and ^{241}Am during the autumn, in much the same way as ^{137}Cs is known to behave (Bunzl and Kracke, 1986; Liden and Gustafsson, 1967; Lindner *et al.*, 1992; Rudge, 1989).

3.8.3 Tissue Distribution of ^{137}Cs

Animals caught during the last trapping session were dissected to provide information about the tissue distribution of radionuclides. It was necessary to bulk the component parts: body hair, gut (with contents), lungs, muscle, organs (comprising kidneys, liver, spleen, heart and sexual organs) and skeleton, from different animals to maintain the sample mass used previously for radionuclide determinations. Of the ten animals caught, five were combined to produce two replicates for each sample category. During the subsequent analysis, time constraints limited the ability to measure activity parameters other than ^{137}Cs.

Table 3.32 presents data from six body components. Random number tables were again used to select the combination of animals, and males and females were pooled randomly. Spatial differences should be negligible in this case, because the traps were set only along the leading edge of the woodland. Prior to analysis, small mammals caught during the first three trapping campaigns were washed in a dilute 0.2% solution of 'Teepol' to remove any surface contamination. It was expected that there would be soil particles present on the body hair as a result of resuspension of soil by the animals' movement, particularly when underground. However, the results from hair analysis reported in Table 3.32 suggest otherwise, with ^{137}Cs activities around 100 Bq kg^{-1} for unwashed animals. It is possible that the natural grooming activity was accentuated as a displacement response whilst the animals were confined to the trap prior to being killed. Even in the gut, however, the activity levels are much lower than expected originally. When the animals were dissected, the stomach and intestine were removed to produce the sample termed 'gut'. It was noted that the stomachs were bloated due to gorging on the bait placed in the Longworth traps. This will have elevated the weight of the 'gut' samples and diluted any radionuclides present. Such high ingestion rates will also have caused more material to be defecated. The latter will have comprised the natural food items:

Table 3.32: ^{137}Cs in tissues and organs of *A. sylvaticus*.

Tissue	*Combined Wet Weight (g)**	*^{137}Cs (rep 1) Bq kg^{-1}***	*^{137}Cs (rep 2) Bq kg^{-1}***	*^{137}Cs Bq kg^{-1} ± standard deviation*
Body Hair	3.06	105 ± 26	72 ± 30	89 ± 24
Gut (full)	67.35	330 ± 22	130 ± 5.0	230 ± 140
Lungs	3.45	870 ± 110	390 ± 88	630 ± 340
Muscle	10.30	2100 ± 50	550 ± 46	1300 ± 1100
Organs	35.24	930 ± 24	470 ± 18	700 ± 330
Skeleton	5.20**	4.4 ± 2.1	8.4 ± 3.5	6.4 ± 2.8

* All ten animals. ** Dry weight reported. Replicate values are reported ± 2σ counting error.

vegetation, invertebrates, seeds etc. thus, again, artificially reducing the radionuclide content of the gut. Had 'snap-traps' been used instead of live traps, the radionuclide content of the gut samples may have been higher.

Given the behaviour of ^{137}Cs within the body, it was believed that the bulk of the activity would be located in the soft tissues such as muscle, and the various organs extracted, including the spleen, liver, kidneys, and heart. The results support this hypothesis with high levels of ^{137}Cs in the muscle and organs, 470 - 2,100 Bq kg^{-1} but very low ^{137}Cs in bone, only 4 Bq kg^{-1}. In particular, the muscle showed very high concentrations of ^{137}Cs, up to 2,100 Bq kg^{-1}. The variation between replicates was very high, in most samples, but this does not negate the fact that ^{137}Cs does accumulate within soft tissues, and that muscle is particularly important in this regard (ICRP, 1979; Muller and Scheffer, 1982; Richmond, 1980). More replication is needed to cater for the high variation but the data provide a starting point for assessing the dose received internally from the various radiologically significant radionuclides.

The activity results for the lungs are of interest because adsorption of, and the dose received from, radionuclides within the lungs may be an important component of the radiological stress upon small mammals in contaminated ecosystems. Based on concentration data, the activity of ^{137}Cs within the lungs is third in the hierarchy behind the levels of ^{137}Cs in muscle and various pooled organs. Soft tissues will have accumulated radionuclides absorbed across the gastro-intestinal tract; but there will also be a component absorbed across the epithelial lining of the lungs. Activity within the lungs at any given time will depend upon a wide range of parameters, not least how efficiently the ciliated epithelial lining is in removing the deposited particles and associated radionuclides. Given the high activity for ^{137}Cs, it may be that the actinides also show elevated levels in the respiratory system. This could be even more critical because alpha emissions within the lungs can cause considerable radiation damage to surrounding tissues (section 1.4.4.3). Further analysis is required to confirm the tissue

distribution of the actinides, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am, within the body of the small mammal species, *A. sylvaticus*.

Back calculation of the ^{137}Cs levels from the values for individual body components listed in Table 3.32 produced estimates of the whole body burden between 45 and 80 Bq kg^{-1}. It should be noted that these are likely to be underestimates because sample material was discarded during dissection and in the preparation of the skeleton. The estimates are comparable to the body burdens listed in Table 3.31 for individual animals. Therefore the tissue distribution of ^{137}Cs reported should be representative of the mammals caught previously.

3.8.4 Isotopic and Nuclide Ratios for *A. sylvaticus*

Table 3.33 shows calculated isotopic (^{238}Pu:$^{239+240}Pu$) and nuclide (^{137}Cs:$^{239+240}Pu$ and ^{137}Cs:^{241}Am) ratios for *A. sylvaticus* caught in Lady Wood. Lack of ^{134}Cs data prevented calculation of the ^{134}Cs:^{137}Cs ratio. For comparison the ratios obtained for animals caught at the Cheshire reference site are also presented. These latter nuclide ratios are clearly much lower than those reported for Lady Wood mammals.

Examining the ^{238}Pu:$^{239+240}Pu$ ratio, it can be seen that ratio varies considerably over the trapping sessions and that it is higher than that recorded for both soils and leaf litter (Tables 3.5 and 3.13). Much of this is due to the very low transfer of plutonium through the food chains at this site as recorded previously for the invertebrates (Table 3.28). The high standard error reported reflects the large biological variation between individual animals.

The nuclide ratios, ^{137}Cs:$^{239+240}Pu$ and ^{137}Cs:^{241}Am, are much greater than those reported for soils (Table 3.4) and the invertebrates (Table 3.28). This is also true for the leaf litter samples. This was expected because of the low transfer of the actinides, relative to caesium, across the mammalian gut. Given that the ratios in soil were of the order of 10 and 20 for ^{137}Cs:$^{239+240}Pu$ and ^{137}Cs:^{241}Am respectively, it is clear that ^{137}Cs is transferred to the mammals more effectively.

Table 3.33: Isotopic and nuclide ratios for *A. sylvaticus* from Lady Wood.

Trapping Session	^{238}Pu:$^{239+240}Pu$	^{137}Cs:$^{239+240}Pu$	^{137}Cs:^{241}Am
September 1993	0.38 ± 0.08	288 ± 195	158 ± 69
March 1994	1.77 ± 1.18	79 ± 5	63 ± 17
July 1994	0.60 ± 0.32	156 ± 57	348 ± 205
August 1993*	3.43 ± 1.77	8 ± 3	15 ± 9

* Small mammals from the Cheshire reference site. Standard error reported.

3.8.5 Summary of Small mammals results

Activity levels in wood mice (*A. sylvaticus*) from the woodland site were 7 to 150 Bq kg^{-1} (^{137}Cs), 0.1 to 0.3 Bq kg^{-1} (^{238}Pu), 0.1 to 0.6 Bq kg^{-1} ($^{239+240}Pu$) and 0.2 to 0.4 Bq kg^{-1} (^{241}Am). No significant differences ($p>0.05$) were found between sexes in respect of any nuclide, so datasets were pooled for between site comparisons. The range for ^{137}Cs encompasses mean values of 55, 11 and 29 Bq kg^{-1} for the three trapping campaigns, values which compare with <0.9 Bq kg^{-1} for the reference site remote from airborne inputs of industrial or secondary marine-derived radioactivity. Actinide levels at the woodland location were little different from the reference site, where they reflect only residual inputs from historical atmospheric weapons testing.

The clear differences in levels of ^{137}Cs in wood mice from the woodland and reference site reflect not just the differences in contemporary exposure conditions for this nuclide at the two locations, but also the higher mobility and transfer coefficient in food chains of caesium in comparison with members of the actinide group. The clear between-site differences were not matched in temporal patterns, where there were significant ($p>0.05$) but less discriminating seasonal differences in body burdens within the woodland site.

These differences were found in respect of ^{137}Cs, ^{238}Pu and ^{241}Am and are thought to reflect changes in the age structure of the population and also seasonal changes in the diet of wood mice from green and invertebrate material to a diet with a greater prominence of contaminated fruits, berries and especially fungi. The involvement of age-accumulation is evidenced by a positive relationship between body burden of ^{137}Cs and wet weight used as a crude index of age. Body burdens of actinides showed little variation with season, probably due to the low transfer potential of these nuclides regardless of dietary changes over the year.

Activity measurements in single tissues showed high levels of ^{137}Cs in muscle, up to 2,100 Bq kg^{-1}, with elevated levels also in lungs (630 Bq kg^{-1}), and the pooled principal organs (spleen, liver, kidneys and heart) (700 Bq kg^{-1}). Low activity was recorded in the skeleton and gut contents, the latter reflecting the presence of artificial bait. Back calculations gave conservatively estimated whole body concentrations for ^{137}Cs of 45 to 80 Bq kg^{-1}. The concentration of ^{137}Cs in lung tissue suggests that inhalation may be an important route of entry for the actinides.

Chapter Four
A SALT MARSH ECOSYSTEM - RIVER ESK

4.1 INTRODUCTION

4.1.1 Background - previous studies

Salt marshes are dynamic habitats that possess characteristics of both the terrestrial and marine environments. Their formation begins in areas subject to tidal inundation, usually developing from areas of mud flats which are protected from the direct action of waves. In the case of the River Esk salt marshes, they are estuarine in nature and are formed on sheltered inner curves of the estuary. In almost all cases, the areas of developing salt marsh are influenced by the surface relief and hydrology of the tidal flats.

Salt marsh development begins with a very low sedimentation rate in areas of mud flats. As the biological activity within these mud flats increases, so does the rate of sedimentation. Sediment particles then become trapped within algal strands and other organic debris. Filamentous algae, for example, have been shown to trap and then stabilise silt very effectively (Ranwell, 1972). In addition, the biological activity, particularly the burrowing of invertebrate animals, helps to improve the soil aeration and promote the growth of aquatic, and then salt marsh, plant species.

Although salt marshes only cover a small proportion of Great Britain, there is considerable interest in them in the context of radionuclide distribution and behaviour. This is primarily because the accumulated sediment, laid down over time, contains a history of radionuclides released into the environment. Both tidal mud flats and salt marshes have been studied extensively to examine the association of radionuclides with sediment (MacKenzie and Scott, 1993; Oldfield *et al.*, 1993), and to investigate the historical release of radionuclides to the environment (Hamilton and Clarke, 1984; McCaffrey and Thomson, 1980; Murdock, 1996). Sediment cores dated using radiometric methods have also been used to assess the historic releases of heavy metals into the aquatic environment (Jones *et al.*, 1995).

More recently, interest in vegetated salt marshes has developed as these can be important sources of radioactivity. They may be the source of aerosols and/or resuspended material which can be carried inland by wind; also they may be grazed by livestock, and wildfowl that may then be shot and used for food (Howard, 1985; Howard and Lindley, 1985; Lowe and Horrill, 1986; Woodhead, 1986). Salt marshes may be reclaimed for agricultural land such as in the Ribble Estuary, Lancashire; and as levels of radionuclides discharged from nuclear establishments decline, radionuclide material present within the salt marshes may assume greater relative importance as it is released and recycled (Burton and Yarnold, 1988; Mudge *et*

al., 1988). However, little information is available on the transfer behaviour of radionuclides between the natural flora and fauna of salt marshes. The plant species that colonise salt marshes are often coated with sediment and are therefore surrounded by radionuclides. Moreover, much of the invertebrate activity associated with salt marshes can be found within the strand line which consists of accumulated plant debris of salt marsh species. Small mammals, particularly the shrews, *Sorex araneus* and *Sorex minutus*, feed within the strand line and the field vole, *Microtus agrestis*, and the field mouse, *Apodemus sylvaticus*, forage in a corridor above and around the strand line.

4.1.2. Study Objective

The primary aim of this study was to investigate the behaviour and transfer of radionuclides through food chains in a salt marsh ecosystem which was receiving a continual input of radionuclides through tidal inundation. This was achieved through the analysis of sediment, vegetation, strand material, invertebrates and small mammals collected over a period of two years. The following discussion includes a description of the spatial distribution of the radionuclides measured (^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am) and evidence for temporal variation. Transfer factors for the radionuclides to the invertebrate and small mammal species are presented.

4.1.3 Deposition Processes

Following the release of radioactivity from BNFL, Sellafield to the Irish Sea, the radionuclides are dispersed throughout the aquatic environment and may be returned to the terrestrial environment through the processes of sea to land transfer, described briefly in Chapters 3 and 5, and through tidal inundation of areas of land such as salt marshes. The actual dispersion and mobility of the radionuclides is dependent upon their interactions with the water body itself. The two isotopes of caesium released from the pipelines remain mostly in solution and are rapidly dispersed by the circulation patterns of currents in the Irish Sea (Jefferies *et al.*, 1973, 1982; McDonald *et al.*, 1990). The residual current in this region carries the radioactivity southward along the Cumbrian coastline towards Liverpool Bay (Jefferies *et al.*, 1973). This has obvious implications for the transfer of radioactivity to the Esk Estuary. A fraction of the caesium will become attached to particles in the sea, and measurements have shown that between 5 and 30% of the seawater inventory was associated with suspended sediment (McKay *et al.*, 1987).

The behaviour of the actinides, plutonium and americium, contrasts notably with that of caesium. It has been shown that these radionuclides are non-conservative in their behaviour and are mainly deposited locally on the sea bed around the discharge pipeline (Hetherington,

1978; McCartney *et al.*, 1994; Pentreath *et al.*, 1983). It is thought that 95% of the discharged plutonium is lost from the water column close to the end of the pipeline (Hetherington, 1978). This may be because both the plutonium and americium are attached to a ferric hydroxide floc in the effluent, that is deposited to the sea bed. The actinides may then slowly desorb from the floc to the sediment (Howorth and Eggleton, 1988; Pentreath *et al.*, 1983). Subsequent transport of the actinides is then dependent upon general sediment transport processes (Hunt, 1985; Mackenzie *et al.*, 1987; Pentreath *et al.*, 1986). The effect of particle size on the adsorption of both the actinides and caesium has already been described in section 1.4.1. The net effect of this is that the specific activity of radionuclides increases with decreasing particle size, so that the fine clays are generally more active than sands (Aston and Stanners, 1982; Hetherington and Jefferies, 1974; Hetherington, 1978).

When the sediment is resuspended from the sea bed, it will be subject to the general circulation patterns within the Irish Sea and as such will also be moved southwards along the Cumbrian coastline towards the Esk Estuary. A small proportion (<5%) of the plutonium and americium remains unattached to particles and will behave in much the same way as the discharged caesium (McKay *et al.*, 1987).

Within the Esk Estuary itself there have been several studies which have investigated the transport and deposition of radionuclides to salt marshes, for example: Assinder *et al.*, 1985; Davis and Shaw, 1993; Hamilton and Clarke, 1984; Kelly and Emptage, 1991. These studies have shown that it is the areas of fine grained sediments which act as the main reservoirs for radionuclides. For example, Kelly and Emptage (1991) estimated that mud banks, salt marshes and intertidal pasture contribute 46% to the total volume of sediment within the estuary and yet contain 76% of the radionuclide inventory. These studies have also indicated that changes in the physico-chemical conditions in estuarine environments may result in caesium becoming more strongly adsorbed to sediments (Santschi *et al.*, 1983). This transfer of radionuclides to salt marsh sediment has been shown to play an important role in the return of radioactivity to the terrestrial environment. In particular it forms an entry route into food chains (Howard, 1985, 1987; Howard and Lindley, 1985), although the actual rate of transfer is determined by the physico-chemical conditions in which the radionuclides reside.

4.2 SITE DESCRIPTION

There are several areas of ungrazed mature or high salt marsh (Long and Mason, 1983) located within the Esk Estuary, Cumbria. The area selected for this study was chosen partly because of prior knowledge of the radionuclide inventory and history of the site (Aston and Stanners, 1981, 1982; Hamilton and Clarke, 1984; Horrill, 1983; Kelly and Emptage, 1991), partly because of the size of the marsh, and also because it is dissected by a railway viaduct

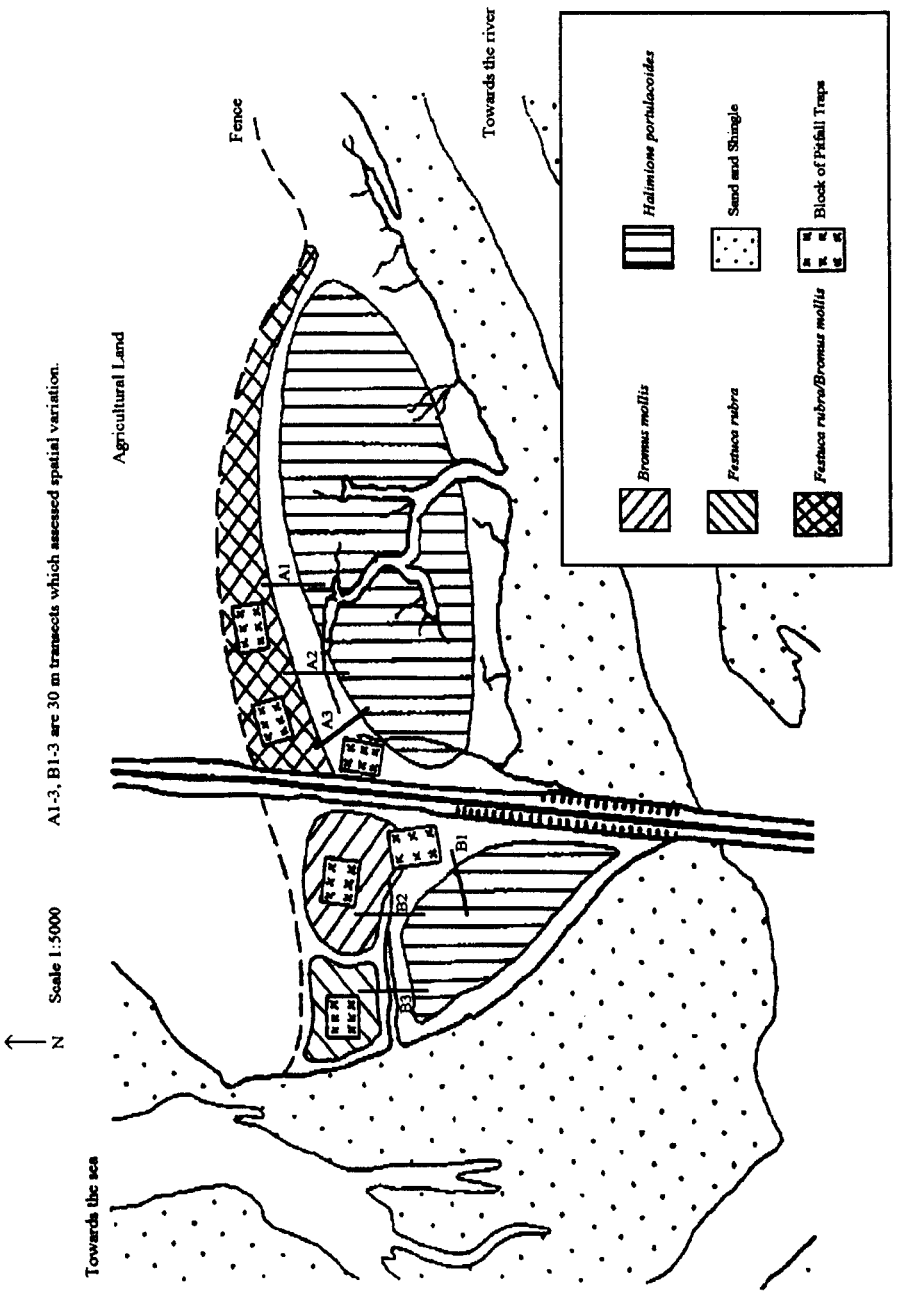
which conveniently forms two sampling areas. The selected area (OSGR: SD 089948) covers approximately 5.4 ha on the riverward side (A) and ≈3.8 ha on the seaward side (B). Figure 4.1 presents an outline sketch of the study site. The history of the Esk Estuary has been described elsewhere (Kelly and Emptage, 1991).

The marsh is approximately 12 km south of BNFL, Sellafield, and receives a continual low level radionuclide input from tidal inundation throughout the year. The marsh is regularly inundated on a daily basis. Given its proximity to the sea, it was expected that limited sea to land transfer would also occur. The salt marsh is separated from the sea by the River Esk itself and also by a strip of sand dunes, approximately 1 km wide, which are dominated by marram grass, *Ammophila arenaria*. Some aerial deposition also occurs across the site but the dominant mechanism for deposition of radionuclides is tidal inundation. Horrill (1983) showed the importance of tidal inundation across this site following determination of the spatial distribution of a range of radionuclides. It was shown, for example, that short lived radionuclides such as ^{95}Nb and ^{95}Zr exhibit greatest deposition in areas most frequently inundated, unlike the long lived radionuclides such as ^{137}Cs and ^{241}Am which also accumulate in the older regions of the marsh. Horrill (1983) concluded that the radionuclide accumulation across the marsh was dominated by physical processes, particularly tidal inundation. In addition, the form of the vegetation is important in determining deposition rates.

The study site is privately owned by the Muncaster Estate and has been undisturbed for at least forty years. It is believed that the salt marsh began to form after the railway viaduct was built as it created areas of slack water which generated an increased sedimentation rate leading to the marsh development. To the rear of the marsh, there is agricultural land used for crops and grazing. Plates 4.1 and 4.2 show the two sampling corridors on either side of the railway viaduct.

On both sides of the viaduct, similar vegetation species were found on and around the margins of the marsh. On both sides, the marsh plant community was dominated by sea purslane, *Halimione portulacoides*, with marginal areas co-dominated by red fescue, *Festuca rubra*, and soft brome, *Bromus mollis*. A list of common species found growing across the site is given in Table 4.1. In terms of food chain transfer, those species growing around the margin of the marsh which are infrequently inundated are favoured in the diet of the herbivorous and omnivorous small mammal species. Small mammals are unlikely to traverse and feed within the marsh area itself partly because the vegetation is usually covered with sediment and also because of the greater risk of predation due to increased exposure. Moreover, tidal inundations keep the marsh almost permanently wet and unattractive to small mammals. Beeftink and Rozema (1988) have reported that wild mammals usually visit marshes for foraging only and

Figure 4.1: **Outline sketch of the River Esk salt marshes indicating the sampling locations and vegetation distribution.**



find residence habitats in the uppermost regions beyond the tidal reach. As a result, a study site was marked out on each side of the railway viaduct to include the very top of the marsh, the strand line and the vegetation above the strand line up to the edge of the agricultural land. This area is rarely inundated completely by the tide.

There is a strand line around the edges of the marsh. The area covered by, and the depth of, the strand layer depend upon the time of year and the height of the most recent inundation. The strand material consisted mainly of leaves of *H. portulacoides* and other detritus from the marsh vegetation plus biological and non-biological debris washed in from the sea, and from upstream along the river. There is considerable invertebrate activity within this strand layer and this community surely forms the bulk of the diet for insectivorous shrews, *S. araneus*, and part of the diet of field mice, *A. sylvaticus*, during the spring and summer periods. Stewart *et al.* (1989) showed that *Sorex* species travelled up to 60 m to forage in the littoral region of a beach.

It is probable that much of the radionuclide burden of the vegetation in the marginal region arises from occasional direct tidal inundation, wind blown water, strand material and from historic deposition across the site when the marsh was younger. Under normal tide conditions, only the front part of the marginal area is inundated, as indicated by the configuration of the strand line. However, on three occasions during the study period strand material was found right up to the boundary of the neighbouring agricultural land. This arises from accentuated high tide events whereby heavy rainfall and strong winds from specific directions cause deposition of debris beyond the normal range.

From the observations of Horrill (1983) it was thought that the deposition of ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am would increase along a transect up the marsh to a point when the activity levels would start to fall because regular inundations no longer occurred. It was also expected that the actual distribution of radioactivity would be modified by the presence of deep water channels (Plates 4.1 and 4.2; Figure 4.1).

The marsh has formed through the increased sedimentation of material in parallel with colonisation of mud flats by tolerant species of vegetation. The marsh is still expanding further into, and above, the base of the river channel. It is approximately two metres above the base of the river channel in its upper reaches and as such only the highest tides reach this area. However, there are deep channels or creeks which dissect the marsh, and whose cliff surfaces undercut the marsh development. Across the bulk of the area the vegetation was visibly covered with sediment, grey coloured when dry. Near the sampling area, approximately 95% of the marsh is covered with vegetation, as are the margins. The exposed bare marsh is mainly in the form of patches near to, and within, the creeks.

Plate 4.1: **Photograph taken of the riverward side of the salt marsh site. The vegetation distribution and large creek discussed in the text is clearly shown.**

Plate 4.2: **Photograph taken of the seaward side of the salt marsh site.**

Table 4.1: Common vegetation species found at the River Esk salt marsh study site.

Salt marsh Vegetation	
Sea Purslane	*Halimione portulacoides*
Sea Rush	*Juncus maritimus*
Glasswort	*Salicornia europaea*
Common Cord Grass	*Spartina anglica*
Sea Aster	*Aster tripolium*
Sea Plantain	*Plantago maritima*
Common Salt marsh Grass	*Puccinellia maritima*
Vegetation Found Around the Margins	
Sweet Chestnut	*Castanea sativa*
Bramble	*Rubus fruticosus*
Hawthorn	*Crataegus monogyna*
Blackthorn	*Prunus spinosa*
Hazel	*Corylus avellana*
Common Vetch	*Vicia sativa*
Soft Brome	*Bromus mollis*
Common Scurvy Grass	*Cochlearia officinalis*
Dandelion	*Taraxacum officinale*
Red Fescue	*Festuca rubra*
Gorse	*Ulex europaeus*
White Deadnettles	*Lamium album*

It was originally thought that the two sides of the salt marsh separated by the railway viaduct would be broadly similar in their properties and therefore in the activity of the radionuclides. However, this is now known not to be entirely the case. The differences are due partly to variations in the tidal inundation on the two sides and partly because the carrying capacity or kinetic energy of the water is different as a consequence of the water flowing around the railway viaduct. For example, tidal inundation on side B usually covers more of the marginal area than on side A, and because the water levels rise from the seaward direction, the water flowing over the marsh on side B is generally more energetic. As a consequence, there is less deposition of material on side B compared to A.

For reference purposes, soil, vegetation, invertebrate and small mammal samples were collected from the University of Liverpool's Botanical Gardens at Ness, Wirral (OSGR: SJ 304754). The nearest nuclear establishment is BNFL, Capenhurst which releases no radionuclides of relevance (section 3.2). However, since terrestrial soils are physico-chemically different from salt marsh sediment, some additional samples of soil and vegetation were collected from a salt marsh in Wales: Llandridian Marsh (OSGR: SS 488934). This site is owned by National Trust and was sampled during August 1994. The Welsh site was selected using data from the MAFF aquatic monitoring report (Camplin, 1993), with knowledge of the discharges from nearby nuclear establishments such as Amersham International located near Cardiff, and the Berkeley and Oldbury power stations operated by

Nuclear Electric further along the River Severn. The liquid discharges from these sites were assessed (Table 4.2) and were shown not to include the radionuclides of interest in significant quantities.

Table 4.2: Annual radioactive discharges from nuclear establishments near the Welsh sampling site.

Nuclide	*Site*		
	Amersham	*Berkeley*	*Oldbury*
^{137}Cs	-----	0.122	0.006
Tritium	555	0.200	0.069
Beta/Gamma	0.018	-----	-----
Carbon-14	1.330	-----	-----

Values are in TBq
Data provided for 1991 as an example (Camplin, 1993).

4.3 MATERIALS AND METHODS

4.3.1 Soil Core Sampling

Short sediment cores were obtained on four occasions between April 1993 and September 1994. An area of vegetation and/or strand material was carefully cleared to the surface of the marsh before a 10 cm diameter soil corer was used to extract sediment cores to a depth of 12 cm. The extracted core was compared to the remaining hole for any signs of vertical compaction. In this case, there was none. The corer was washed between extractions. Depending upon the use of the sediment core, either two sets of two cores were collected and bulked together or three cores were extracted and then sectioned in the field into 4 cm slices which were then combined. The sediment samples were then prepared as described for the soils in section 3.3. Typically, a Marinelli beaker held about 450 g of sediment and the samples required short count times of the order 5,000 to 10,000 seconds (1.5 to 3 hours) to obtain sufficient data on ^{134}Cs, ^{137}Cs and ^{241}Am. Sub-samples of 5 g were collected for radiochemical separation using the method described in Appendix A and then analysed for the plutonium isotopes. Organic matter content and pH measurements were also determined on some of the sediment samples. The methods employed are described in section 3.3.

In addition to the short cores, during November 1993 one 50 cm core was taken on each side of the railway viaduct just below the strand line. The core was extracted using the petrol driven vibro-corer described in section 3.3. The samples were then sectioned into 5 cm slices and prepared for radionuclide determination as described in section 3.3.

4.3.2 Strand line material and Vegetation Sampling

Vegetation and strand samples were collected at approximately monthly intervals from April 1993 to May 1994, and then bi-monthly until November 1994. Vegetation samples were collected by clipping to within 2 to 3 cm of the sediment surface to avoid any contamination of the sample. Two species were targeted for analysis: *F. rubra* and *B. mollis*. In addition, less frequently samples of *Juncus maritima* and *H. portulacoides* were collected to assess the recent radionuclide deposition across the salt marsh and below the strand line. Figure 4.1 presents a sketch map of the disposition of the major vegetation species across the marsh. Samples were stored in a cold room (<5°C) until they were sorted into live and senescent material of the same species, and the wet weights recorded. They were then placed in an oven to dry at 85°C for at least 48 hours or until constant dry weight was achieved. Samples were then homogenised in a Waring commercial blender before being placed into a Marinelli beaker for presentation to the germanium detectors. A Marinelli beaker held between 40 and 90g of sample, depending upon the plant species. Depending upon the sampling location, samples typically required between 30,000 and 300,000 seconds (between 8 and 85 hours) to obtain sufficient data on ^{137}Cs and ^{241}Am. ^{134}Cs was rarely measured in vegetation above the detection limits for the germanium detectors used during this study. A sub-sample of 25 g was collected from each sample for radiochemical separation and analysis for the plutonium isotopes of interest and, occasionally, ^{241}Am.

During August 1993, a series of additional samples of sediment and vegetation were collected after the preliminary results from the first three months of sampling indicated that there was a problem with the sampling strategy chosen. Originally, sampling involved the collection of 4 replicates per side, selected at random from a study area which was approximately 10 m wide by 40 m long centred around the strand line material. However, it became obvious that sampling randomly within this area meant that some samples were collected from areas recently inundated and therefore contained significant quantities of radionuclides while some were sampled above the strand line and contained barely any radioactivity. Consequently, the sampling strategy was changed, and post-August 1993 two samples of sediments and vegetation were collected from three parallel transects marked out using the position of the strand line material (Figure 4.1). The number of replicates per transect was limited to two because of the increase in the number of samples requiring analysis.

The samples taken during August 1993 were collected on each side of the railway viaduct at 5 m intervals along three 30 m 'marsh' transects which ran perpendicular to the back edge of the study area. The positions of the three transects are marked on Figure 4.1 and provide information about the spatial distribution of radionuclides from the field boundary towards the river.

Two strand samples per side, consisting mainly of shed leaves from *H. portulacoides* and other plant detritus, were collected on each sampling visit. The samples were sorted to remove any non-biological material, the wet weights were recorded and the samples were dried in an oven at 105°C for 48 hours or until constant weight was achieved. The samples were then homogenised in a Waring commercial blender before being placed into a Marinelli beaker for presentation to the germanium detectors. Typically, the Marinelli beakers held between 70 and 90 g of material and count times of 5,000 to 10,000 seconds were required to measure ^{134}Cs, ^{137}Cs and ^{241}Am. A sub-sample of 25 g was collected from each sample and analysed for plutonium.

Comparison soil and vegetation samples were collected from Ness Gardens, Wirral during August 1993 and August 1994, and from Llandridian Marsh, Wales in August 1994. The samples were collected and prepared as already described. All the samples were decay-corrected to the date of sampling in later analysis of the data.

4.3.3 Sampling of Invertebrates

Invertebrates were collected using pitfall traps. Full details are given in section 3.3. In this case, 3 blocks of 6 pitfall traps were set (Figure 4.1), placed around the strand line on each side of the railway viaduct. The traps were sunk into the ground until they were level with the surface of the marsh and then charged with 100 ml of 2% formalin solution. Ingress of water was prevented by the use of a hardboard lid. The pitfalls were first exposed in April 1993 and remained in place until November 1994. They were changed regularly, usually at monthly intervals, but during hot weather they were changed or recharged more frequently as described in section 3.3. The pitfalls were lifted and re-set when tidal inundations over a pre-determined height (9.5 m) were expected. For the majority of the time, this was successful in preventing flooding. However, during the winter of 1993 and the spring of 1994, unexpected high tides (mainly as a consequence of heavy rainfall and strong winds) swamped the traps and resulted in the loss of sample material. This was the primary reason for the early lifting of pitfalls in November 1994.

Originally, it was intended that each block of six pitfalls would form one replicate. Figure 4.1 shows the three pitfall blocks along each side of the viaduct. Unfortunately the low sample biomass for most of the invertebrate species and the loss of samples in the field necessitated the pooling of all 18 pitfall traps on each side of the viaduct. Samples were also pooled quarterly in an effort to generate sufficient material for counting. After collection, the samples were stored in fresh 2% formalin solution as described in section 3.3 until they were sorted into appropriate taxonomic groupings (section 4.7) and then prepared for radionuclide determination as previously. Typically, invertebrate samples were counted for between 85,000

and 200,000 seconds to obtain measurements of ^{134}Cs, ^{137}Cs and ^{241}Am. A sub-sample was taken for radiochemical separation for the analysis of plutonium, the quantity being dependent upon the mass of the whole sample.

4.3.4 Sampling of Small Mammals

Small mammals were caught using live trapping techniques and suitable animals were culled in the field, weighed and immediately placed in a cold box, before being frozen to prevent tissue degradation. Small mammals were caught using baited Longworth live traps on four occasions during the course of this study. Section 3.3 describes the trap details. On each occasion, 30 traps were placed around the margins of the marsh on both sides of the viaduct and within the strand line. Trapping sessions were somewhat determined by the predicted tide heights. Where possible, the traps were placed along or at right angles to 'runs' within the vegetation and strand line. Trapping success varied greatly with each trapping session (section 4.8). The samples were then prepared for radionuclide determination as described previously in section 3.3.

4.4 SEDIMENT RESULTS

During a tidal inundation sediment is deposited across a salt marsh thus increasing its height. As the height increases, only higher tides are able to completely submerge the marsh. Thus the accretion rate of the marsh will change over time. In addition, changes in the source of radioactivity will be reflected in the depth profile of the radionuclides; for example, the historic change in the marine discharges from BNFL, Sellafield (section 1.3.1) is reflected in the radionuclide concentrations in sediment core profiles (Murdock, 1996; Hamilton and Clarke, 1984; Oldfield *et al.*, 1993).

Spatial variation in the distribution of radionuclides across the narrow study area was anticipated, partly because the study area was at the furthest point from the river and thus unlikely to be inundated by any except the highest tides. Also, because the study area was split around the strand line, a difference in the radionuclide distribution on either side of the strand line was expected. Defining the actual foraging area for the invertebrates and small mammals is therefore important in determining both their direct exposure to, and their uptake of, the radionuclides.

Section 4.1 discusses of the behaviour of, and processes that affect, radionuclides within the marine environment, and it also describes how salt marsh sediment acts as a 'sink' for radionuclides. Within the Esk Estuary, radionuclides scavenged by sediment particles from the water body are laid down during tidal inundations. It has been shown that once bound to

the sediment and held within the marsh there is little, if any, radionuclide movement within sediment cores if the marsh is not subjected to significant physical and/or chemical disturbance (French, 1993; Mackenzie and Scott, 1993; Marcus *et al.*, 1993). This suggests that radionuclides bind strongly to sediment and therefore become unavailable for uptake and transfer to higher trophic levels. However, there is recent evidence that radionuclides, particularly the actinides, are becoming remobilised from the surface of sediment (Burton and Yarnold, 1988), and it has also been shown that 2 to 3% of the plutonium bound within the sediment can be released back to the water body (Mudge *et al.*, 1988). Burton and Yarnold (1988) went so far as to suggest that sediment within the Ravenglass Estuary is now acting as a source of actinides flowing into the Irish Sea, presumably since the discharge levels of these nuclides from Sellafield have declined in recent years.

The remobilisation of plutonium has been shown to be greatest under conditions of low salinity (<4%) (Hamilton-Taylor *et al.*, 1987) and this has implications for (a) the long term removal and redistribution of the actinides from the salt marsh sediments, and (b) the availability and uptake of radionuclides into food chains, both aquatic and terrestrial.

4.4.1 Spatial and Temporal Variation

As already mentioned, the initial sampling plan for the River Esk site utilised a simple strategy with point samples being selected at random within a defined study area. The area selected was rarely inundated fully. In fact, during the two years of the project the tides reached the boundary edge of the fields on just two occasions following strong winds and heavy rainfall. However, because the samples were collected from, and just above, the limit of regular tidal inundation, there was considerable variation in the radionuclide burden of samples taken close together.

As a consequence of using point sampling, there was some variation in the spatial and temporal activity of the sediment cores taken during April and June 1993. Figures 4.2 and 4.3 show the variation in the activity of these sediment cores. The spatial variation was mainly due to the extraction of cores from different parts of the marsh margin, particularly the position relative to distance from the river. For example, the ^{137}Cs levels for side A during April 1993 varied from 3,100 to 8,000 Bq kg^{-1} and similar levels of variation were seen for ^{238}Pu, $^{239+240}Pu$, and ^{241}Am. Only the ^{134}Cs values were reasonably stable across both sides. As a consequence, samples collected post-August 1993 were taken from transects sited on the hypothesis that there are bands of radioactivity in the sediment profile parallel to the river which depend upon the number and height of tidal inundations occurring per year. The marsh transects are shown on Figure 4.1. The data from these transects were used to generate line graphs for each radionuclide showing the change in activity over distance (Figures 4.4 to 4.5).

Figure 4.2: **^{137}Cs, $^{239+240}Pu$ and ^{241}Am activities in individual soil core samples collected during April and June 1993.**

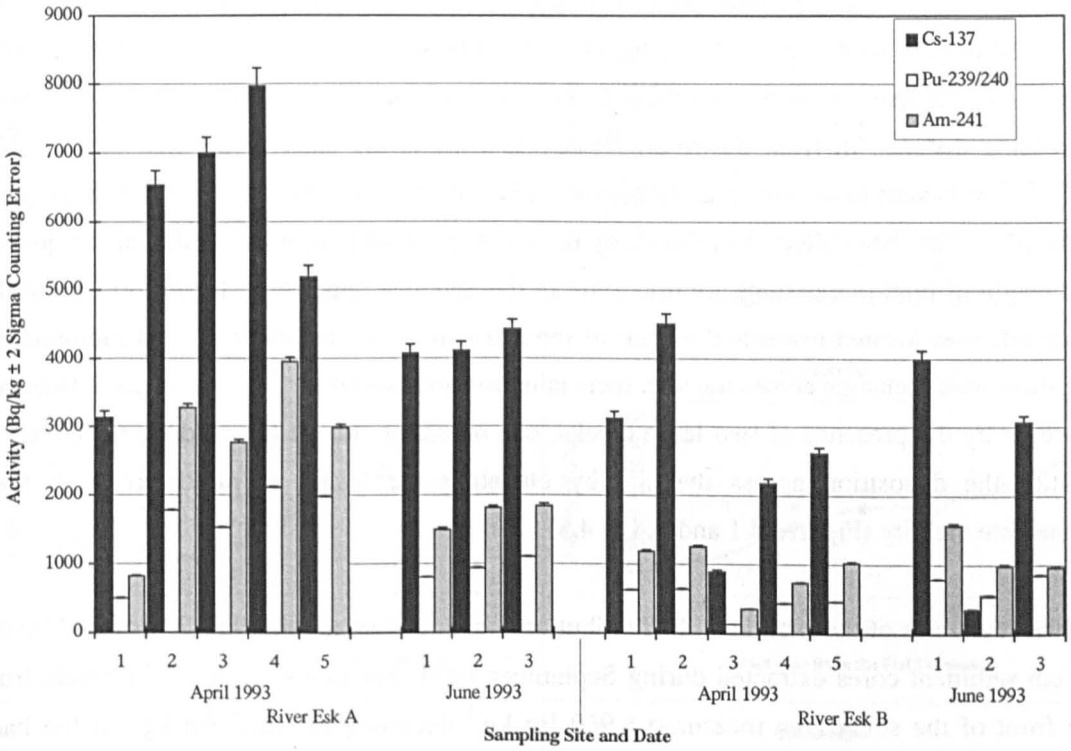
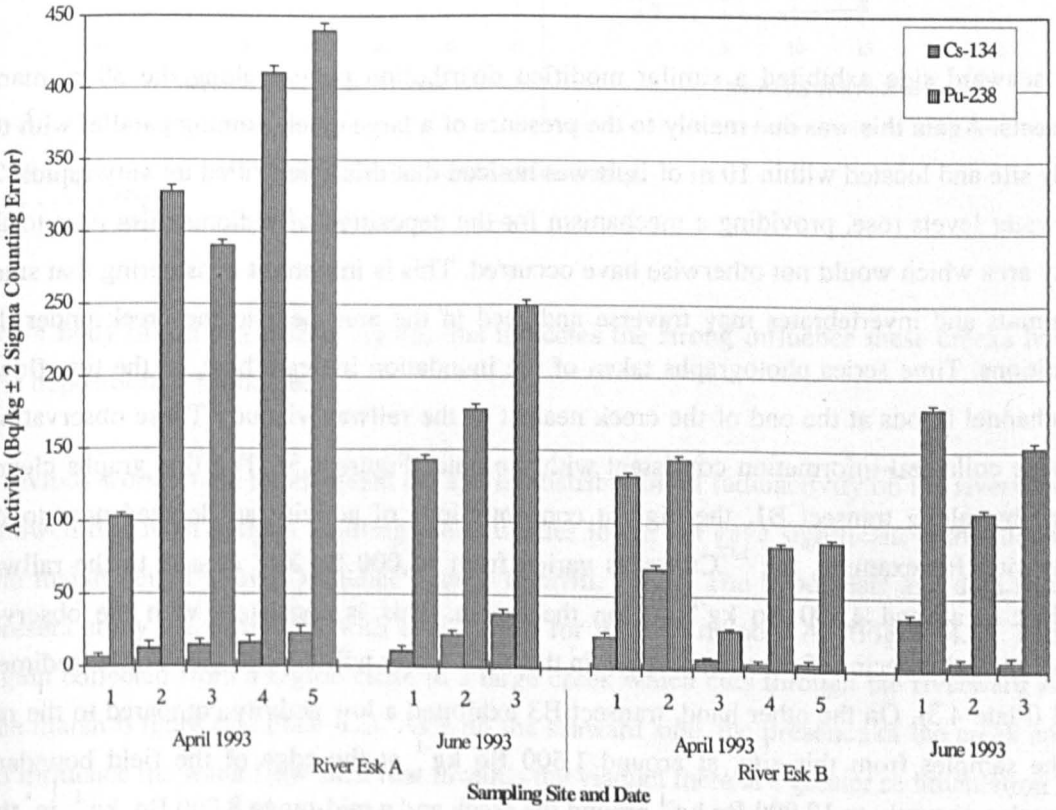


Figure 4.3: **^{134}Cs and ^{238}Pu activities in individual soil core samples collected during April and June 1993.**



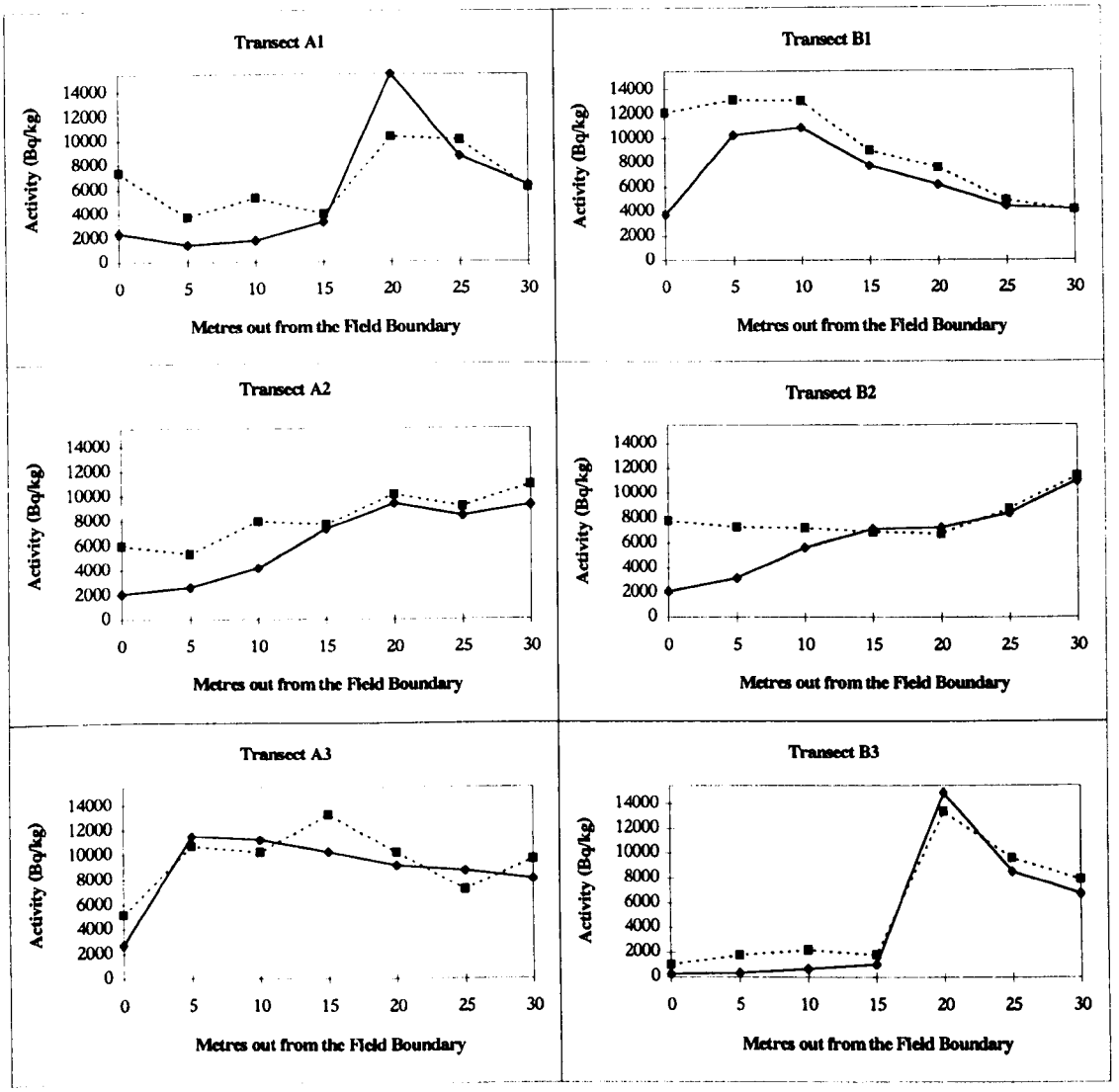
4.4.1.1 Spatial Distribution across the marsh

Although complex, the general pattern of deposition across the two marshes follows the predicted pattern with a greater amount of each radionuclide found nearer the river. Horrill (1983) showed that across the site there is an increase in activity of, for example, ^{137}Cs with increasing distance in from the river. However, towards the back of the marsh, the ^{137}Cs activity levels start to decline, mainly because this part of the marsh is subject to less frequent inundation. The data collected in this study therefore agree with the concept that the frequency and height of tidal inundations are important in radionuclide deposition. In addition, since the study site was located towards the back of the salt marsh, it was predicted that radionuclide activities would change across the site, increasing towards the river. The results are influenced however by the presence of two large creeks, one on either side of the viaduct, which act to modify the deposition across the site by elevating the levels of radioactivity in their immediate vicinity (Figures 4.1 and 4.4 to 4.5).

Further evidence of the radionuclide distribution for the riverward side (A) is provided by the 12 cm sediment cores extracted during September 1994. For example, the ^{137}Cs levels from the front of the study area measured 8,960 Bq kg^{-1} declining to 5,510 Bq kg^{-1} at the back (Figures 4.6 and 4.7; Table 4.3). Similar measurements were seen for the other radionuclides (Table 4.3). One interesting feature shown in the line graphs, Figures 4.4 and 4.5, is the similar pattern of distribution of the four reported radionuclides, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am. This lends support to the hypothesis that the deposition mechanism is tidal.

The seaward side exhibited a similar modified distribution pattern along the 30 m marsh transects. Again this was due mainly to the presence of a large creek running parallel with the study site and located within 10 m of it. It was noticed that this creek filled up very rapidly as the water levels rose, providing a mechanism for the deposition of radionuclides near to the study area which would not otherwise have occurred. This is important considering that small mammals and invertebrates may traverse and feed in the area near to the creek under dry conditions. Time series photographs taken of the inundation indicate how, as the tide flows, the channel floods at the end of the creek nearest to the railway viaduct. These observations provide collateral information consistent with the data (Figure 4.5). The line graphs clearly show that along transect B1, the highest concentrations of activity are located near to the study site; for example, the ^{137}Cs levels varied from 12,000 Bq kg^{-1} closest to the railway viaduct to around 4,000 Bq kg^{-1} out on the marsh. This is consistent with the observed flooding of this sector of the marsh, wherein the slack water will deposit much of its sediment load (Plate 4.3). On the other hand, transect B3 exhibited a low activity compared to the rest of the samples from this site, at around 1,500 Bq kg^{-1} at the edge of the field boundary, increasing abruptly to 12,000 Bq kg^{-1} around the creek and a mid-range 8,000 Bq kg^{-1} in the

Figure 4.4: **Line graphs of the marsh transects (A and B, riverward and seaward sides respectively) showing ^{137}Cs and ^{241}Am sediment results.**

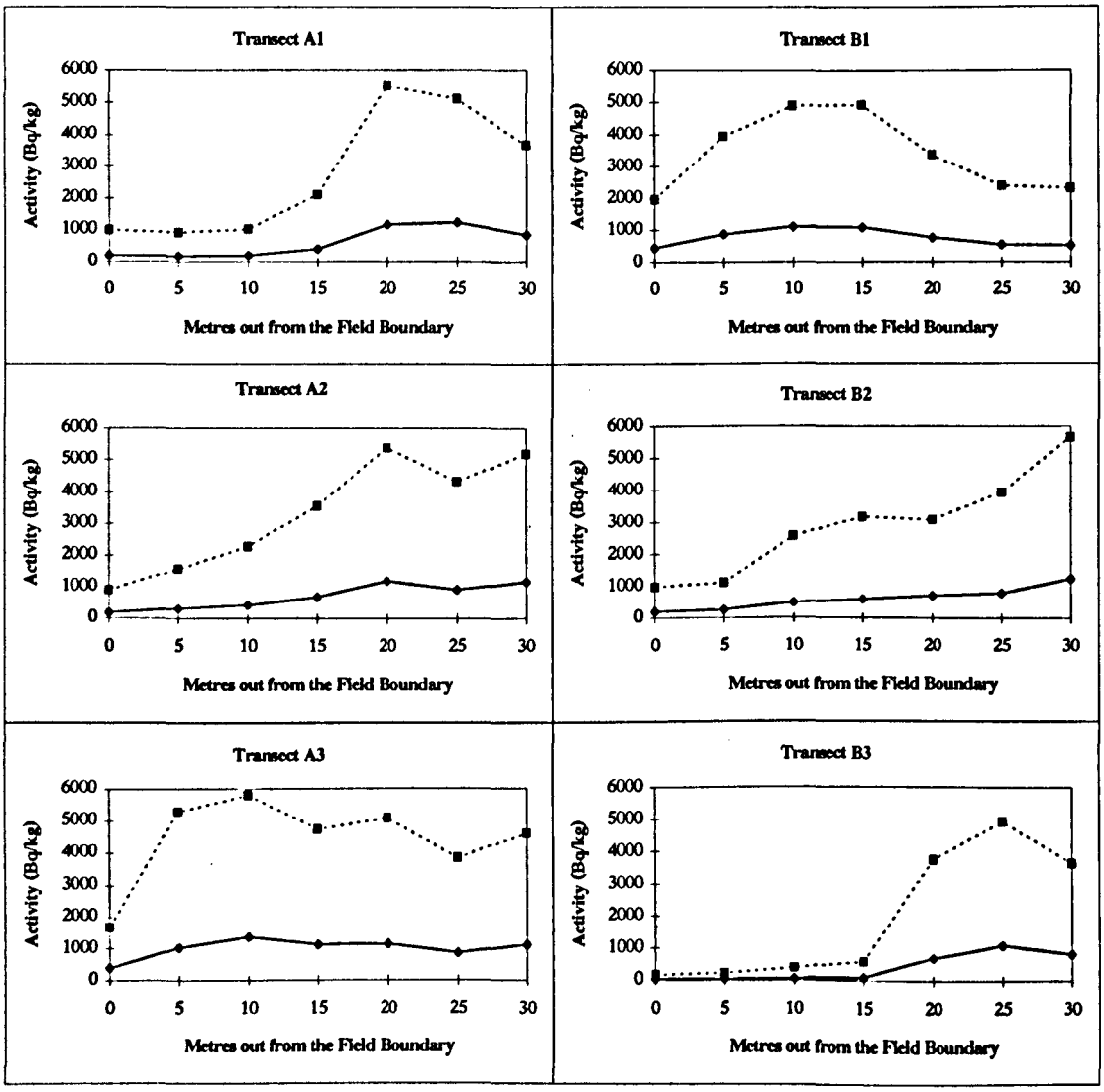


Counting errors all <3% (2 σ). Error bars omitted for clarity.
- - - - - - = ^{137}Cs.
——— = ^{241}Am

main body of the salt marsh. Again, this indicates the strong influence these creeks have on the deposition of sediment.

Previous work which investigated the spatial distribution of radioactivity on the riverward site showed that most gamma emitting radionuclides in the silt gave significant correlations with the marsh height above Ordnance Datum (Horrill, 1983). The hypothesis and data from the present study are consistent with this, except for transect number A3 (Figure 4.4). This was again collected from a region close to a large creek which cuts through the riverward side of the marsh (Figure 4.1; Plate 4.2). As with the seaward side, the presence of the creek appears to influence the water flow such that towards the viaduct there is a greater sedimentation rate,

Figure 4.5: Line graphs of the marsh transects (A and B, riverward and seaward sides respectively) showing ^{238}Pu and $^{239+240}Pu$ sediment results.



Counting errors all <3% (2 σ). Error bars omitted for clarity.
- - - - - - = $^{239+240}Pu$.
——— = ^{238}Pu.

and hence a greater amount of radioactivity being deposited. This is shown in Figure 4.4.

Figures 4.6 and 4.7 present the data from sediment cores taken in September, 1994. The charts show change in the distribution pattern for the two sides of the marsh. The riverward side follows the predicted pattern, with a greater amount of radioactivity towards the centre of the marsh. This agrees with the transect data discussed above.

Additional sediment samples were collected in September, 1994 from the centre of the salt marsh on the riverward side to confirm that the activity levels still rise towards the centre compared to the margins. The measured activities ranged from 10,800 to 12,300 Bq kg^{-1} for

Figure 4.6: **^{137}Cs, $^{239+240}$Pu and ^{241}Am activities in soil core samples collected during September 1994.**

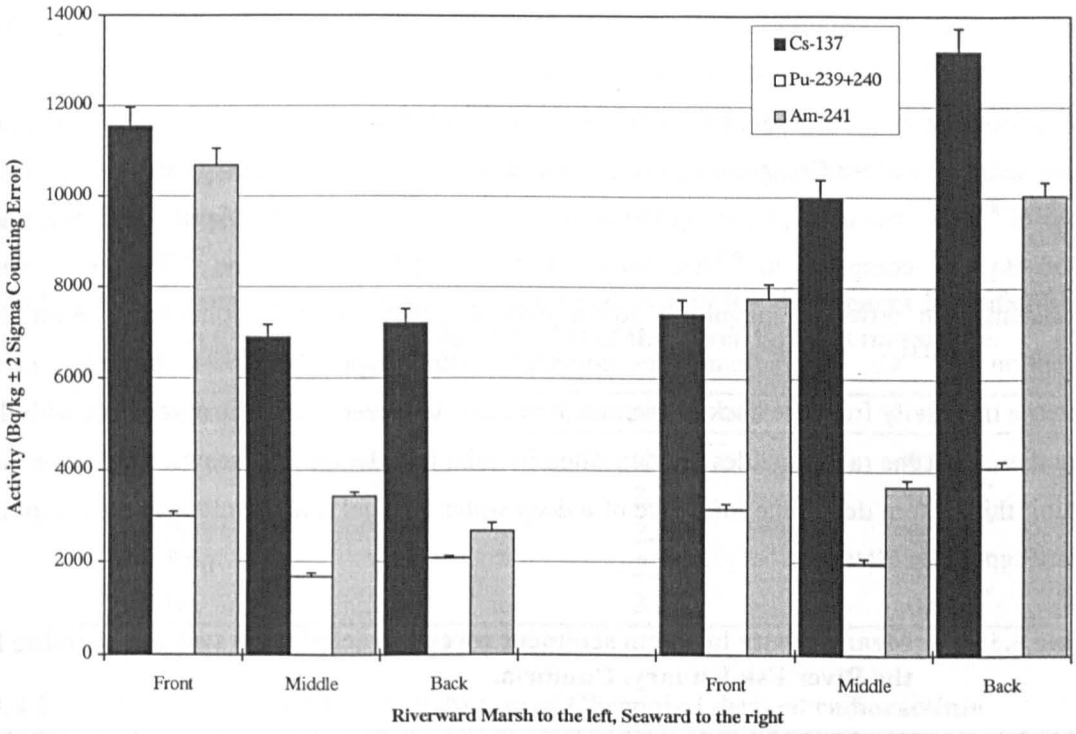
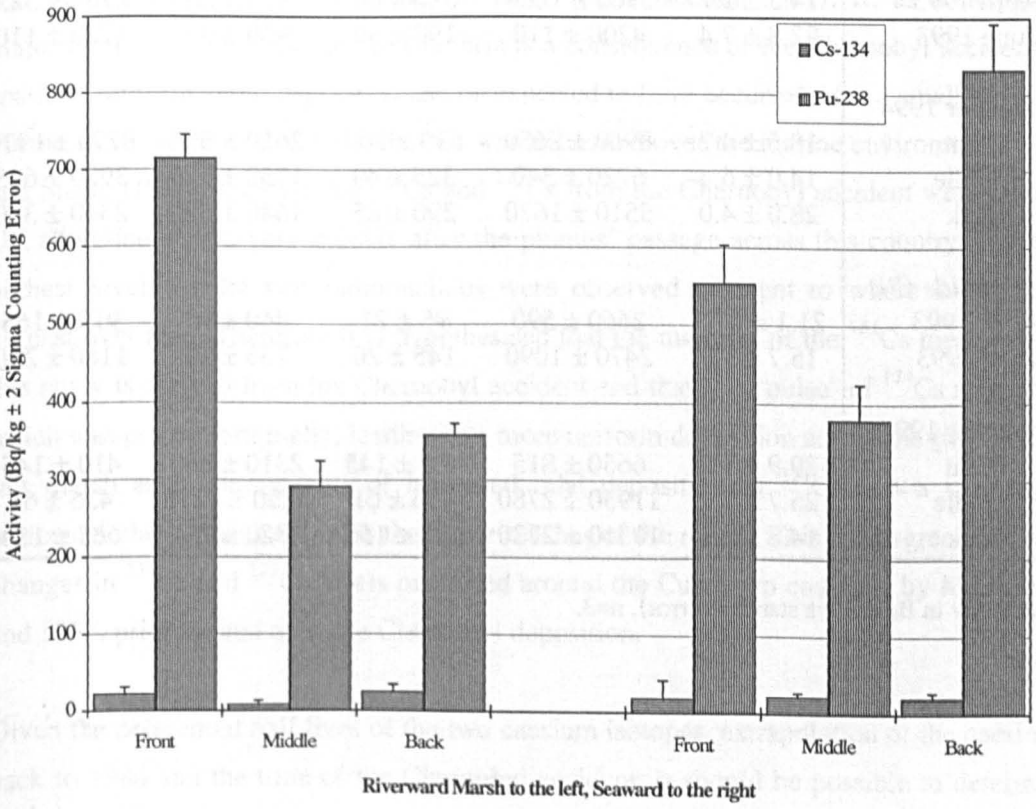


Figure 4.7: **^{134}Cs and ^{238}Pu activities in soil core samples collected during September 1994.**



^{137}Cs; 12,500 to 15,000 Bq kg^{-1} for ^{241}Am; 1,100 to 1,450 Bq kg^{-1} for ^{238}Pu; and 5,700 to 7,000 Bq kg^{-1} for $^{239+240}Pu$. These data agree with the hypothesis that the radionuclide deposition across the site continues to increase as the river is approached and compare favourably with the data presented in Table 4.3.

As Table 4.3 shows, the level of activity for ^{137}Cs, ^{241}Am and, to a lesser extent, $^{239+240}Pu$, are all of a similar order of magnitude; but in this case the ^{137}Cs levels are approximately twice those of ^{241}Am. For example, along the riverward side ^{137}Cs levels for April, 1993 averaged 5,960 Bq kg^{-1} compared to ^{241}Am levels of 2,760 Bq kg^{-1}. ^{134}Cs and ^{238}Pu levels were consistently an order of magnitude lower than the other three radionuclides. With the exception of ^{134}Cs, the radionuclides collected during September 1994 showed a clear increase in activity from the back of the marsh towards the river. This is in agreement with the hypothesis that the radionuclides are deposited by tidal inundation. The seaward side does not exhibit this pattern due to the influence of a deep water channel running close to the sampling area (Figure 4.1; Plate 4.3).

Table 4.3: Mean activity in 12 cm sediment cores extracted from two salt marshes in the River Esk Estuary, Cumbria.

Sampling Date and Location	*^{134}Cs*	*^{137}Cs*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*
Riverward Side (A):					
April 1993	14.3 ± 2.7	5960 ± 1880	315 ± 60	1580 ± 285	2760 ± 525
June 1993	22.8 ± 7.4	4200 ± 110	190 ± 30	950 ± 85	1720 ± 110
September 1994					
Front	18.5 ± 4.7	8960 ± 2650	610 ± 100	2610 ± 555	8770 ± 1970
Middle	14.0 ± 6.4	6720 ± 340	325 ± 40	1750 ± 125	3930 ± 635
Back	28.0 ± 4.0	5510 ± 1670	290 ± 85	1640 ± 620	2360 ± 340
Seaward Side (B):					
April 1993	21.1 ± 12.2	2660 ± 590	95 ± 21	460 ± 90	910 ± 165
June 1993	15.7 ± 9.9	2470 ± 1090	145 ± 20	735 ± 90	1180 ± 200
September 1994					
Front	20.9 ± 3.2	6650 ± 815	410 ± 145	2310 ± 880	410 ± 145
Middle	25.7 ± 7.0	11950 ± 2780	436 ± 61	3250 ± 1290	436 ± 61
Back	14.8 ± 4.1	10510 ± 2730	685 ± 150	3320 ± 770	685 ± 150

Mean Activity in Bq kg^{-1} (± standard error), n=3.

4.4.1.2 Comparison of radionuclide levels on the two sides of the marsh

Figures 4.2, 4.3, 4.6 and 4.7 show some evidence of a difference in the activity levels of each of the radionuclides between the two sides of the viaduct. The mean data presented in Table 4.3 provide further comparisons of the radionuclide data for the two sides. The results of a series of Student *t* tests show that the activity levels of four of the five radionuclides are significantly different ($p<0.01$) across the two sides of the marsh. Table 4.4 summarises the results of the *t* tests.

Table 4.4: Summary of Student *t* test output examining differences in radionuclide activities between sides A and B of the River Esk salt marsh site.

Radionuclide	*t statistic*	*t critical (two tail)*	*P value*
^{134}Cs	-0.18	2.26	0.858
^{137}Cs	3.47	2.26	0.004**
^{238}Pu	3.30	2.26	0.009**
$^{239+240}Pu$	3.50	2.26	0.007**
^{241}Am	3.48	2.26	0.007**

4.4.1.3 ^{134}Cs Activity and the contribution of Chernobyl derived radiocaesium

The anomalous result is that of ^{134}Cs and it is suggested that this is a function of the deposition of ^{134}Cs when compared to the other four radionuclides in much the same way as was described for the soil activities in Lady Wood (section 3.4.1). If, as anticipated, the majority of the ^{134}Cs deposited in Cumbria is a consequence of the Chernobyl accident then a spatially uniform aerial deposition can be expected to have occurred over a small area of land. Furthermore, additional ^{134}Cs and ^{137}Cs was deposited over the marine environment. Mitchell and Steele (1988) reported that ^{134}Cs and ^{137}Cs from the Chernobyl accident were detected in UK shoreline waters very quickly after the plumes' passage across this country, and that the highest levels of the two radionuclides were observed adjacent to where deposition was highest over land. Therefore it is hypothesised that the majority of the ^{134}Cs measured during this study is derived from the Chernobyl accident and that this 'pulse' of ^{134}Cs swamped that which was present originally, leading to a more uniform deposition across the two sides of the salt marsh as a consequence of increased tidal deposition of ^{134}Cs and a spatially more uniform aerial deposition across the upper reaches of the marsh. This is in agreement with the changes in ^{134}Cs and ^{137}Cs levels measured around the Cumbrian coastline by MAFF in 1985 and 1987, prior to- and after the Chernobyl deposition.

Given the deferential half lives of the two caesium isotopes, extrapolation of the caesium data back to 1986 and the time of the Chernobyl accident, it should be possible to determine the ^{134}Cs and ^{137}Cs inventory attributable to Chernobyl in the salt marsh sediments. Table 4.5

presents data extrapolated back to 1986 and compares it to the present activities for ^{134}Cs and ^{137}Cs. It is evident from the data that there is a significant change in the ^{137}Cs:^{134}Cs ratio with an average value for the two sides being 34 and 30 for A and B respectively. This compares to the present average ^{137}Cs:^{134}Cs ratio recorded during this study for the sediment samples of 307 and 270. There is an order of magnitude difference between the two sets of data but more importantly the ratio values of 30 to 34 are significantly closer to the measured ratio obtained in 1986 for sites around BNFL, Sellafield. It should be noted that the Chernobyl-derived caesium was deposited on to the sediments and, using the coring technique employed in this study, this deposition will have been mixed with those deposits from previous years. This will raise the ^{137}Cs activity levels and therefore elevate the ratio obtained.

An upper estimate of the ^{137}Cs component derived from the Chernobyl accident can be obtained by multiplying the corrected ^{134}Cs values by 1.6 (Fulker, 1987; section 3.4.1). From these calculations, it can be estimated that the contribution of the Chernobyl accident to the total ^{137}Cs inventory (to the depth of 15 cm) in the sediments is 5% and 11% for sides A and B respectively. Again, this reflects differences in the way the tidal inundation deposits sediment across the two sides of the marsh as the water flow is affected by the presence of the viaduct. On side A, the higher radionuclide activities suggest that there has been a greater sedimentation rate in the past (and may still be).

The similarity in activity levels of ^{137}Cs, $^{239+240}Pu$ and ^{241}Am agrees with the known behaviour of the actinides and ^{137}Cs in a water body (section 4.1). Liquid discharge data for Sellafield (section 1.3.1) show that ^{137}Cs levels are between 15 and 30 times greater than for total plutonium alpha and ^{241}Am respectively (BNFL, 1992). In previous years these levels

Table 4.5: ^{134}Cs and ^{137}Cs Activities (Bq kg^{-1} ± 2σ counting error) decay corrected to the Chernobyl accident deposition over the UK on 2nd May 1986.

Site	*April 1993 Data*			*Decay Corrected Data*		
	^{137}Cs	^{134}Cs	^{137}Cs:^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs:^{134}Cs
	Riverward (A)					
Core 1	3,130 ± 100	6 ± 0.4	512	3,670 ± 120	64 ± 4.2	58
Core 2	6,530 ± 210	12 ± 0.7	533	7,670 ± 250	128 ± 7.6	60
Core 3	7,000 ± 230	14 ± 0.7	486	8,220 ± 270	150 ± 7.8	55
Core 4	7,970 ± 260	16 ± 0.8	494	9,360 ± 310	168 ± 8.5	56
Core 5	5,180 ± 170	23 ± 0.6	229	6,080 ± 200	236 ± 5.8	26
	Seaward (B)					
Core 1	31,310 ± 100	11 ± 0.9	142	3,670 ± 120	230 ± 5.6	16
Core 2	4,510 ± 500	68 ± 2.3	66	5,290 ± 170	711 ± 23.7	7
Core 3	890 ± 30	7 ± 0.6	127	1,050 ± 34	73 ± 6.5	14
Core 4	2,180 ± 70	4 ± 0.3	531	550 ± 84	43 ± 3.4	60
Core 5	2,620 ± 90	4 ± 0.4	595	3,070 ± 100	46 ± 3.8	67

Data used taken from April 1993 samples.

have been much greater still. From the discharge data, high levels of ^{137}Cs might be expected in the environment. However, it is well known that ^{137}Cs is conservative in a water body, remaining in solution and dispersing with the flow of the currents (Assinder *et al.* 1985; section 4.1). In contrast, the actinides are readily scavenged from solution by adsorption on to particulate material (McKay and Pattenden, 1993). The particle grain size is also important, partly because it affects sedimentation rate and also because it influences adsorption of radionuclides to sediment particles (Hamilton-Taylor *et al.*, 1993); hence deposition of the actinides by tidal inundation of the salt marsh leads to an increased level of activity compared to ^{137}Cs. The effects of the particle size of the sediments from the Esk Estuary have been examined in detail elsewhere (Kelly and Emptage, 1991) and therefore were not specifically investigated in this study.

4.4.1.4 Deposition values, Bq m^{-2}

Table 4.6 presents data calculated for the deposition (Bq m^{-2} to 3 significant figures) for the radionuclides: ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am. The data clearly show an elevated deposition of the same radionuclides when compared to grassland sites within the locality (Jones *et al.*, 1996). Given the different deposition mechanisms this is not surprising. Moreover, the data provide evidence of the conservative behaviour of ^{137}Cs in water bodies when compared to the actinides because the ^{137}Cs activity on the grassland site is between 15 and 200 times higher than the actinides, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am. This contrasts sharply

Table 4.6: Comparison of deposition values (Bq m^{-2}) from samples collected in September 1994 with previous studies.

Location	*Sample Date*	*^{134}Cs*	*^{137}Cs*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*	*Reference*
Riverward (A)							
Front	1994	490	269,000	16,800	67,800	250,000	This study
Middle	1994	205	182,000	7,700	44,700	90,500	"
Back	1994	240	90,300	4,330	24,700	33,900	"
Seaward (B)							
Front	1994	410	170,000	12,700	73,000	180,000	This study
Middle	1994	110	55,300	2,250	11,700	20,400	"
Back	1994	75	91,600	5,870	28,500	69,600	"
Seascale Hall*	1987		14,250	65	939	386	Jones *et al.*, 1996
Ravenglass**			5,370 to 171,000		520 to 50,400	539 to 47,000	Curtis *et al.*, 1991
Solway Firth	1992		35,200				McDonald *et al.*, 1992

* Radionuclide deposition to a grassland site (aerial deposition).

** Collected annually between 1979 and 1985; the upper and lower values obtained over the period are reported.

with the values obtained from the Esk sediment where the ^{137}Cs values range from 13.4 to 24.6, 2.3 to 4.0 and 0.9 to 2.7 times higher than ^{238}Pu, $^{239+240}Pu$ and ^{241}Am respectively.

The reported levels of activity in this study compare favourably with measurements made in previous work around the North West coastline, particularly for Ravenglass (Curtis *et al.*, 1991). The exception is the high ^{241}Am values at the front edge of the study area on both sides of the River Esk salt marsh. However, Horrill (1983), sampling on the riverward side, measured ^{241}Am values with a mean silt value of 5,650 Bq kg^{-1} which is between the extremes of measured values for ^{241}Am of between 1,720 and 8,770 Bq kg^{-1} on the riverward side (Table 4.3).

4.4.1.5 Isotopic and Nuclide ratios

Table 4.7 presents data on the mean isotopic and nuclide ratios across the site for samples collected in September, 1994. The values were calculated for each transect across the front, middle (strand line) and back of the study areas. The results are variable and there appears to be no pattern or consistency to the data. Given the similar pattern of deposition for each radionuclide in the 30 m marsh transects, and the decline in activity for each radionuclide from the front to the back of the study area in the short core data, a constant isotopic or nuclide ratio was expected. The ^{137}Cs:^{134}Cs ratio is also inconsistent with the same ratio in the liquid waste discharged through the Sellafield pipeline. For 1992, the ^{137}Cs:^{134}Cs ratio in the annual discharge was 18.4. The ratios measured in the salt marsh sediments are considerably greater than this value. There is in fact a 25 fold difference. This is due to the difference in the half lives of ^{134}Cs and ^{137}Cs and the fact that the caesium deposit in the sediment, being aged, includes a higher proportion of ^{137}Cs present.

Table 4.7: Mean isotopic and nuclide ratios for River Esk sediment samples from September 1994.

Sample Site	*Isotopic Ratios*		*Nuclide Ratios*	
	^{137}Cs:^{134}Cs	^{238}Pu:$^{239+240}Pu$	^{137}Cs:$^{239+240}Pu$	^{137}Cs:^{241}Am
Riverward (A)				
Front	547 ± 122	0.25 ± 0.06	3.96 ± 0.73	1.08 ± 0.03
Middle	926 ± 215	0.17 ± 0.01	4.09 ± 0.23	2.06 ± 0.38
Back	299 ± 84^{+}	0.18 ± 0.03	3.99 ± 2.14	2.70 ± 0.21
Seaward (B)				
Front	421 ± 139	0.18 ± 0.02	2.35 ± 0.16	0.97 ± 0.13
Middle	491 ± 88^{+}	0.19 ± 0.00	5.29 ± 3.19	2.69 ± 0.21
Back	667 ± 144^{+}	0.20 ± 0.02	3.23 ± 0.21	1.32 ± 0.00

Values reported to 3 significant figures as average (± standard deviation), n=2.
$^{+}$ Values calculated using < limit of detection values and consequently could be an overestimation of the true activity.

4.4.1.6 Behaviour of radionuclides

It was noted by Horrill (1983) that ^{134}Cs levels increased progressively up the marsh from the river. This was related to the tidal inundation cycle. During each cycle, fresh ^{134}Cs is deposited up to the height of the inundation. Since total inundation across the study area is rare, there is little replacement of the ^{134}Cs within the study area and the levels are declining as radioactive decay occurs. The longer half-life of ^{137}Cs gives rise to a diferential decline in the activity levels of the caesium isotopes and hence the ^{137}Cs:^{134}Cs ratio is much greater than that of the liquid discharges from Sellafield.

The nuclide ratios, ^{137}Cs:$^{239+240}Pu$ and ^{137}Cs:^{241}Am, reflect the increase in the activity of actinides across the River Esk salt marshes. The seaward side of the marsh shows less consistent isotopic and nuclide ratios compared to the riverward side. One possible reason for the small differences in the ratios might be the way that ^{137}Cs and the actinides behave in the presence of silt or clay sediment within the water body. It is well known that the actinides are readily adsorbed on to sediment particles with distribution coefficients (kd) for Pu in the order of $1x10^5$ and Am $2x10^6$, unlike ^{137}Cs which is closer to 3,000 (IAEA, 1985). Grain size of the sediment particles and the presence of organic matter will also affect the radionuclide adsorption (section 4.1), and topographical differences between the two sides of the marsh will then influence the sedimentation rate for different sized particles, affecting radionuclide deposition.

4.4.1.7 ANOVA results

Table 4.8 presents the results from a series of Analysis of Variance tests that examined the spatial variation across the study site using the data collected from September, 1994. The test examined the data for differences between the front, middle and back transects on each side of the railway viaduct. Because of changes in the sampling strategy, temporal variation cannot accurately be assessed using the present data set. It was expected that the salt marsh sediments would change in activity because as salt marshes develop and age, new sediment containing contemporary radioactivity will be laid down across the surface (Hamilton and Clarke, 1984; McCaffrey and Thompson, 1980; Murdock, pers. comm.). However, in this study, because samples were collected around the margins of the marsh in the areas thought to be important for small mammal and invertebrate activity, little 'new' sediment was laid down during the study period since deposition here will only have occurred on the two occasions when the marsh was completely submerged.

This means that the sediment activity in the cores extracted at different times is unlikely to change. The exception will be material undergoing radioactive decay and new material being

Table 4.8: **Summary of results from One-way Analysis of Variance Tests for spatial variation in whole sediment cores on ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities.**

	Riverward Side			*Seaward Side*		
Nuclide	*F-ratio*	*P*	*5% LSD*	*F-ratio*	*P*	*5% LSD*
^{134}Cs	4.04	0.141		0.14	0.877	
^{137}Cs	13.09	0.033*	2850.0	1.01	0.462	
^{238}Pu	13.65	0.031*	243.6	31.46	0.010**	161.2
$^{239+240}Pu$	1.29	0.395		17.52	0.022*	1003.6
^{241}Am	49.33	0.005**	2494.0	9.95	0.047*	4071.9

deposited across the site via aerial deposition and sea to land transfer, both of which are of minor importance compared to the tidal effects and therefore the historic deposition contained within the core. It is suggested that the variation in activity for the sediments as recorded are due to changes in the physical coring locations between April/June 1993 and September 1994. Because of this, and the changes in the way samples were collected between April 1993 and the next two occasions, an additional set of samples is required to utilise the Analysis of Variance (ANOVA) test for exploring temporal variation. For those spatial ANOVA results which showed significant differences, a 5% Least Significant Difference (LSD) figure was calculated using Bonferroni's method and the result is also reported in Table 4.8.

Further examination of the ANOVA results reveals that there are significant differences in the activity along the three transects running parallel to the strand line, but only for some radionuclides. Analysis of the 5% LSD values shows that the deposition pattern is different on the two sides of the marsh. For example, the riverward side conforms to the expected distribution pattern that the concentration of each radionuclide declines towards the back of the marsh. The ANOVA data confirm this for ^{137}Cs, ^{238}Pu and ^{241}Am, with the front transect results being significantly different from both the middle (strand line) and back ($p<0.01$ for ^{241}Am; $p<0.05$ for ^{137}Cs and ^{238}Pu). The seaward side again exhibits the effects of the large creek. In this case it is the middle (strand line) transect which is significantly different from both the back and front transects ($p<0.01$ for ^{238}Pu; $p<0.05$ for $^{239+240}Pu$ and ^{241}Am).

The sediment cores provide information on the radionuclide concentrations incorporated into the sediment. Information concerning the spatial distribution of ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am across the site indicates that each side of the marsh is subject to a distinct pattern of deposition caused by differences in the way the tide inundates the two sides of the marsh. To examine the historic deposition to the marsh and to determine the radionuclide concentration in the rooting zone of the vegetation, additional soil cores were collected and sectioned into 4 cm slices.

4.4.2 Radionuclide Distribution within Soil Core Profiles

Data from the 12 cm sectioned cores are presented in Figures 4.8 and 4.9. For each radionuclide the levels of historic deposition across the site are of a similar order of magnitude on both sides of the marsh. This provides confirmation that the two sides have received a similar radionuclide input in the past, although the spatial distribution of radionuclides is different across the two sides.

Commonly, sediment core samples collected from salt marshes on the North West coast have been analysed to produce vertical radionuclide profiles. These profiles have been shown to reflect changes in the liquid discharge history of BNFL, Sellafield (Hamilton and Clarke, 1984; Murdock, 1996) because of the stratification obtained from the mechanism of salt marsh sedimentation. However, it is evident from these short core samples that the 4 cm resolution is, not surprisingly, too crude to accurately model the Sellafield discharges. Further evidence of this, is provided by the 50 cm cores taken in November 1993 (Figures 4.10 and 4.11).

The 50 cm core data show a very rapid decline in activity levels for both sides of the marsh between the 20 and 30 cm depth. This reflects the fact that the cores were taken 10 to 15 m below the strand line, in an area with low sedimentation rate. This suggests that the active discharges from Sellafield have been laid down in the top 20 cm of sediment. Further work involving detailed core analysis with a much finer resolution would provide sufficient data to successfully determine the sedimentation rates in this region of the marsh over the last 40 years. With a low sedimentation rate, each slice in the core contains sediment laid down over a number of years. This explains the presence of sub-surface maxima for ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am between 5 and 10 cm down the sediment profile in both the 50 cm core and the 12 cm cores taken from the seaward side and from the front transect on the riverward side of the marsh. It is hypothesised that these sub-surface maxima are representative of the highest radioactive liquid discharges released from BNFL, Sellafield in the early 1970s (section 1.3.1). ^{134}Cs is the only nuclide not to show this clearly and it is postulated that this is related to the short half-life of ^{134}Cs compared to the other radionuclides.

The distribution pattern for radionuclides down the 50 cm cores taken on the seaward side is different with no sub-surface maxima and, also, the bulk of the activity (>97%) present in the top 15 cm. This contrasts with the riverward side where >99% of the radionuclides are in the top 25 cm. A probable explanation of these differences is that the sedimentation rate on the seaward side is much lower. A low sedimentation rate would mean an inferior resolution using a 5 cm core slice which may obscure the presence of any sub-surface maxima.

Figure 4.8: **^{137}Cs, $^{239+240}Pu$ and ^{241}Am activities in River Esk sectioned sediment cores (12 cm). Data from August 1993.**

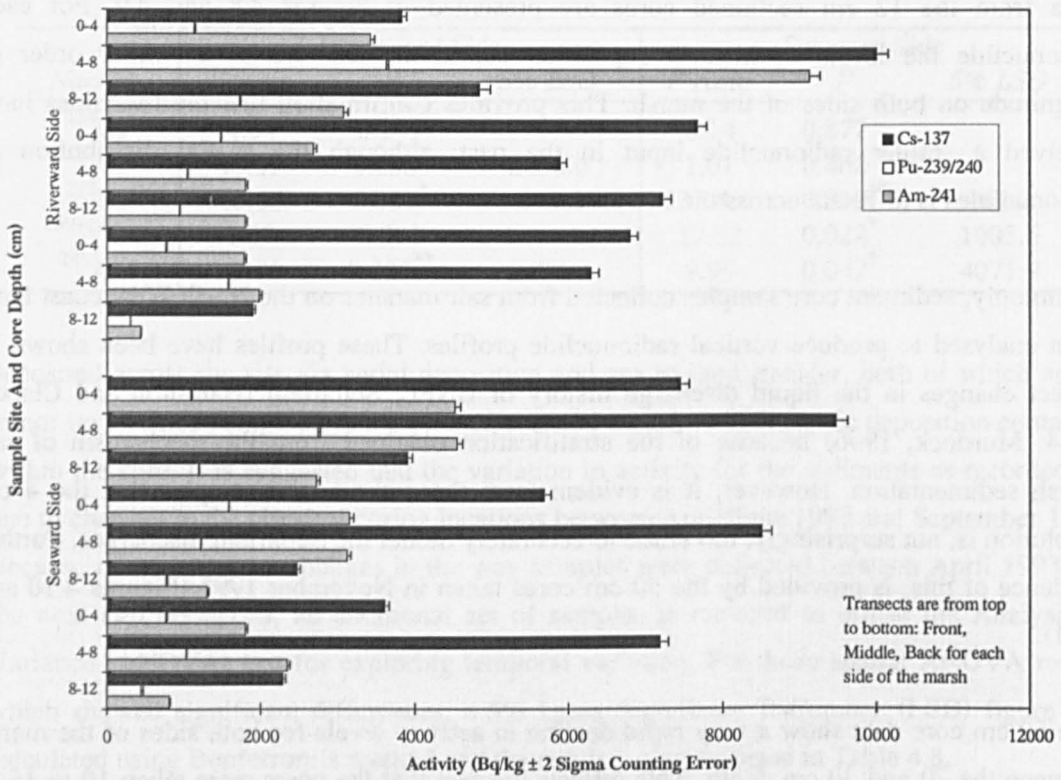


Figure 4.9: **^{134}Cs and ^{238}Pu activities in River Esk sectioned sediment cores (12 cm). Data from August 1993.**

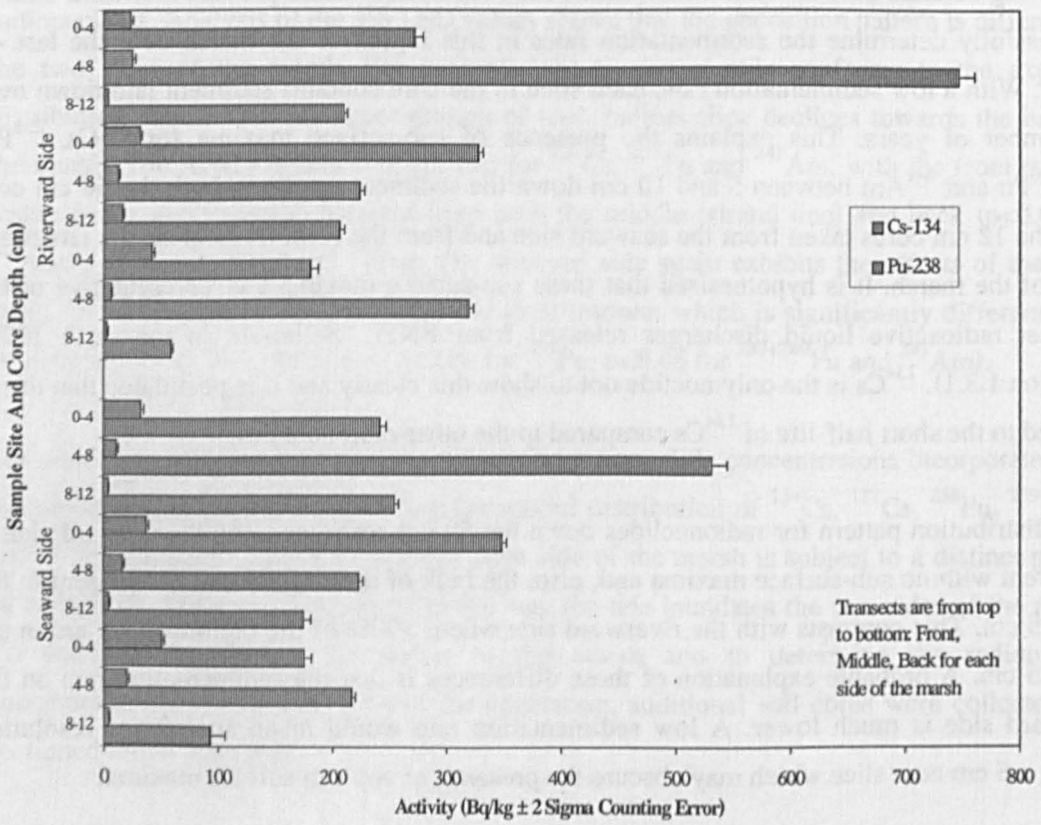


Figure 4.10: ^{137}Cs, $^{239+240}Pu$ and ^{241}Am activity in 50 cm sediment cores extracted during November 1993.

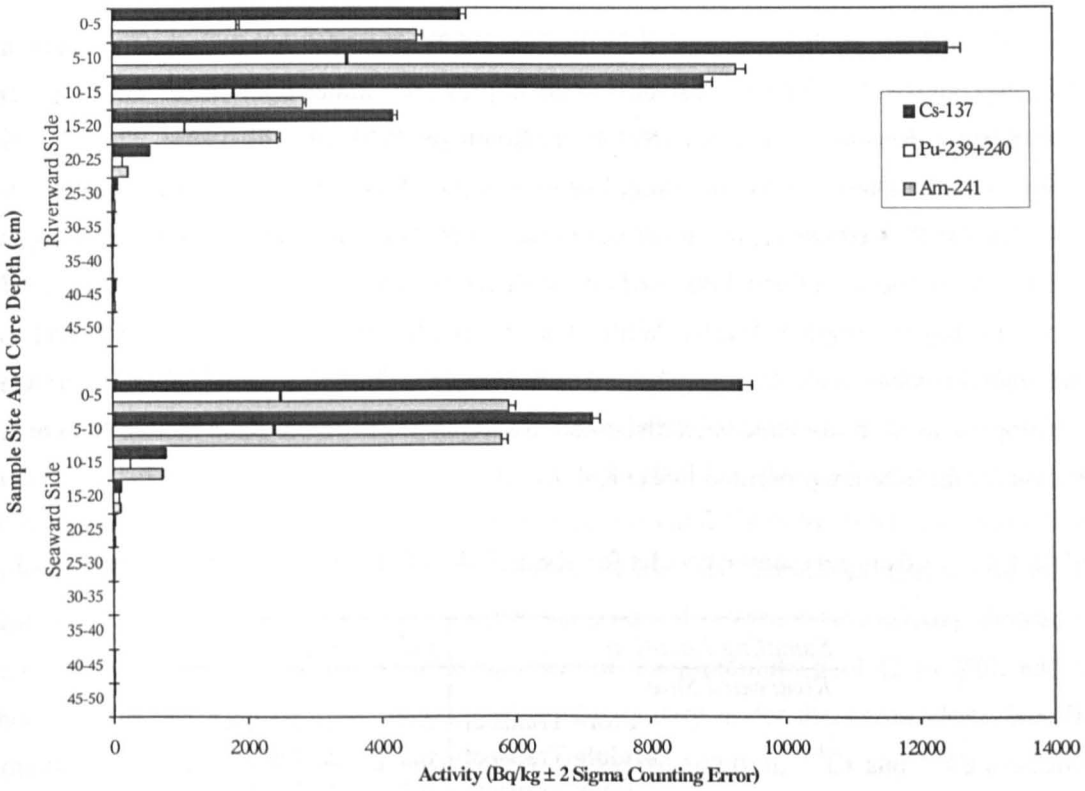
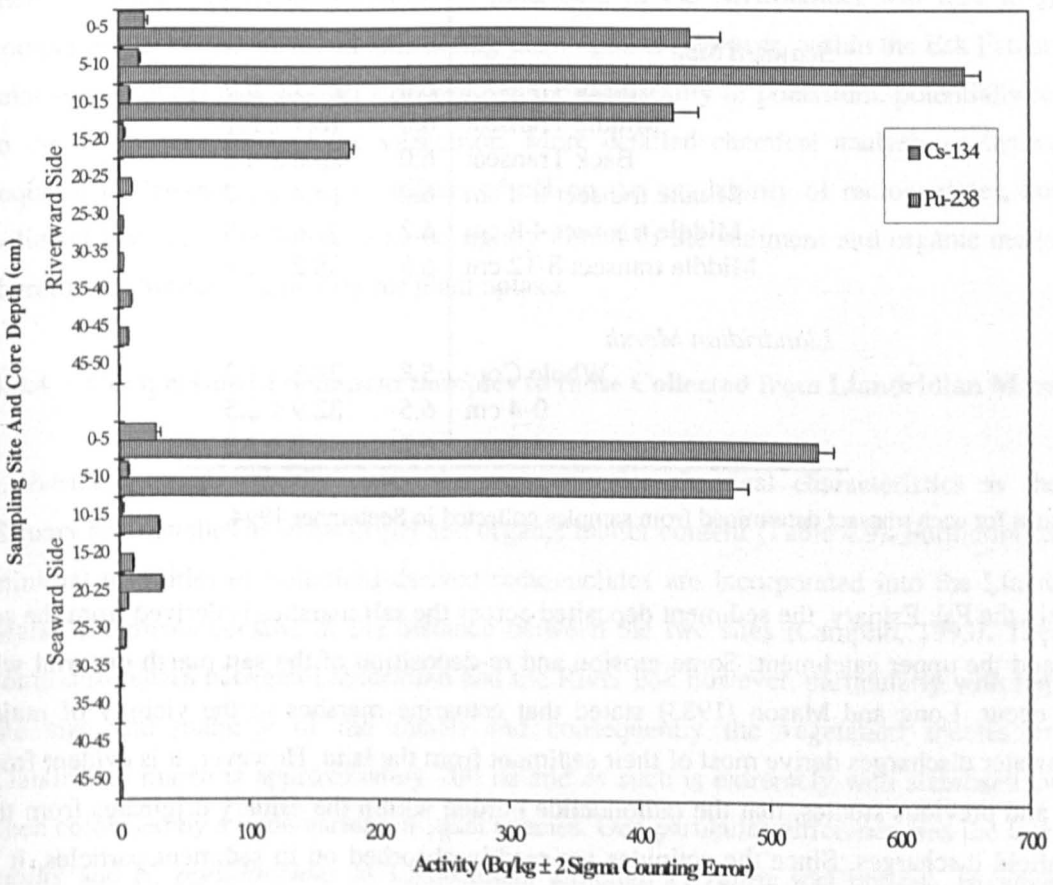


Figure 4.11: ^{134}Cs and ^{238}Pu activity in 50 cm sediment cores extracted during November 1993.



4.4.3 Sediment Characterisation

The radionuclide activities measured in both the 50 cm and 12 cm sediment cores are reasonably consistent with measurements made in previous studies both on the salt marsh site and within the Ravenglass Estuary (Kelly and Emptage, 1991; Horrill, 1983). This provides evidence of a long term, and continuing, low level input of radionuclides into the salt marshes of the Esk Estuary. However, for the present study, it is also important to assess the biological availability of these radionuclides within the environment and consequently the transfer factors to higher trophic levels. With this in mind, organic matter content and pH measurements were made on a small number of samples. The influences of these parameters on biological availability have been discussed in section 1.4.1. The organic matter content and pH data for this site are presented in Table 4.9.

Table 4.9: Soil parameter results for River Esk sediments (± standard error), n=3.

Sampling Location	*pH*	*% LOI**
Riverward Side		
Front Transect	6.0	23.3 ± 0.4
Middle Transect	6.3	28.7 ± 3.3
Back Transect	6.1	48.1 ± 13.7
Middle transect 0-4 cm	6.3	34.8 ± 4.9
Middle transect 4-8 cm	5.5	29.8 ± 1.7
Middle transect 8-12 cm	6.0	21.6 ± 1.6
Seaward Side		
Front Transect	6.0	27.1 ± 5.9
Middle Transect	6.0	18.7 ± 2.1
Back Transect	6.0	25.8 ± 0.8
Middle transect 0-4 cm	6.0	14.4 ± 1.7
Middle transect 4-8 cm	6.2	37.6 ± 6.6
Middle transect 8-12 cm	6.5	38.2 ± 5.2
Llandridian Marsh		
Whole Core	5.8	29.5 ± 1.3
0-4 cm	6.5	32.9 ± 2.5
4-8 cm	6.7	46.2 ± 3.8

* Results for each transect determined from samples collected in September 1994.

Within the Esk Estuary, the sediment deposited across the salt marshes is derived from the sea bed and the upper catchment. Some erosion and re-deposition of the salt marsh material will also occur. Long and Mason (1983) stated that estuarine marshes in the vicinity of major freshwater discharges derive most of their sediment from the land. However, it is evident from this, and previous studies, that the radionuclide burden within the estuary originates from the Sellafield discharges. Since the actinides are readily absorbed on to sediment particles, it is

suggested that the bulk of the sediment within the Esk salt marshes derives from the sea. Kelly *et al.* (1991) reported a similar interpretation. This implies that the bulk of the organic matter present within the marsh is derived also from the sea. Moreover, it has been reported that the organic matter content of a marsh will increase with elevation (Long and Mason, 1983) mainly because of the increase in vegetation cover as the marsh ages. Organic matter has been shown to be important in the adsorption of radionuclides within soil or sediment profiles and, given the measured organic matter content, it is expected that much of the radionuclide load within the salt marshes will be tightly bound.

The pH results show consistency between the transects and the two sides of the marsh. Around this level of pH 6.0 most metal ions, particularly the radionuclides of interest in this study, will be insoluble and remain tightly adsorbed to the sediment particles. The remobilisation of small quantities of plutonium (Burton and Yarnold, 1988; Hamilton-Taylor *et al.* 1987) only accounts for a small proportion of the total plutonium held within the Esk Estuary. In fact measurements of the sediment and tidal flows within the estuary showed that remobilisation accounted for a small proportion of the plutonium pool (2 to 3%), and that there was in fact a net gain of plutonium into the estuary under the normal low river flow conditions seen within this estuary (Kelly *et al.* 1991). In addition, ^{134}Cs and ^{137}Cs availability are affected by the high levels of potassium and sodium in salt marsh soils compared to typical grassland (Long and Mason, 1983). Since potassium and caesium are analogous in their behaviour, high concentrations of potassium in the environment will lead to greater competition between the metal ions during plant uptake. However, within the Esk Estuary, the relatively high pH may lead to a decrease in the availability of potassium, potentially leading to uptake of caesium into the vegetation. More detailed chemical analysis of the soils is required to determine the exact effects of pH on the availability of radionuclides, but it is believed that they will, in general, be tightly bound to the sediment and organic matter and therefore of limited availability for plant uptake.

4.4.4 Comparison of Sediment Samples to those Collected from Llandridian Marsh

Llandridian marsh exhibits much the same physico-chemical characteristics as the Esk Estuary salt marshes in terms of pH and organic matter content (Table 4.9). Furthermore, only minimal quantities of Sellafield-derived radionuclides are incorporated into the Llandridian Marsh sediments because of the distance between the two sites (Camplin, 1993). There are some differences between Llandridian and the River Esk however, particularly with regard to the size and maturity of the marsh and consequently the vegetation species present. Llandridian marsh is approximately 700 ha and as such is extremely well stabilised and has been colonised by a wide variety of plant species. One particular difference was the lack of *B. mollis* and *H. portulacoides* at Llandridian, although *F. rubra* was present. However, the

samples collected from Llandridian provide a measure of the background radionuclide levels which were produced from weapons testing fallout, the Chernobyl accident and other nuclear accidents.

Table 4.10 compares soil activity data from the River Esk salt marshes with results from Llandridian Marsh. It is evident that the radionuclide content of samples from the River Esk salt marsh is much greater than for Llandridian Marsh, in the majority of cases by two orders of magnitude. This provides strong confirmation that the dominant radionuclide source is Sellafield since the Llandridian Marsh provides a measure of the background radionuclide levels as described above and only contains a small measure of Sellafield radioactivity. For example, Camplin (1993) reported declining ^{137}Cs concentrations in the Irish Sea with increasing distance from Sellafield as seen in the data presented here.

Table 4.10: Comparison of ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities from River Esk and Llandridian salt marshes.

Transect/Site	^{134}Cs	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
Riverward Side					
Front transect	18.5 ± 4.7	8,960 ± 2,650	610 ± 100	2,610 ± 555	8,770 ± 1,970
Middle transect	14.0 ± 6.4	6,720 ± 340	325 ± 40	1,750 ± 125	3,930 ± 635
Back transect	28.0 ± 4.0	5,510 ± 1,670	290 ± 85	1,640 ± 620	2,360 ± 340
Middle transect 0-4 cm*	26.5 ± 5.1	6,390 ± 155	387.0 ± 17.4	1,860 ± 40	4910 ± 90
Middle transect 4-8 cm*	13.9 ± 2.3	14,800 ± 430	5,75.0 ± 5.2	3,120 ± 12	7,770 ± 130
Middle transect 8-12 cm*	<2.1**	2,780 ± 95	96.9 ± 4.7	660 ± 13	1,340 ± 45
Seaward Side					
Front Transect	20.9 ± 3.2	6,650 ± 815	410 ± 145	2,310 ± 880	410 ± 145
Middle Transect	25.7 ± 7.0	11,950 ± 2,780	436 ± 61	3,250 ± 1,290	436 ± 61
Back Transect	14.8 ± 4.1	10,510 ± 2,730	685 ± 150	3,320 ± 770	685 ± 150
Middle transect 0-4 cm*	34.1 ± 5.7	15,900 ± 430	550 ± 45	5,810 ± 160	6,180 ± 120
Middle transect 4-8 cm*	18.1 ± 7.9	18,700 ± 525	490 ± 30	2,550 ± 75	8,270 ± 160
Middle transect 8-12 cm*	15.3 ± 6.5	7,760 ± 230	1,400 ± 40	7,010 ± 85	4,190 ± 120
Llandridian Marsh					
Whole Core	<0.54**	78.9 ± 37.0	3.21 ± 2.13	6.34 ± 3.75	5.93 ± 0.87
0-4 cm*	<0.23**	48.8 ± 1.95	5.34 ± 0.33	2.59 ± 0.23	3.05 ± 0.92
4-8 cm*	<0.19**	89.4 ± 3.13	0.36 ± 0.06	3.67 ± 0.14	3.61 ± 0.78
8-12 cm*	<0.17**	22.2 ± 0.93	0.23 ± 0.04	2.07 ± 0.13	1.66 ± 0.24

Mean Activity in Bq kg^{-1} (± standard error), n=3 for whole core data. Values reported to 3 significant figures.

* Sectioned core error is a 2σ counting error.

** Limit of detection values.

Only ^{137}Cs levels appear to be affected by the presence of material from the Sellafield discharges again reflecting differences in the behaviour of the actinides and caesium as described in section 4.1. The nuclide ratio shows that there are significant differences ($p<0.05$) between the actinide and caesium activities at Llandridian compared to the River Esk. For example, ^{137}Cs:^{241}Am at the River Esk lies between 1.3 and 2.8. Comparing this to the Llandridian site, where the ratio is between 13 and 23, indicates that the ^{241}Am is rapidly adsorbed to the sediment particles in the vicinity of Sellafield, as expected (Hetherington, 1978; McCartney *et al.*, 1994; Pentreath *et al.*, 1983).

4.4.5 Summary of Sediment Results

Sediment core samples from the River Esk salt marsh showed temporal and spatial variation in radionuclide activity, the latter being related mainly to the distance from the river. The spatial variation is exemplified by the range of 3,000 to 8,000 Bq kg^{-1} (^{137}Cs) for the riverward side of the marshland ecosystem. Lower activity levels were characteristic of the landward section of the marsh , and were consistent with the less frequent inundations of this area.

That the main deposition mechanism for activity is the influence of tidal patterns is evidenced by the similar distribution pattern for ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am. Measured activities for surface sediments from the central marsh ranged from 10,800 to 12,300 Bq kg^{-1} (^{137}Cs), 1,100 to 1,450 Bq kg^{-1} (^{238}Pu), 5,700 to 7,000 Bq kg^{-1} ($^{239+240}Pu$), and 12,500 to 15,000 Bq kg^{-1} (^{241}Am) with the ranges reflecting proximity to the river. ^{134}Cs showed no discernible pattern between the landward sectors of the salt marsh and the River Esk. This is attributed to the deposition of radiocaesium following the Chernobyl accident exhibiting a uniform deposition mod ified by airborne and tidal mechanisms.

The seaward side of the marsh showed a similar distribution pattern, but is more influenced by deeply incised creeks that facilitate inundation of the study area earlier and more intensively within the tidal cycle, than would occur in their absence. Thus levels of ^{137}Cs activity varied from 4,000 Bq kg^{-1} well out on the marsh to a peak of 12,000 Bq kg^{-1} at the study site close to the route of the creek.

Sediment core profiles showed a rapid decline in activity below the 20 to 30 cm sector of the 50 cm cores. This reflects accumulation of Sellafield-derived activity in the surface horizons over the last 40 years of deposition history due to accretion of sediment. Peak activity levels at 5 to 10 cm depth on the seaward side were not evident in the riverward profiles, the difference probably reflecting the patterns of sedimentation rate. Much of the radioactivity is

thought to be sediment- and organic matter bound, which will limit its labile properties and bioaccumulation potential.

4.5 STRAND LINE MATERIAL RESULTS

Strand line samples were collected from both sides of the River Esk salt marsh. The importance of the strand line in the ecology of a salt marsh has already been stressed (sections 1.4.3 and 4.2) and examination of this marsh showed just how much invertebrate activity there was and also the evidence of small mammal foraging through, and within, the strand line. In particular, species of *Orchestia* (section 4.7) were extremely common, their habit of jumping when disturbed making them very visible within the strand material. *Orchestia* spp. are considered to be omnivorous scavengers (Fish and Fish, 1989) and combined with other invertebrate species (section 4.7), they play an important in the decomposition of the strand material.

The amount of strand material varied during the course of the study according to seasonal changes and tidal influence. For example, during the autumn and early winter period, the depth and width of the strand layer was considerably greater, covering approximately four times the area observed during the summer. Also, because the depth of the strand line increased by a similar factor, invertebrate activity was still recorded during the winter months deep within the layer. This then provides a source of prey items to small mammals foraging locally.

4.5.1 Composition of strand line material

As already mentioned, the strand line consists mainly of the leaves of *H. portulacoides*, to the extent of 70% of each sample collected. Shortly after the flowering season, the flowering parts were also present in the strand line material, making a total contribution of *H. portulacoides* of over 80%. This is of considerable importance because it has been shown that vegetation in general, and because of its perennial nature *H. portulacoides* in particular, has a considerable effect upon the deposition of radionuclides within the marsh (Horrill, 1983; Jones *et al*, 1994). Horrill (1983) observed higher concentrations of radionuclides within sampling plots on the riverward side of these salt marshes which contained *H. portulacoides* as opposed to other vegetation species. This is mainly because the physical form and structure of *H. portulacoides* is very effective at removing and holding sediment from the water during inundation. Consequently, there is a very large quantity of sediment on both the leaves and woody plant stems. Since it has been shown that nearly all the vegetation contamination on a salt marsh is due to externally adhering sediment (Jones *et al.*, 1994), a very large component of the radioactivity is moved to the strand line when the leaves of *H. portulacoides* are shed. This

provides a very important mechanism for the transfer of radionuclides to an area utilised extensively by invertebrates and small mammals for feeding and hence it is of primary importance in the food chain transfer of radionuclides.

The remaining strand material consists of other plant and animal detritus brought in by the tides and from upstream; or from the other plant species present on the salt marsh. Also, there is a 3 to 5% component of litter and flotsam which has been washed up on the shore. Where litter was present in a sample, it was discarded as being non-biological and therefore of little consequence. The presence of litter is however of importance to small mammals. Animal runs or feeding sites were frequently observed within or under pieces of litter or flotsam as they afford protection against predation and will, to some extent, encourage the use of the strand for foraging. Another important factor in determining the time spent foraging within the strand material is the number of tidal inundations which reach the upper parts of the marsh. Using predicted tide height data (Laver, 1993; 1994; 1995), there are on average six inundations over 9 m each month, usually occurring over a period of 3 to 5 days. This means that foraging is possible for long periods each month without the animals being disturbed by tides.

4.5.2 Temporal and Spatial Distribution

Samples of strand material were analysed and the results are presented in Figures 4.12 to 4.15. The overall activity data for the two sides of the marsh are remarkably similar, again indicating the importance of *H. portulacoides* which is present in large pure stands on both sides of the railway viaduct. On both sides the ^{137}Cs activity ranged between 180 Bq kg^{-1} to 600 Bq kg^{-1}. Ranges for ^{238}Pu, $^{239+240}Pu$ and ^{241}Am were in the order of 20 to 60 Bq kg^{-1}; 80 to 250 Bq kg^{-1}; and 150 to 450 Bq kg^{-1}, respectively. In general, the levels on the riverward marsh were fractionally greater than the seaward side, again a function of the increased sedimentation rate on the riverward side as a consequence of water flow around the viaduct as described earlier (section 4.41). Student *t* tests for each of the radionuclides show that the activities are not significantly different for the two sides ($p>0.05$). The actinides however are significantly different ($p<0.05$ for ^{238}Pu and $^{239+240}Pu$; $p<0.01$ for ^{241}Am), probably a reflection of the behavioural differences of caesium and the actinides in a water body.

4.5.3 Results of temporal variation ANOVA

The two sides also exhibited a different pattern of activity over time. The riverward side (A) showed some evidence of a cyclic trend (Figures 4.12 and 4.13) in each of the radionuclides activities, with an increase during the autumn to spring period and a decline during the summer months. Results from a One-way ANOVA presented in Table 4.11 show significant

Figure 4.12: **^{137}Cs, $^{239+240}Pu$ and ^{241}Am mean activities (Bq kg^{-1} ± standard deviation, n=2) in flotsam samples collected from the riverward (A) marsh.**

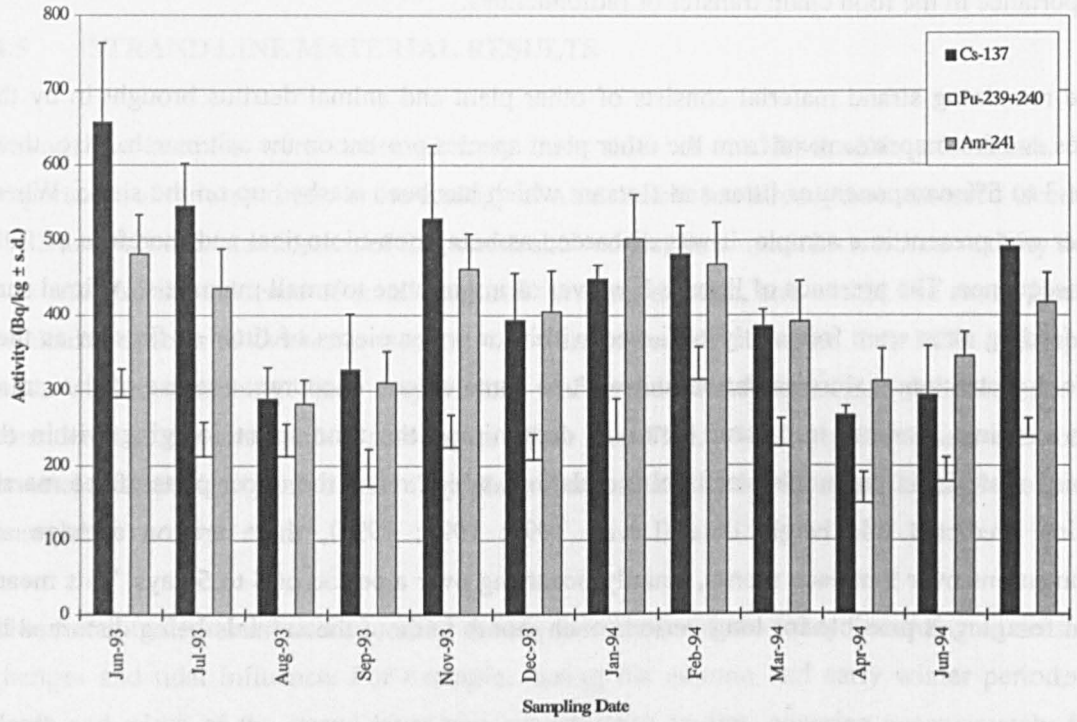
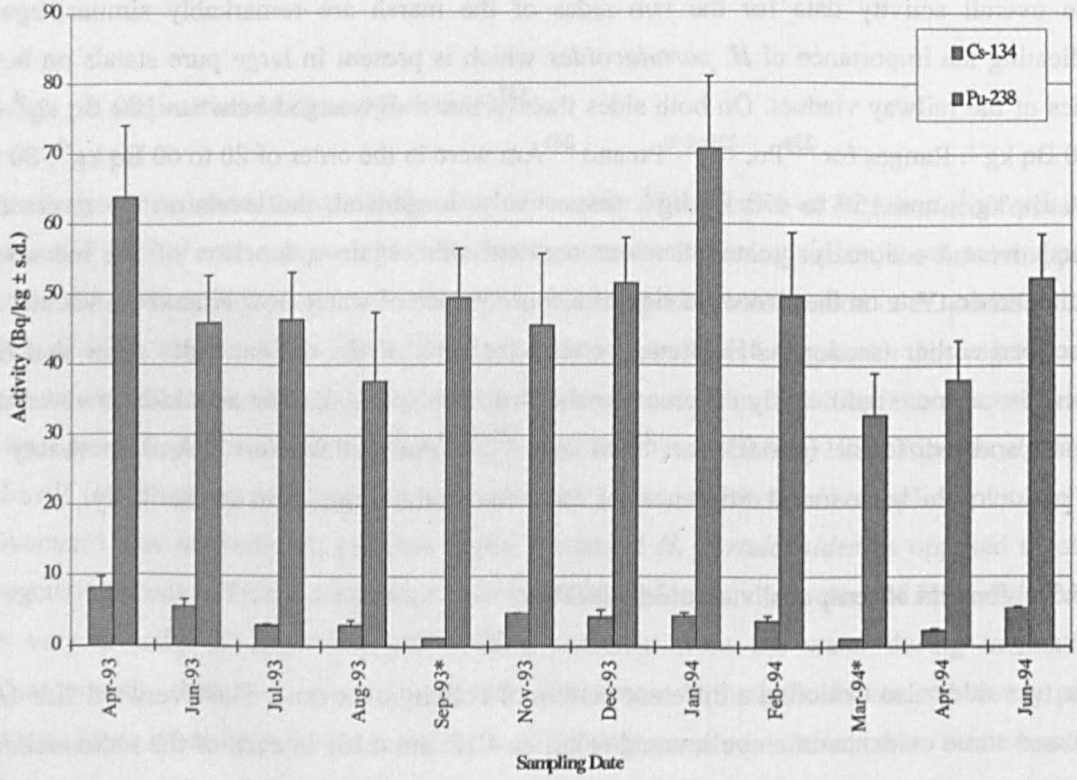


Figure 4.13: **^{134}Cs and ^{238}Pu mean activities (Bq kg^{-1} ± standard deviation, n=2) in flotsam samples collected from the riverward (A) marsh.**



* = ^{134}Cs values reported are < LOD values.

Figure 4.14: **^{137}Cs, $^{239+240}Pu$ and ^{241}Am mean activities (Bq kg^{-1} ± standard deviation, n=2) in flotsam samples collected from the seaward (B) marsh.**

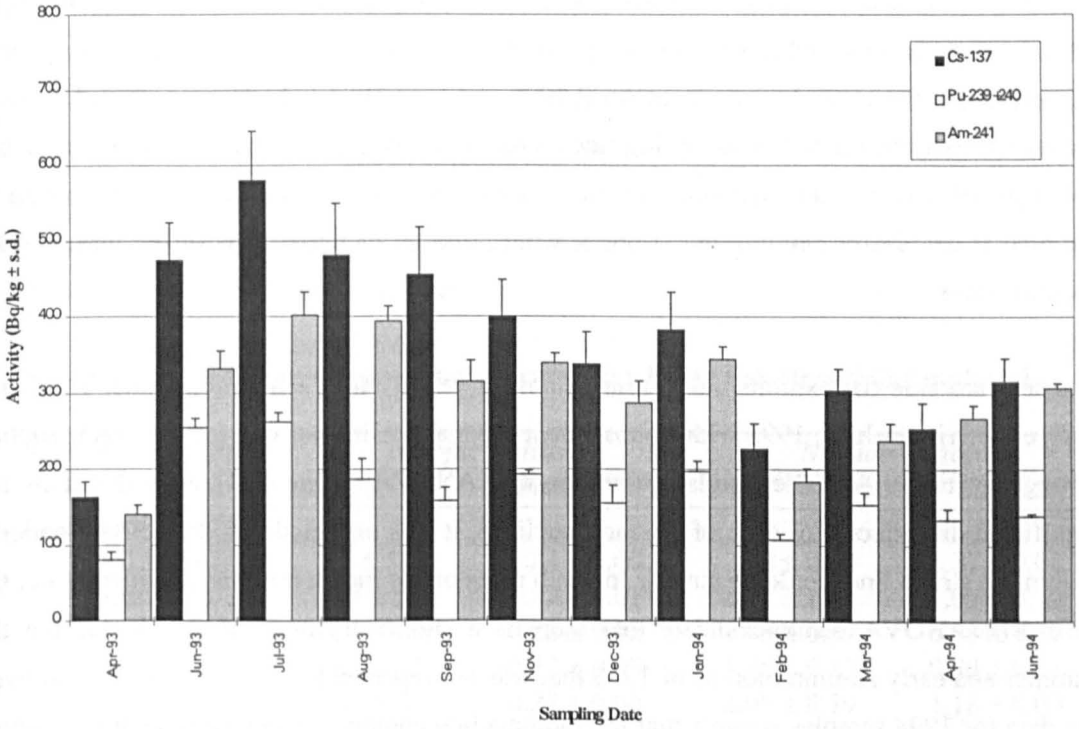
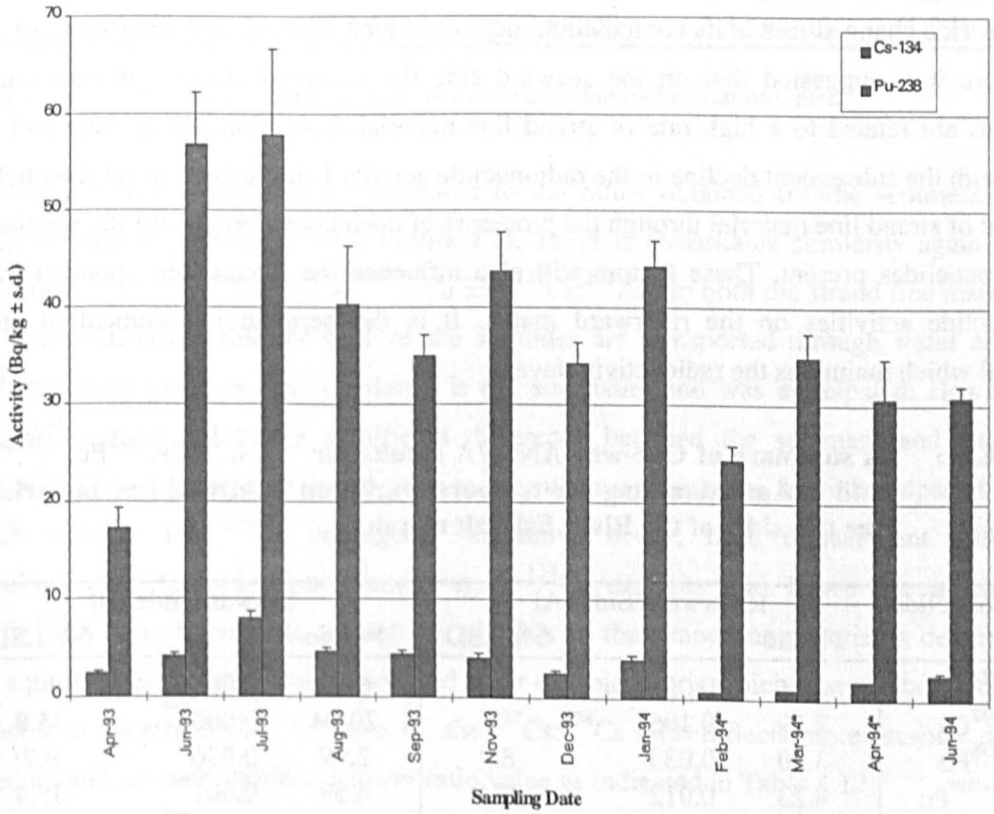


Figure 4.15: **^{134}Cs and ^{238}Pu mean activities (Bq kg^{-1} ± standard deviation, n=2) in flotsam samples collected from the seaward (B) marsh.**



* = ^{134}Cs values reported are < LOD values.

temporal differences for three of the radionuclides, ^{134}Cs, ^{238}Pu and $^{239+240}Pu$ ($p<0.05$). It is clear from the comparison of the 5% LSD values that it is specific months which are significantly different. In general, April, September, November and December 1993 and June 1994 are significantly higher than the other months, as indicated in Figures 4.12 and 4.13. It is suggested that the peaks in activity in April 1993 and June 1994 (which was sampled early in the month) may be related to the spring tides. From the data it is evident that there are higher levels of radionuclides in the strand line during the winter period. This reflects the increase in the tidal reach, higher rainfall and stronger winds, and an increased loss of the leaves of *H. portulacoides*.

The seaward side (B) exhibits a different pattern of activity for each radionuclide. There is a peak of activity in June, 1993 with a subsequent gradual decline activity for each radionuclide through to June, 1994. The results of the One-way ANOVA (Table 4.11) show that there are significant differences for each of the radionuclides. It was expected that the activity patterns within the strand line would be similar for both sides of the marsh but this is evidently not the case. The ANOVA results indicate that there is a significant peak of activity during the summer and early autumn months of 1993 that was not repeated the following year. However, the data for 1994 samples suggest that the radionuclide content is more consistent throughout the year.

During the two year sampling period, it was observed that the strand line material on the seaward side changed less in its composition, depth and area covered than the riverward side. Therefore it is suggested that on the seaward side the observed changes in radionuclide activities are related to a high rate of strand line material deposition during June and July 1993, with the subsequent decline in the radionuclide activity being related to (a) the physical removal of strand line material through the processes of decomposition and (b) the weathering of radionuclides present. These factors will also influence the fluctuations apparent in the radionuclide activities on the riverward marsh. It is the periodic replacement of strand material which maintains the radioactivity levels.

Table 4.11: A summary of One-way ANOVA results for ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am examining for temporal variation in strand line material on the two sides of the River Esk salt marsh.

Radionuclide	Riverward Side (A)			Seaward Side (B)		
	F-ratio	P	5% LSD	F-ratio	P	5% LSD
^{134}Cs	4.16	0.021*	1.4	6.19	0.005**	1.1
^{137}Cs	2.19	0.105		20.94	0.000***	35.8
^{238}Pu	3.20	0.033*	8.9	2.99	0.036*	9.7
$^{239+240}Pu$	4.23	0.012*	32.9	4.59	0.007**	35.4
^{241}Am	2.37	0.077		26.98	0.000***	21.2

4.5.4 Isotopic and Nuclide Ratios

Table 4.12 presents data on the mean isotopic and nuclide ratios for five of the months in which strand line material was sampled. The results exhibit little temporal variation, with only the ^{137}Cs:^{134}Cs ratio fluctuating by very much. However, most of this fluctuation is masked by the high associated standard deviation which is also reported. Similarly, there is little difference in the radionuclide ratios for the two sides of the marsh. For example, the plutonium isotopic ratio is remarkably similar between the two sides and over time, averaging 0.22 with a very low standard deviation.

Table 4.12: Mean isotopic and nuclide ratios for River Esk strand line material.

Sample Site/Month	*Isotopic Ratios*		*Nuclide Ratios*	
	^{137}Cs:^{134}Cs	^{238}Pu:$^{239+240}Pu$	^{137}Cs:$^{239+240}Pu$	^{137}Cs:^{241}Am
Riverward Side (A)				
April 1993	81 ± 15	0.22 ± 0.00	2.24 ± 0.40	1.37 ± 0.02
August 1993	113 ± 50	0.22 ± 0.01	2.04 ± 1.02	1.05 ± 0.10
December 1993	105 ± 4.9	0.22 ± 0.02	1.88 ± 0.57	0.88 ± 0.23
March 1994	240 ± 2.1	0.22 ± 0.03	1.86 ± 0.12	0.88 ± 0.01
June 1994	156 ± 95	0.22 ± 0.00	2.08 ± 0.19	1.18 ± 0.09
Seaward Side (B)				
April 1993	68 ± 6.3	0.22 ± 0.01	2.06 ± 0.34	1.16 ± 0.17
August 1993	103 ± 5.7	0.22 ± 0.00	2.59 ± 0.03	1.22 ± 0.11
December 1993	148 ± 57	0.22 ± 0.02	2.21 ± 0.24	1.18 ± 0.09
March 1994	283 ± 0.7	0.23 ± 0.02	2.01 ± 0.32	1.28 ± 0.05
June 1994	108 ± 17	0.22 ± 0.00	2.32 ± 0.13	1.04 ± 0.05

Values reported to 3 significant figures as an average (± standard deviation), n=2.

The data in Table 4.12 can be compared to the ratios obtained for the sediment samples collected during September 1994 (Table 4.7). There is remarkable similarity again between the ratios, ^{238}Pu:$^{239+240}Pu$, ^{137}Cs:$^{239+240}Pu$ and ^{137}Cs:^{241}Am in both the strand line material and the sediment. Given that the bulk of the actinides are transported through water associated with sediment particles, this similarity is not surprising and was anticipated. However, the ^{137}Cs:^{134}Cs ratio exhibits a significant difference between the sediment and strand line material. This was also anticipated since sediment cores were taken from the edges of the tidal reach and, as the ^{134}Cs undergoes radioactive decay, little replacement will occur. Consequently, the sediments contain aged ^{134}Cs deposits and hence the much higher ^{137}Cs:^{134}Cs ratio. In contrast, the activity of ^{134}Cs on the strand line material is determined by the activity present on the vegetation and other organic debris which is water-borne during its transfer to the strand line. In this case, the ^{137}Cs:^{134}Cs ratio reflects more recently deposited caesium and, as such, exhibits a lower ratio value as indicated in Table 4.12.

4.5.5 Summary of Strand Line Material

The extent and biomass of the strand line varied seasonally and comprised mainly the leaf litter of *H. portulacoides*. Activity levels were similar for the seaward and riverward sectors of the marsh, with the ranges being : 180 to 600 Bq kg^{-1} (^{137}Cs), 20 to 60 Bq kg^{-1} (^{238}Pu), 80 to 250 Bq kg^{-1} ($^{239+240}Pu$) and 150 to 450 Bq kg^{-1} (^{241}Am).

Temporal differences between the riverward and seaward sectors of the marsh were more obvious than were spatial differences. The riverward sector showed higher activity levels between autumn and spring, peaking in mid-winter, compared to the summer. For the seaward side the activity peak was in June, a fact that is attributed to high rates of deposition in the summer and subsequent attrition and weathering later in the year.

4.6 VEGETATION RESULTS

The vegetation species across the upper tidal reaches of this salt marsh are representative of normal grassland communities as they have succeeded the more salt-tolerant species. As a consequence it was expected that small mammals, particularly *Microtus agrestis* and *Apodemus sylvaticus*, will feed within this 'grassland corridor'. On the marsh itself, halophytic species such as *Halimione portulacoides, Puccinellia maritima, Salicornia europaea* and *Spartina anglica* dominate. These species contribute significantly to the strand line material as described in section 4.5.

Across the marsh, different vegetation species are present in distinct communities or zones. This zonation of vegetation is a common feature of all salt marshes and occurs throughout their development. Three basic zones can be distinguished: low, middle and upper marsh (Long and Mason, 1983). The boundaries between them depend upon the tidal reach and wave action. In this study, the particular interest was in the middle and upper marsh areas. Although the vertical zonation of plant species is determined mainly by tidal amplitude, on a local scale the presence of creeks can influence the plant communities. For example, around the fringes of the creeks, *H. portulacoides* was observed, whilst it was absent from other areas at the same height but distant from the creeks. Other species located around the fringes of creeks included *P. maritima* and *S. europaea*. Such observations are common on salt marshes and are reported to be caused by differences in the drainage of the sediments (Long and Mason, 1983).

In the upper zone, particularly at the edge of the tidal reach, the distribution of the vegetation varies across the two sides of the marsh. However, the same species are present. For example, *Festuca rubra* is more abundant on the seaward side of the marsh while *Bromus mollis* forms

the dominant vegetation on both sides. These two species dominated the vegetation samples collected and are believed to form the bulk of the vegetation in the herbivorous small mammal diet. During the second year of study, additional vegetation samples were collected from the main part of the marsh to clarify the deposition of activity across the site as a whole. These samples consisted primarily of *H. portulacoides*.

4.6.1 Spatial and Temporal Variation

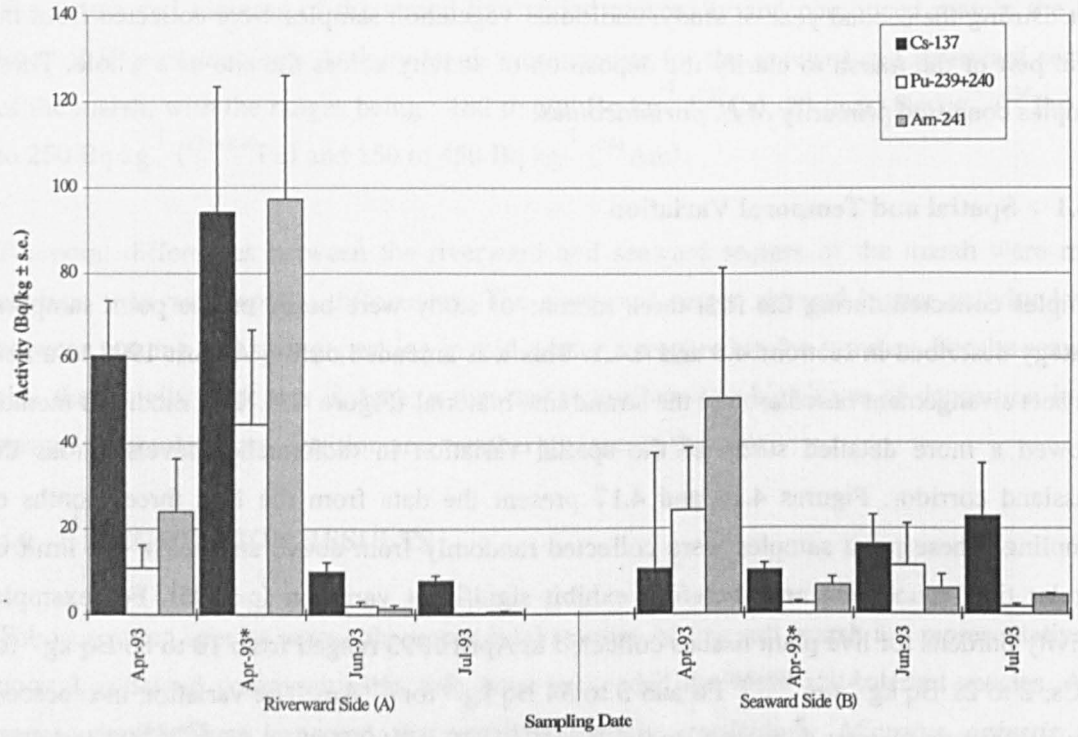
Samples collected during the first three months of study were based on the point sampling strategy described in sections 4.3 and 4.4.1. This was amended during August 1993 to a new transect arrangement based around the strand line material (Figure 4.1). This modified method allowed a more detailed study of the spatial variation in radionuclide levels across the grassland corridor. Figures 4.16 and 4.17 present the data from the first three months of sampling. These point samples were collected randomly from above and below the limit of regular tidal inundation and therefore exhibit significant variation ($p<0.05$). For example, activity burdens for live plant tissues collected in April 1993 ranged from 10 to 80 Bq kg^{-1} for ^{137}Cs, 2 to 21 Bq kg^{-1} for $^{239+240}Pu$ and 3 to 54 Bq kg^{-1} for ^{241}Am. The variation in senescent tissue was even greater; 44 to 166 Bq kg^{-1} for ^{137}Cs, 10 to 108 Bq kg^{-1} for $^{239+240}Pu$ and 50 to 174 Bq kg^{-1} for ^{241}Am. The bulk of these samples (>90%) comprised *B. mollis*, the most abundant species at that time. Senescent *B. mollis* was collected during April, 1993. Observations of *B. mollis* over the two years of study showed that during the late autumn/winter period, senescent material dominated to the extent that live tissue samples could not easily be collected.

The data in Figures 4.16 and 4.17 contributed to the decision to amend the sampling strategy. When the six marsh transects were laid down during August 1993, vegetation samples were collected every five metres as were the sediment cores. These composite vegetation samples comprised the available plant material at each set position. The results are presented in Figures 4.18 and 4.19. Samples were counted to produce 2σ counting errors (<3%).

4.6.2 Vegetation Marsh transects

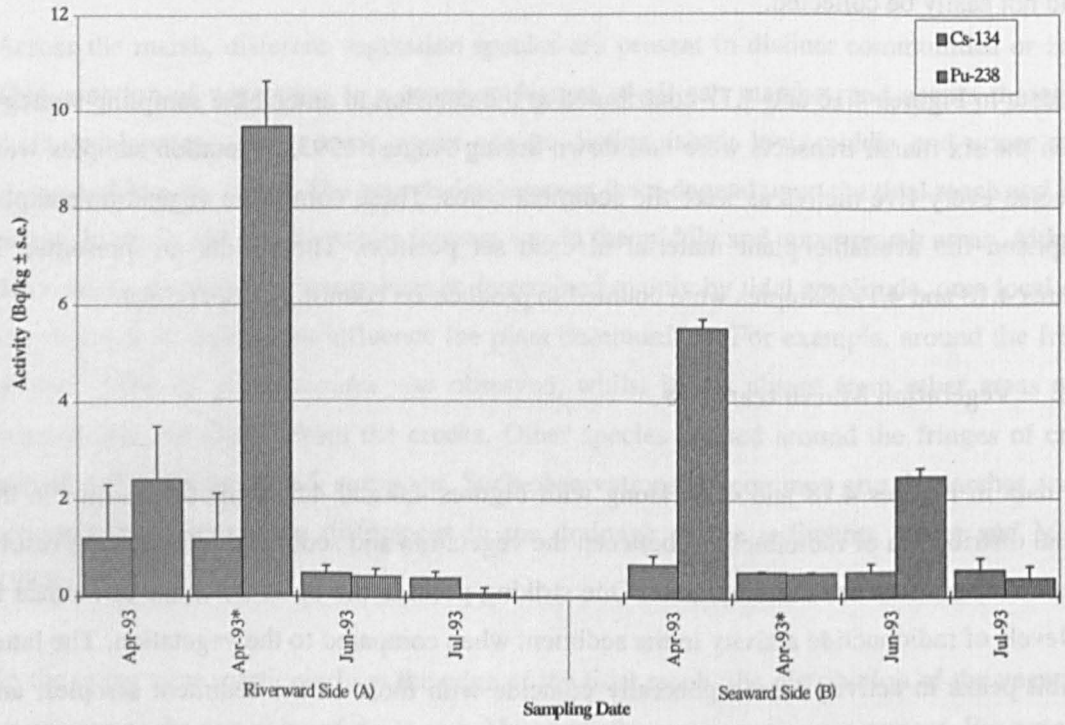
The data in Figures 4.18 and 4.19, along with Figures 4.4 and 4.5, show similarities in the spatial distribution of radionuclides between the vegetation and sediment, although the results from the vegetation are more variable. One striking point is the up to 30 times difference in the levels of radionuclide activity in the sediment when compared to the vegetation. The latter exhibit peaks in activity which generally coincide with those in the sediment samples, and there is an increase in radionuclide levels from the back of the marsh towards the river. This is in agreement with the hypothesis in section 4.4.1 regarding the spatial distribution of

Figure 4.16: **^{137}Cs, $^{239+240}Pu$ and ^{241}Am activities from composite grass samples collected during the first three months of study.**



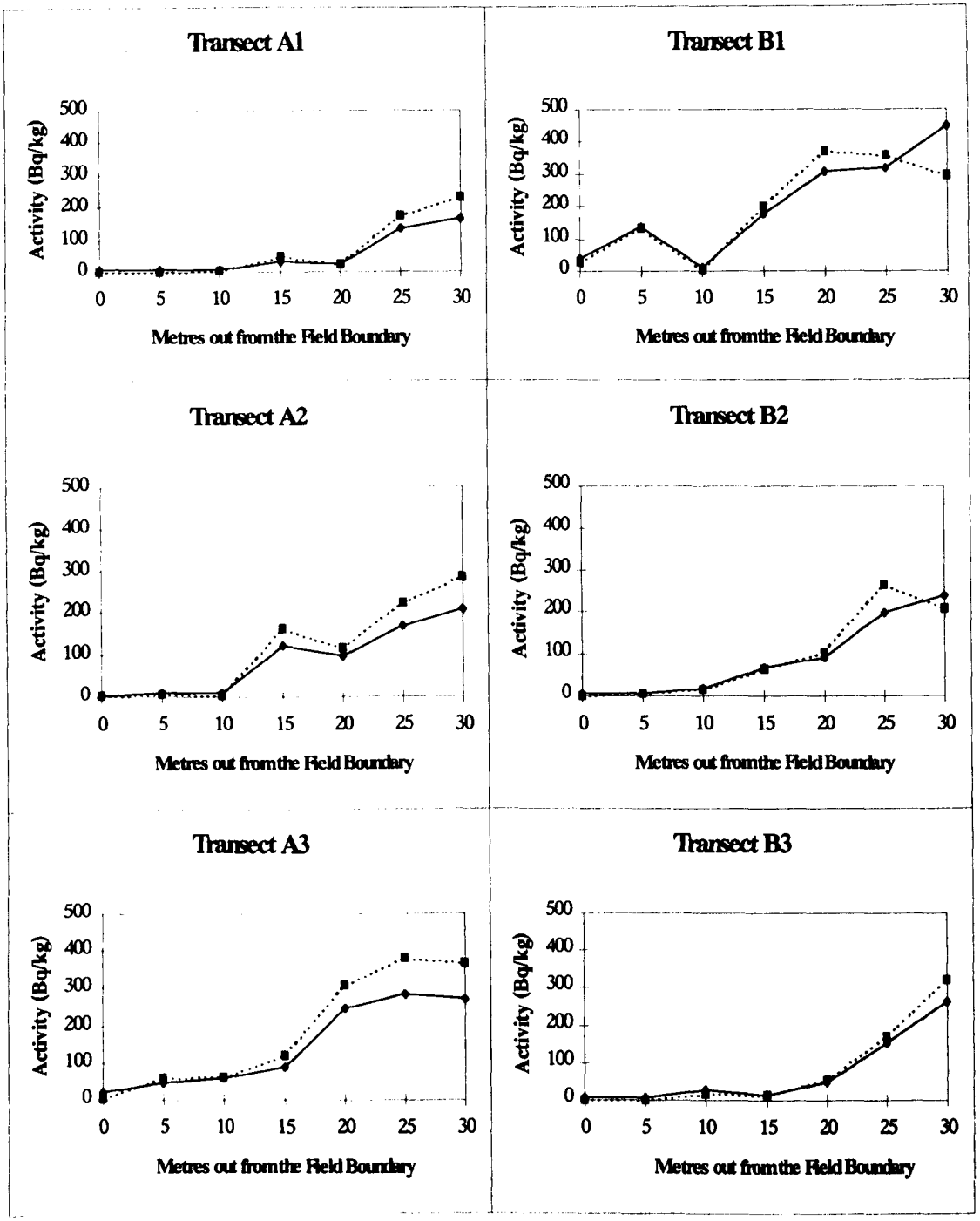
* = Samples of senescent material.

Figure 4.17: **^{134}Cs and ^{238}Pu activities from composite grass samples collected during the first three months of study.**



* = Samples of senescent material.
LOD values were used during the calculation of the means for all the ^{134}Cs results.

Figure 4.18: **Line graphs of the River Esk marsh transects (A and B, riverward and seaward sides respectively) showing ^{137}Cs and ^{241}Am vegetation results.**

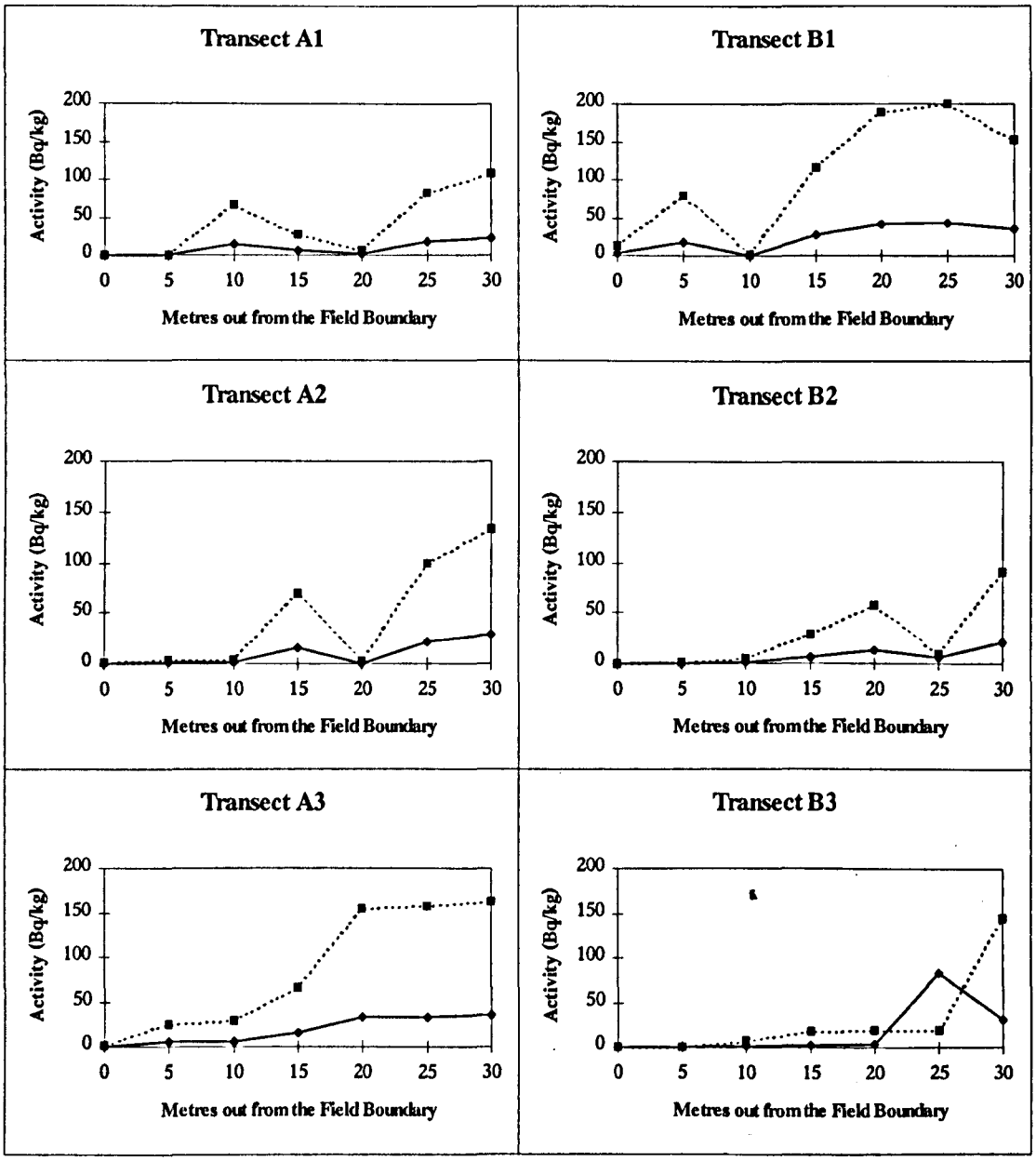


Counting errors all <3% (2σ), error bars omitted for clarity.
——— = ^{137}Cs.
..... = ^{241}Am.

radionuclides across the marsh. It is also notable that all four radionuclides (^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am) behaved in a similar manner.

The vegetation activity data from the transects differ from those of the sediment with regard to the presence of the large creeks. Whereas the sediment samples exhibited peaks in activity towards the back of the marsh, there is little evidence of a similar occurrence in the vegetation. This may be due to the long term accumulation of radionuclides in the sediment,

Figure 4.19: Line graphs of the River Esk marsh transects (A and B, riverward and seaward sides respectively) showing ^{238}Pu and $^{239+240}Pu$ vegetation results.



Counting errors all <3% (2σ), error bars omitted for clarity.
——— = $^{239+240}Pu$.
..... = ^{238}Pu.

with additional factors such as rainfall, wind and wave action weathering the radionuclides associated with plant foliage. The action of rain and wind will be particularly important towards the back of the marsh where tidal inundation is less frequent. A recent study on Ince Marsh in the Mersey Estuary has shown that the bulk of the radionuclide contamination was associated with sediment attached to foliage (Jones *et al.*, 1994). Furthermore, marked temporal variation was observed consistent with the deposition of suspended sediments during tidal inundation, with a subsequent decline in contamination due to weathering with a half life of 20 to 30 days (Jones *et al.*, 1994). At the limit of the tidal reach, the amount of radioactivity

present on the plant foliage will be related to the time elapsed since the last full inundation. Moreover, the concentration of radionuclides will depend upon the activity in the suspended sediments and also the frequency of tidal inundations. It is thus feasible that the concentration of radionuclides associated with vegetation at the back of the marsh would be low and that the observed activity may not reflect the influence of the creeks.

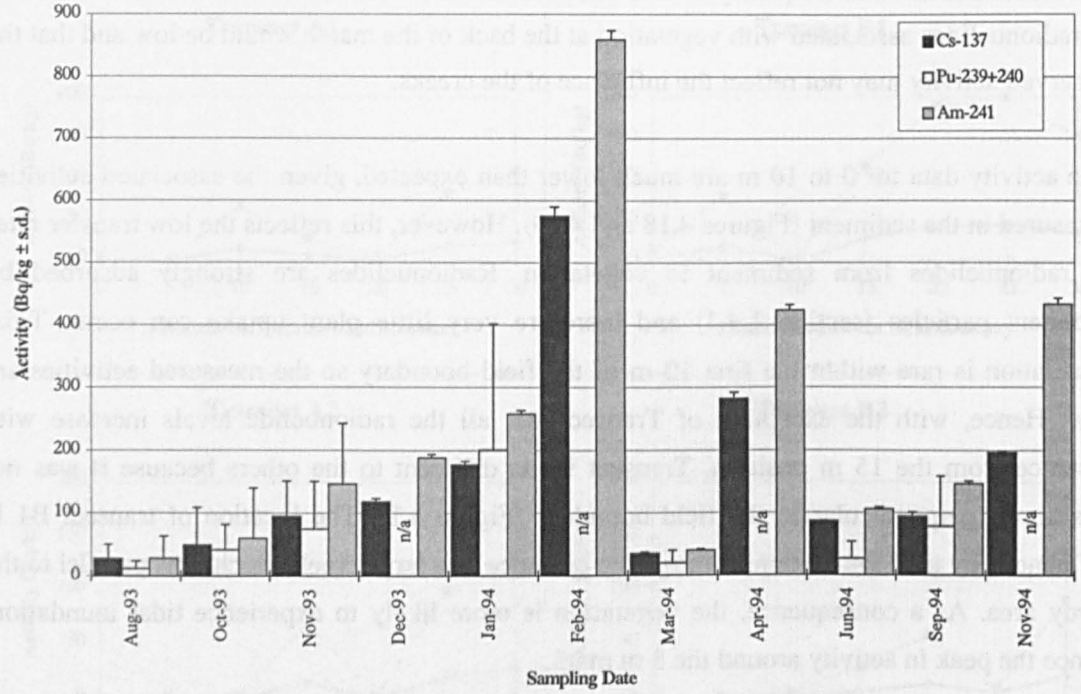
The activity data for 0 to 10 m are much lower than expected, given the associated activities measured in the sediment (Figures 4.18 and 4.19). However, this reflects the low transfer rates of radionuclides from sediment to vegetation. Radionuclides are strongly adsorbed by sediment particles (section 1.4.1) and therefore very little plant uptake can occur. Tidal inundation is rare within the first 10 m of the field boundary so the measured activities are low. Hence, with the exception of Transect B1, all the radionuclide levels increase with distance from the 15 m position. Transect B1 is different to the others because it was not positioned perpendicular to the field boundary (Figure 4.1). The location of transect B1 is influenced by tidal inundations through the presence of a large creek which runs parallel to the study area. As a consequence, the vegetation is more likely to experience tidal inundation, hence the peak in activity around the 5 m mark.

Further information on the spatial distribution of radionuclides was derived from the amended sampling strategy. By sampling vegetation using transects running behind, along and in front of the strand line (relative to the river), the area believed to be utilised by small mammals and invertebrates was more accurately assessed. Figures 4.20 to 4.32 present the ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am results for *F. rubra* and *B. mollis* across the two sides of the marsh. Figures 4.22 to 4.32 clearly show increased activities along the front transect when compared to the middle (strand line) and back transects.

4.6.2 Radionuclide levels in *Festuca rubra* on the Riverward marsh

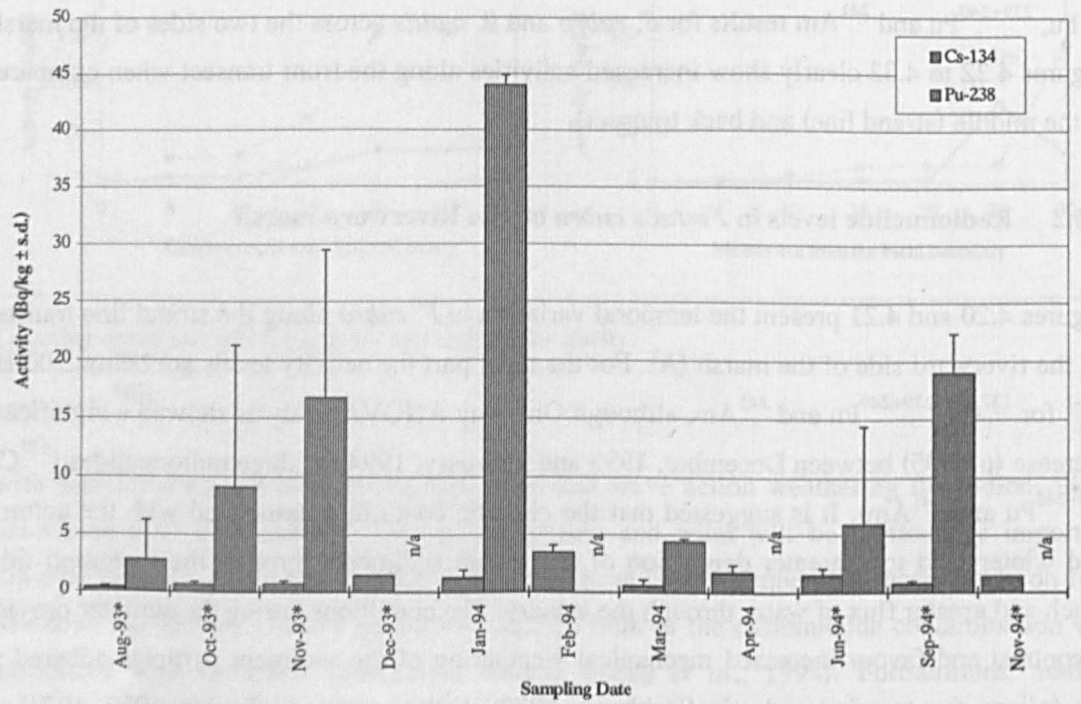
Figures 4.20 and 4.21 present the temporal variation in *F. rubra* along the strand line transect on the riverward side of the marsh (A). For the most part the activity levels are below 100 Bq kg^{-1} for ^{137}Cs, $^{239+240}Pu$ and ^{241}Am, although One-way ANOVA analysis showed a significant increase ($p<0.05$) between December, 1993 and February, 1994 for three radionuclides (^{137}Cs, $^{239+240}Pu$ and ^{241}Am). It is suggested that the climatic conditions associated with the autumn and winter lead to a greater deposition of suspended sediments through the increased tidal reach and greater flux of water through the estuary. The conditions during the summer provide a contrast and favour increased mechanical weathering of the sediment particles adhered to the foliage due to wind and rain (Dahlman, 1972; Witherspoon and Taylor, 1970, 1971). It was observed that during the autumn/winter much of the vegetation on the marsh is covered by sediment particles, evident as a grey film over the foliage. This could also be important

Figure 4.20: ^{137}Cs, $^{239+240}Pu$ and ^{241}Am activities in *F. rubra* collected from the Riverward Marsh (A).



n/a = Sample not analysed.
Samples only collected from along the strand line.

Figure 4.21: ^{134}Cs and ^{238}Pu activities in *F. rubra* collected from the Riverward Marsh (A).



n/a = Sample not analysed.
* = LOD values used in the calculations.
Samples only collected from along the strand line.

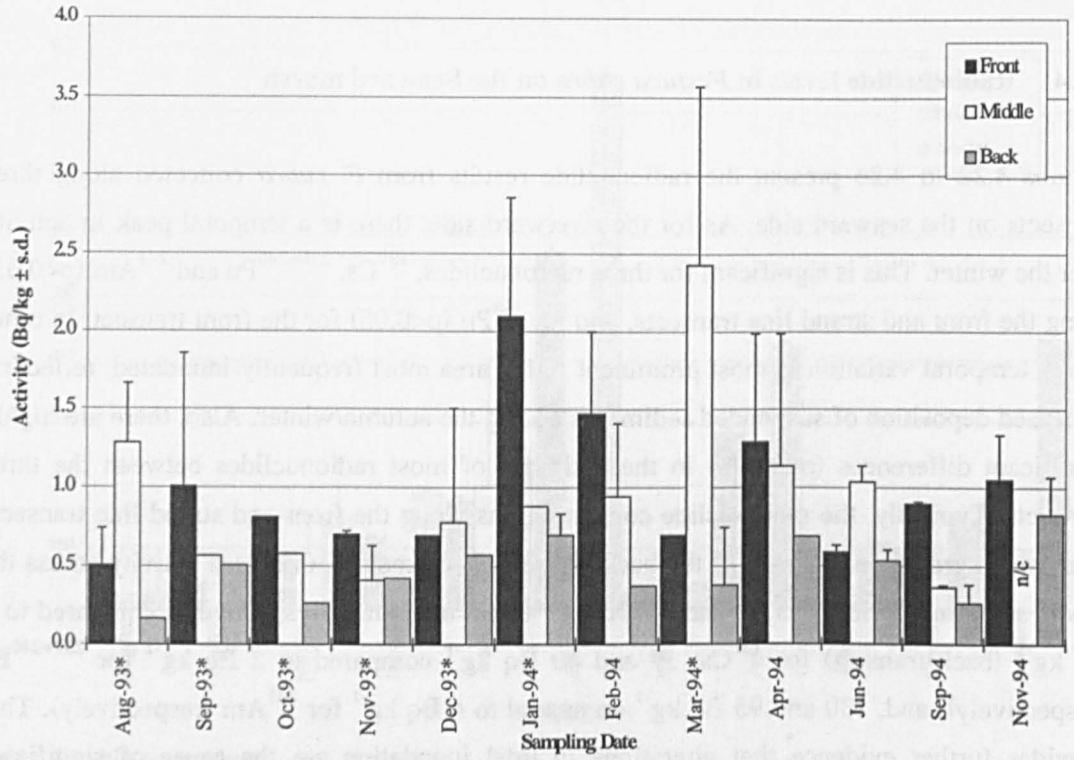
because *F. rubra* is known to be selectively grazed by *M. agrestis*, although animals are unlikely to feed on material which is of 'poor' quality.

4.6.4 Radionuclide levels in *Festuca rubra* on the Seaward marsh

Figures 4.22 to 4.26 present the radionuclide results from *F. rubra* collected along three transects on the seaward side. As for the riverward side, there is a temporal peak in activity over the winter. This is significant for three radionuclides, ^{137}Cs, $^{239+240}Pu$ and ^{241}Am ($p<0.05$) along the front and strand line transects, and for ^{238}Pu ($p<0.05$) for the front transect. In other words temporal variation is most prominent in the area most frequently inundated, reflecting increased deposition of suspended sediments during the autumn/winter. Also, there are highly significant differences ($p<0.001$) in the activities of most radionuclides between the three transects. Typically, the radionuclide concentrations along the front and strand line transects were much greater than those for the back transect. For example, the mean activity across the whole sampling period was 96 and 78 Bq kg^{-1} (front and strand respectively) compared to 9 Bq kg^{-1} (back transect) for ^{137}Cs; 39 and 40 Bq kg^{-1} compared to 2 Bq kg^{-1} for $^{239+240}Pu$ (respectively); and, 130 and 95 Bq kg^{-1} compared to 6 Bq kg^{-1} for ^{241}Am (respectively). This provides further evidence that alterations in tidal inundation are the cause of significant changes in the levels of radionuclides across the site. Tables 4.13 and 4.14 provide summaries of One-way ANOVA results and report the 5% LSD values for those which are significant.

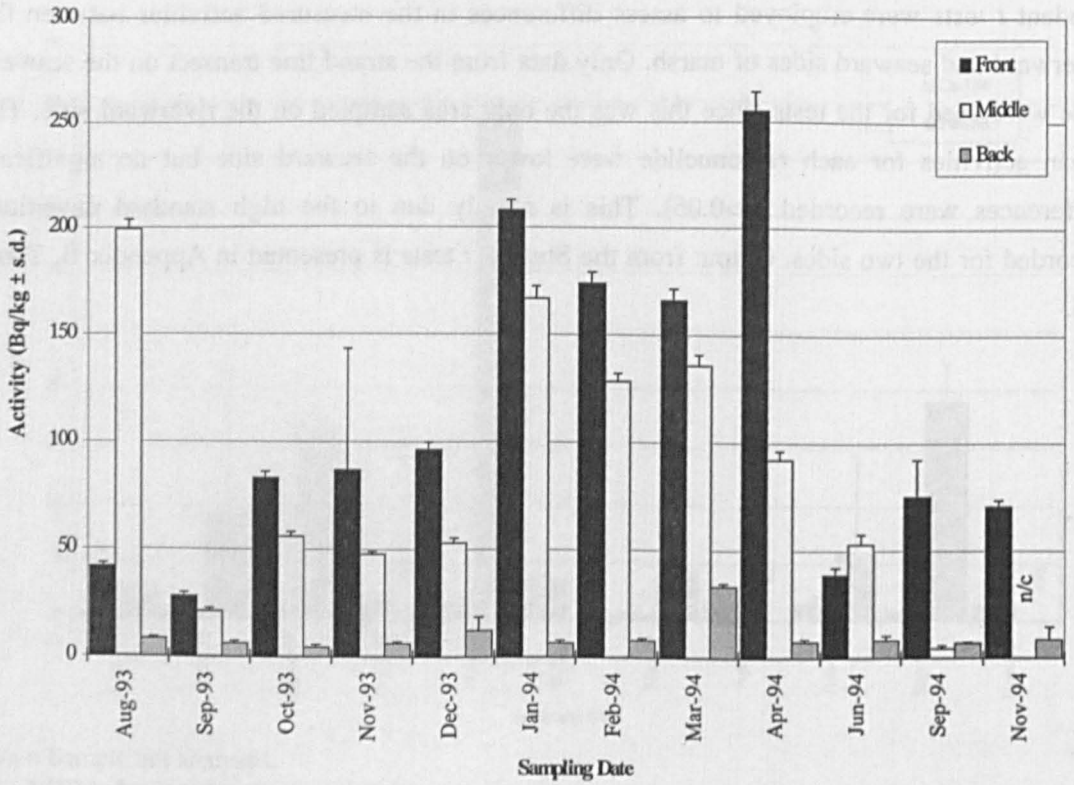
Student *t* tests were employed to assess differences in the measured activities between the riverward and seaward sides of marsh. Only data from the strand line transect on the seaward side were used for the tests since this was the only area sampled on the riverward side. The mean activities for each radionuclide were lower on the seaward side but no significant differences were recorded ($p>0.05$). This is mainly due to the high standard deviations recorded for the two sides. Output from the Student *t* tests is presented in Appendix B, Table B5.

Figure 4.22: ^{134}Cs activity in *Festuca rubra* collected from the Seaward Marsh (B).



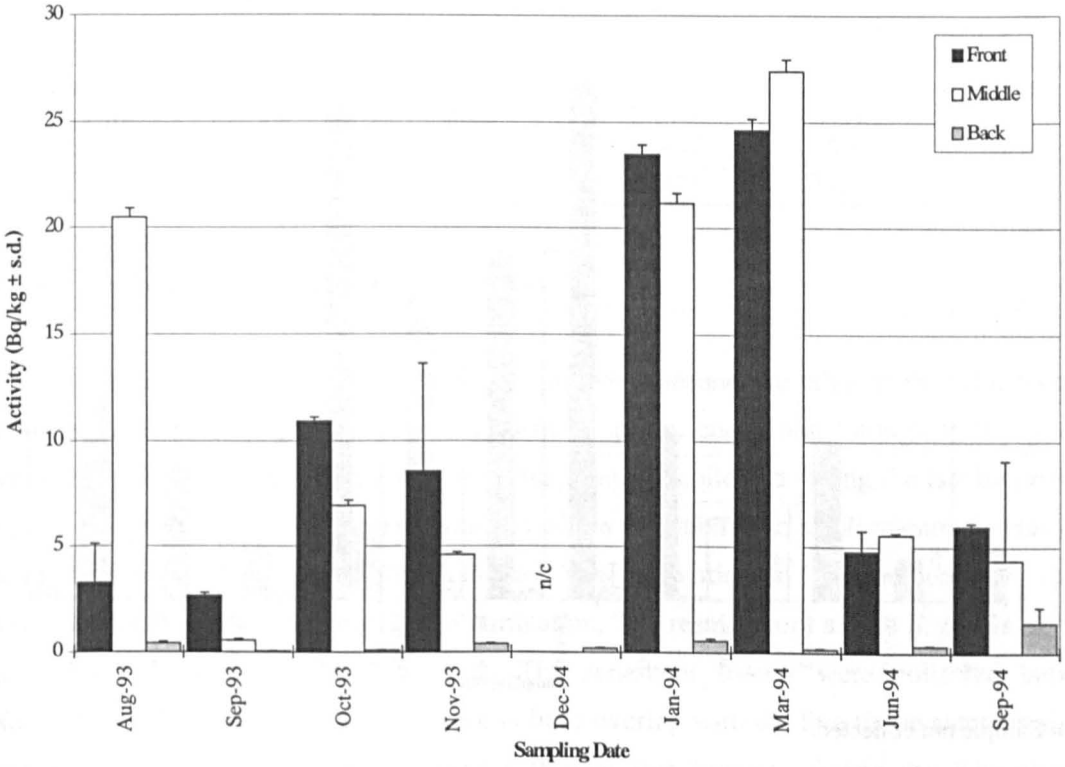
* = LOD values used in the calculations.
n/c = Sample not collected.

Figure 4.23: ^{137}Cs activity in *Festuca rubra* collected from the Seaward Marsh (B).



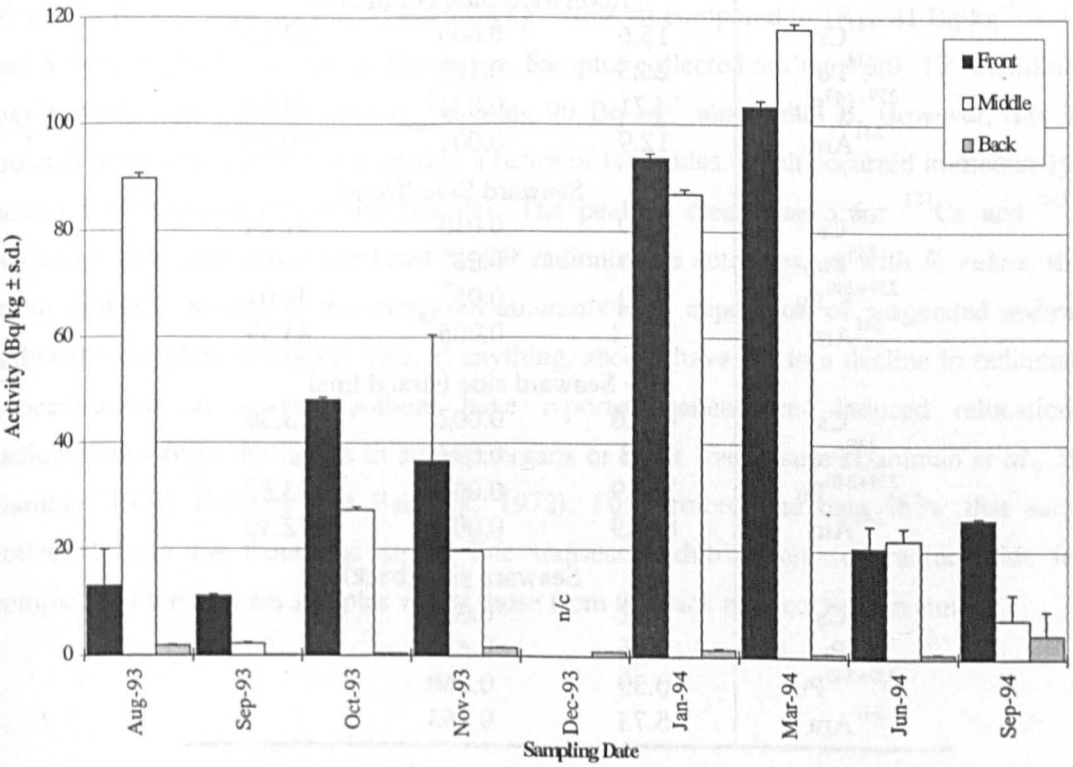
n/c = Sample not collected.

Figure 4.24: ^{238}**Pu activity in *Festuca rubra* collected from the Seaward Marsh (B).**



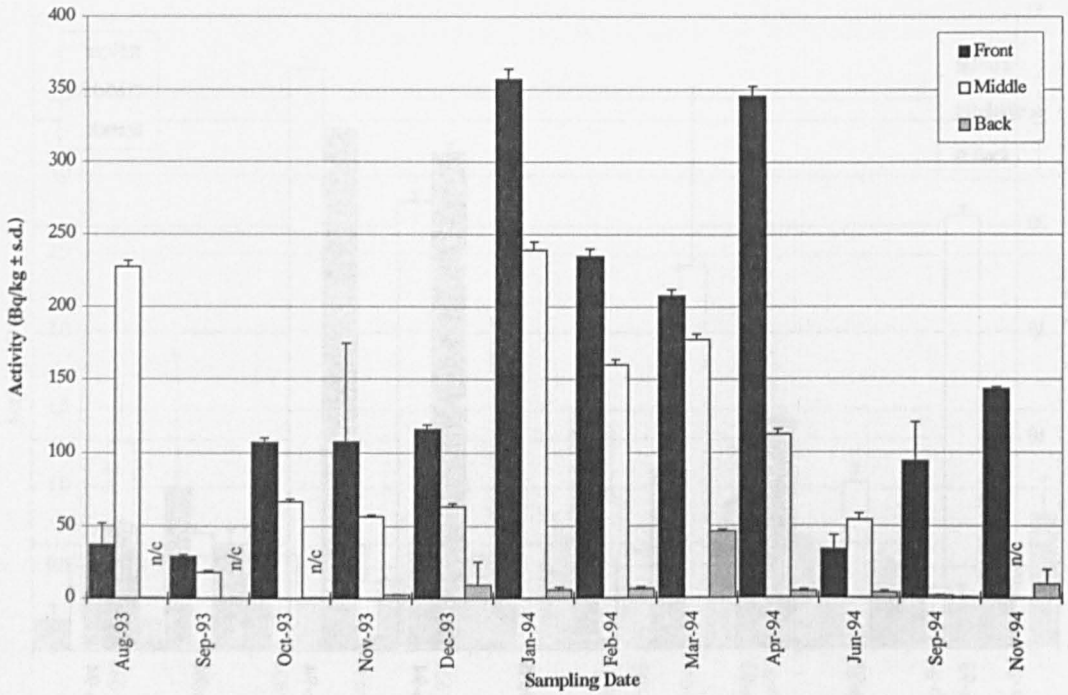
n/c = Sample not collected.

Figure 4.25: $^{239+240}$**Pu activity in *Festuca rubra* collected from the Seaward Marsh (B).**



n/c = Sample not collected.

Figure 4.26: ^{241}Am activity in *Festuca rubra* collected from the Seaward Marsh (B).



n/c = Sample not collected.

Table 4.13: Summary of One-way ANOVA results for temporal variation in *F. rubra* samples from the River Esk salt marsh[a].

Radionuclides	F-ratio	P	5% LSD
	Riverward Side (strand line)		
^{137}Cs	15.6	0.000***	53.15
^{238}Pu	2.23	0.159	
$^{239+240}$Pu	4.71	0.031*	48.92
^{241}Am	12.9	0.000***	90.56
	Seaward Side (front)		
^{137}Cs	8.89	0.013*	41.44
^{238}Pu	8.80	0.05*	7.05
$^{239+240}$Pu	9.50	0.05*	34.01
^{241}Am	13.53	0.005**	83.59
	Seaward side (strand line)		
^{137}Cs	403.0	0.002**	13.36
^{238}Pu	9.93	0.094	
$^{239+240}$Pu	159.9	0.006**	13.87
^{241}Am	824.9	0.001***	12.30
	Seaward side (back)		
^{137}Cs	5.42	0.058	
^{238}Pu	1.45	0.412	
$^{239+240}$Pu	0.39	0.860	
^{241}Am	5.73	0.053	

[a] ^{134}Cs data are not presented due to the large number of < LOD values.

* Significant at $p<0.05$, ** at $p<0.01$ and *** $p<0.001$.

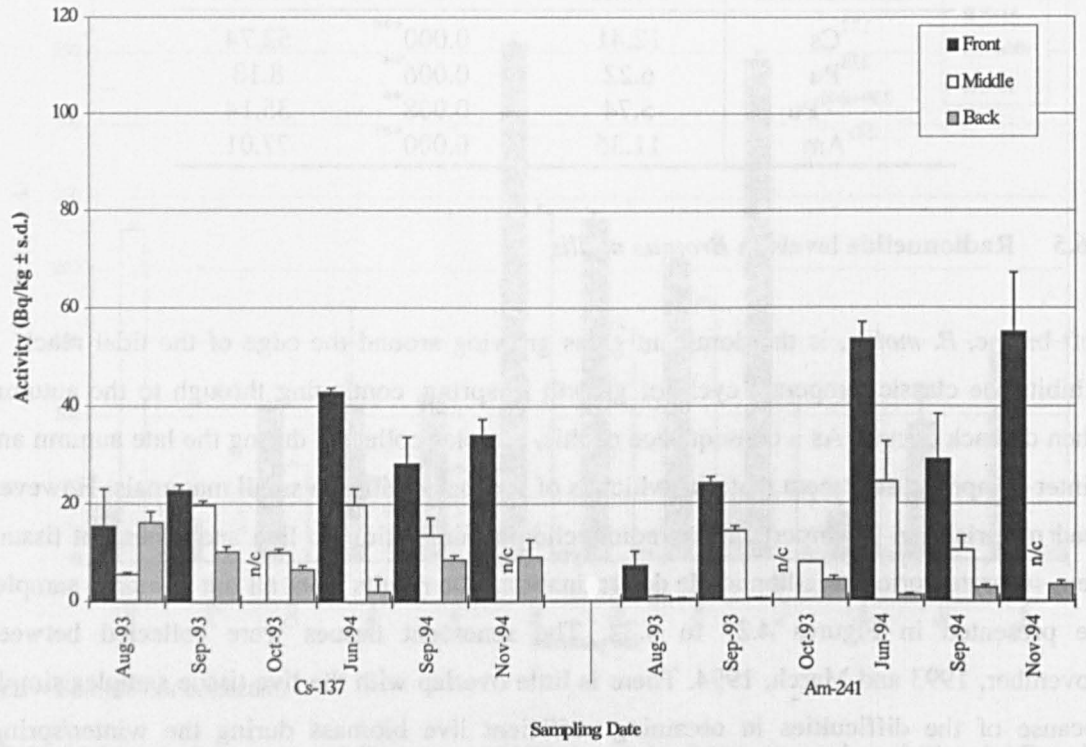
Table 4.14: Summary of One-way ANOVA results for *F. rubra* samples from the River Esk salt marsh. Spatial variation for the seaward side.

Radionuclides	F-ratio	P	5% LSD
^{137}Cs	12.41	0.000^{***}	52.74
^{238}Pu	6.22	0.006^{**}	8.18
$^{239+240}Pu$	5.74	0.008^{**}	35.14
^{241}Am	11.36	0.000^{***}	77.01

4.6.5 Radionuclide levels in *Bromus mollis*

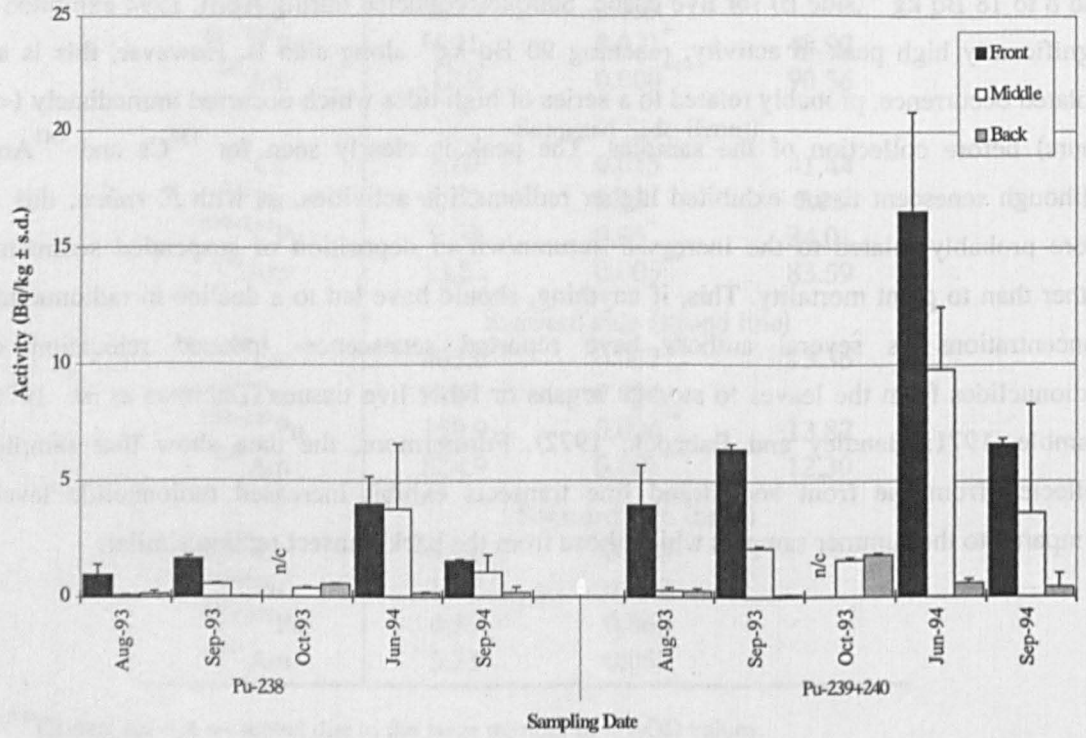
Soft brome, *B. mollis*, is the dominant grass growing around the edge of the tidal reach. It exhibits the classic temperate cycle of growth in spring, continuing through to the autumn, when dieback occurs. As a consequence of this, samples collected during the late autumn and winter comprised senescent material which is of low palatability to small mammals. However, dead material can be important for radionuclide accumulation so live and senescent tissues were separated prior to radionuclide determination. The results from all the *B. mollis* samples are presented in Figures 4.27 to 4.32. The senescent tissues were collected between November, 1993 and March, 1994. There is little overlap with the live tissue samples simply because of the difficulties in obtaining sufficient live biomass during the winter/spring. Figures 4.27 and 4.31, and 4.29 and 4.32 (^{137}Cs and ^{241}Am live and senescent tissue activities) show that the radionuclide levels are higher in the senescent tissue samples on both sides of the marsh; for example, ^{137}Cs levels in senescent tissues along the front transect ranged from 40 to 120 Bq kg^{-1} (side A) and 40 to 75 Bq kg^{-1} (side B) compared to 18 to 41 Bq kg^{-1} (side A) and 8 to 18 Bq kg^{-1} (side B) for live tissue. Samples collected during April, 1994 exhibited a significantly high peak in activity, reaching 90 Bq kg^{-1} along side B. However, this is an isolated occurrence, probably related to a series of high tides which occurred immediately (<6 hours) before collection of the samples. The peak is clearly seen for ^{137}Cs and ^{241}Am. Although senescent tissue exhibited higher radionuclide activities, as with *F. rubra*, this is more probably related to the increased autumn/winter deposition of suspended sediments rather than to plant mortality. This, if anything, should have led to a decline in radionuclide concentrations as several authors have reported senescence- induced relocation of radionuclides from the leaves to storage organs or other live tissues (Dahlman *et al.*, 1975; Gamble, 1971; Handley and Babcock, 1972). Furthermore, the data show that samples collected from the front and strand line transects exhibit increased radionuclide levels compared to the summer samples whilst those from the back transect remain similar.

Figure 4.27: **^{137}Cs and ^{241}Am activities in *B. mollis* (live tissues) collected from the riverward side (A) of the River Esk marsh.**



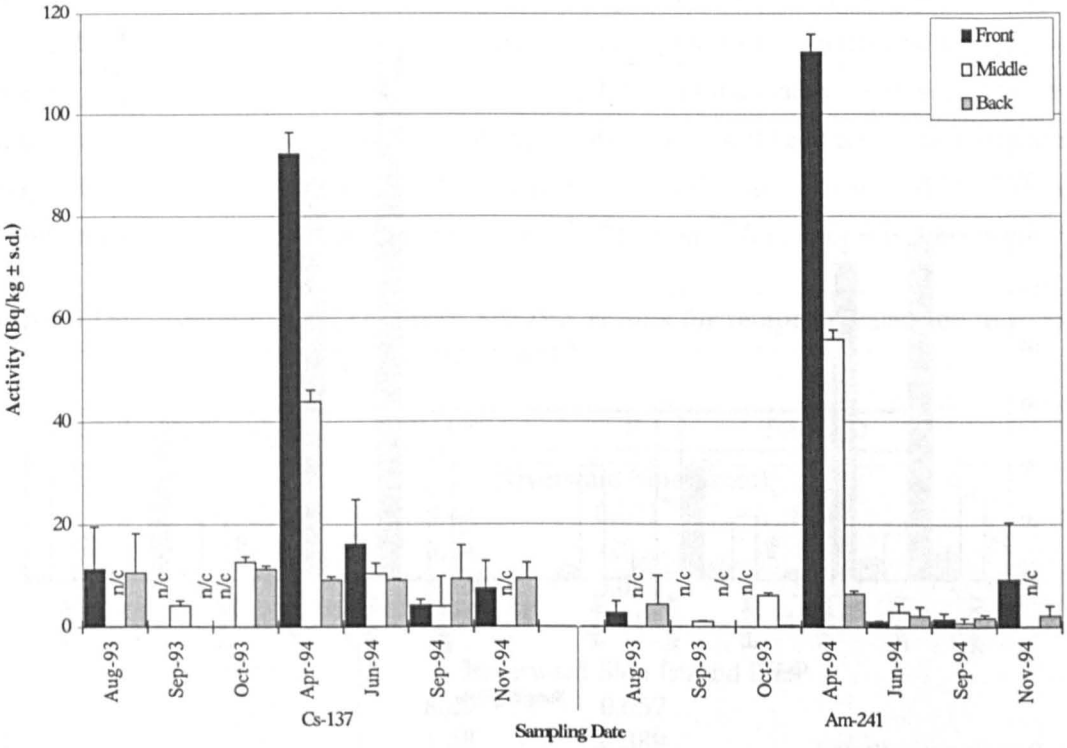
n/c = Sample not collected.

Figure 4.28: **^{238}Pu and $^{239+240}$Pu activities in *B. mollis* (live tissues) collected from the riverward side (A) of the River Esk marsh.**



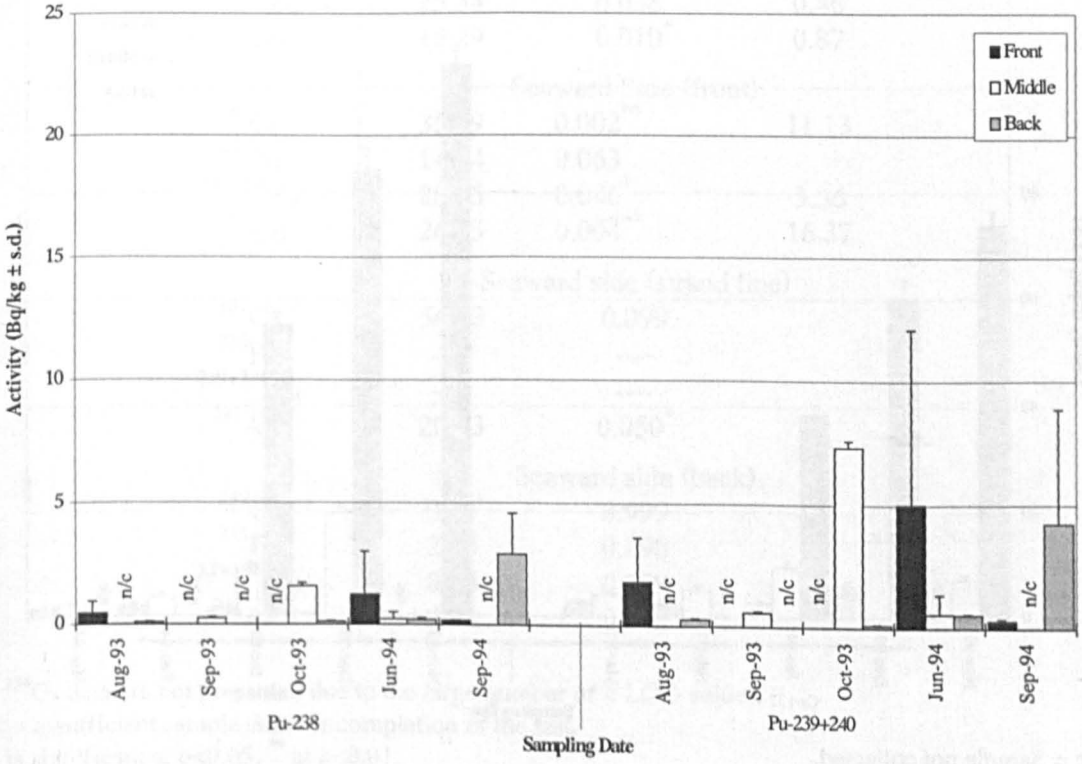
n/c = Sample not collected.

Figure 4.29: **^{137}Cs and ^{241}Am activities in *B. mollis* (live tissues) collected from the seaward side (B) of the River Esk marsh.**



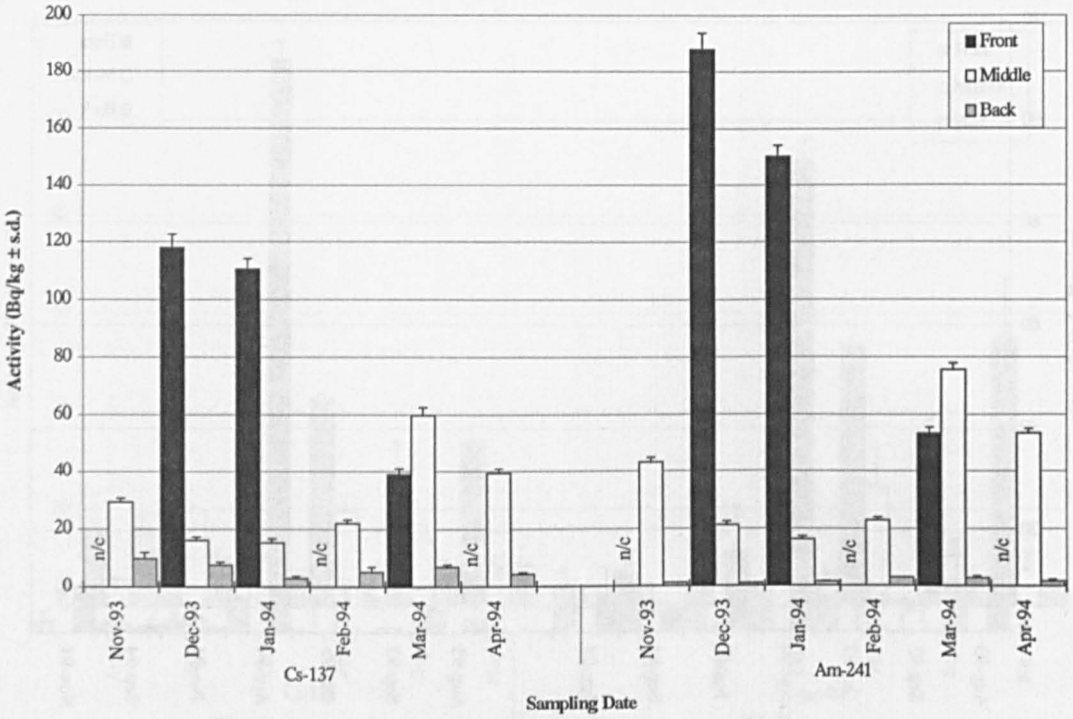
n/c = Sample not collected.

Figure 4.30: **^{238}Pu and $^{239+240}$Pu activities in *B. mollis* (live tissues) collected from the seaward side (B) of the River Esk marsh.**



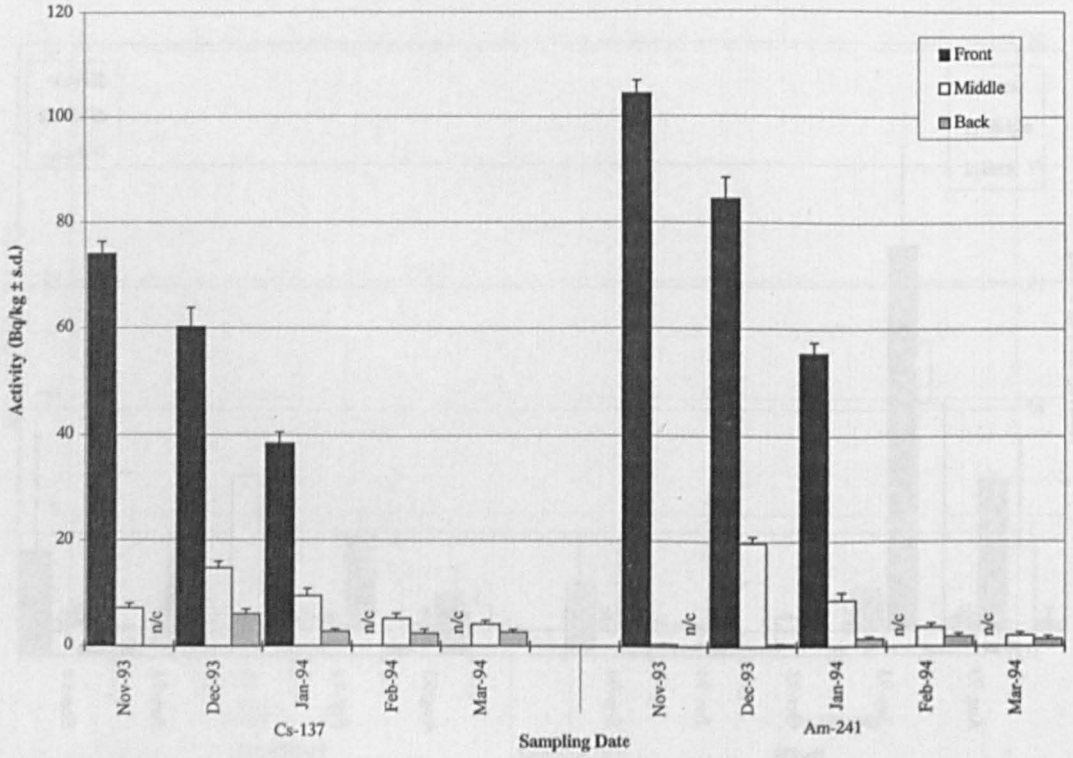
n/c = Sample not collected.

Figure 4.31: **^{137}Cs and ^{241}Am activities in *B. mollis* (senescent tissues) collected from the riverward side (A) of the River Esk marsh.**



n/c = Sample not collected.

Figure 4.32: **^{137}Cs and ^{241}Am activities in *B. mollis* (senescent tissues) collected from the seaward side (B) of the River Esk marsh.**



n/c = Sample not collected.

Data from the live tissue samples were tested for temporal variation between sampling months for the three transects on the two sides of the marsh. A summary of the ANOVA results is presented in Table 4.15 for ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am. The results again show a significant time-based change in activity ($p<0.05$) along the front transect and it is postulated that this is a function of the time elapsed since the last tidal inundation. A less marked change in the radionuclide levels was expected along the strand line and back transects which are less frequently inundated. However, the data from the riverward side also indicate that there are significant differences in the activity levels of $^{239+240}Pu$ and ^{241}Am. This was unexpected and

Table 4.15: Summary of One-way ANOVA results for temporal variation in *B. mollis* from the River Esk salt marsh[a].

Radionuclides	F-ratio	P	5% LSD
	Riverward Side (front)		
^{137}Cs	8.44	0.031*	8.36
^{238}Pu	6.14	0.085	
$^{239+240}Pu$	8.66	0.050*	5.41
^{241}Am	12.77	0.015*	13.33
	Riverward Side (strand line)		
^{137}Cs	8.29	0.057	
^{238}Pu	1.48	0.389	
$^{239+240}Pu$	2.68	0.222	
^{241}Am	5.83	0.090	
	Riverward Side (back)		
^{137}Cs	4.46	0.086	
^{238}Pu	4.34	0.196	
$^{239+240}Pu$	25.34	0.038*	0.46
^{241}Am	15.29	0.010*	0.87
	Seaward Side (front)		
^{137}Cs	35.19	0.002**	11.13
^{238}Pu	14.84	0.063	
$^{239+240}Pu$	20.65	0.046*	3.56
^{241}Am	26.45	0.004**	16.37
	Seaward side (strand line)		
^{137}Cs	56.93	0.099	
^{238}Pu	----	----	
$^{239+240}Pu$	----	----	
^{241}Am	202.3	0.050*	
	Seaward side (back)		
^{137}Cs	0.04	0.999	
^{238}Pu	2.50	0.298	
$^{239+240}Pu$	0.64	0.659	
^{241}Am	0.51	0.762	

[a] ^{134}Cs data are not presented due to the large number of < LOD values.
---- insufficient sample data for completion of the test.
* is significant at $p<0.05$, ** at $p<0.01$.

reflects the high activities recorded during October, 1993 against the low levels recorded for April, 1993. There is no apparent reason for this difference and it is likely to be related to the variation inherent in the sampling procedure.

Table 4.16 presents a summary of the ANOVA examining the spatial variation in activities for *B. mollis* for each transect. Again the test utilised data from live vegetation samples only. The results show significant differences ($p<0.01$) for the riverward side for all nuclides except ^{238}Pu. Interestingly, no differences are evident for the seaward side ($p>0.05$). It is likely that this is a function of the distance separating the three transects. On the riverward side, the study area was larger, accommodating a greater distance between the transects (approximately 8 m). On the seaward side, the transects were located about 5 m apart in the region from which *B. mollis* was sampled. This is different to the seaward area utilised for sampling *F. rubra* and is the most likely explanation for the apparent difference between the two species. A final *t* test was undertaken to determine whether activity levels in *B. mollis* growing on either side of the marsh differed significantly. The results clearly indicate that there are no significant differences ($p>0.05$) when the equivalent transects are compared to each other (Appendix B, Table B6).

Table 4.16: Summary of One-way ANOVA results for *B. mollis* samples from the River Esk salt marsh. Test for spatial variation.

Radionuclides	F-ratio	P	5% LSD
		Riverward Side (A)	
^{137}Cs	17.44	0.000^{***}	7.06
^{238}Pu	2.70	0.093	
$^{239+240}Pu$	5.32	0.010^{**}	4.50
^{241}Am	15.38	0.000^{***}	13.51
		Seaward Side (B)	
^{137}Cs	0.60	0.556	
^{238}Pu	0.11	0.895	
$^{239+240}Pu$	0.21	0.813	
^{241}Am	0.88	0.429	

4.6.6 Radionuclide levels in *Halimione portulacoides* and *Juncus maritimus*

The final set of vegetation samples was to confirm the findings from the 30 m marsh transects and to follow changes in the suspended sediment deposition in a region which was inundated frequently. It was not expected to derive more than an indication of such events and with this in mind the data obtained are presented for *H. portulacoides* and *Juncus maritimus* (Figures 4.33 and 4.34). Clumps of *J. maritimus* are located below the fringes of the tidal reach in areas of standing water on both sides of the marsh. These represent the boundary between the halophytic species and those more characteristic of grassland communities. *H. portulacoides*

Figure 4.33: ^{137}Cs and ^{241}Am activities in samples of *Halimione portulacoides* collected from the River Esk salt marsh.

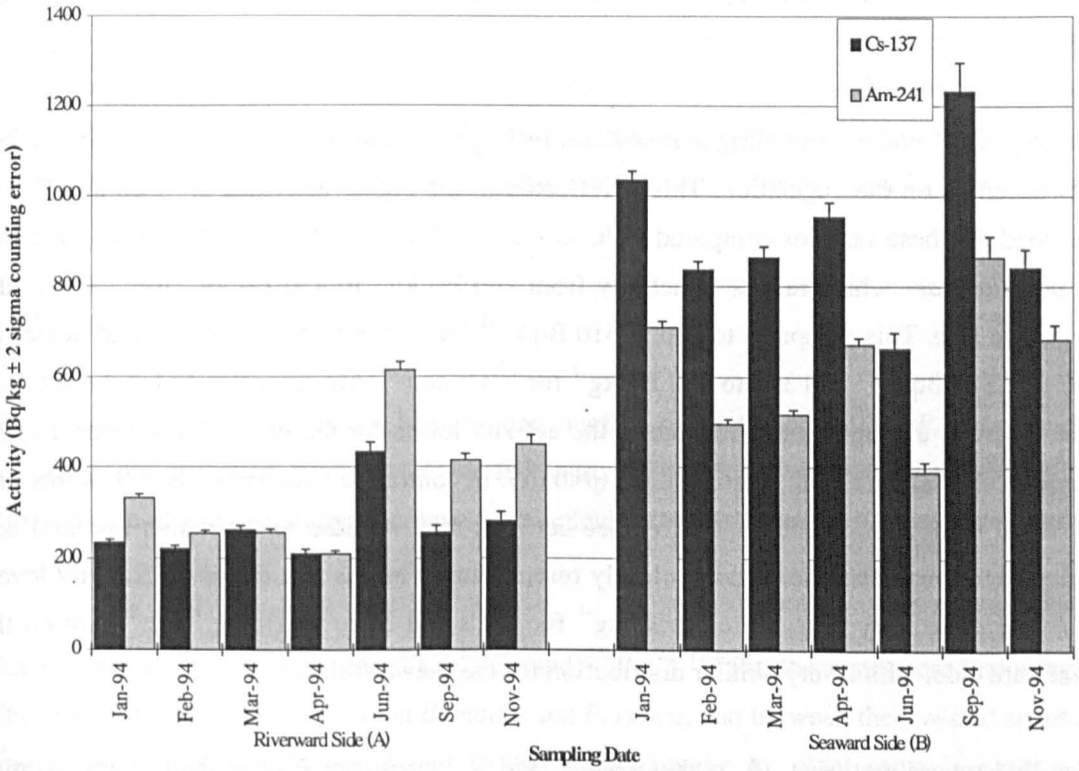
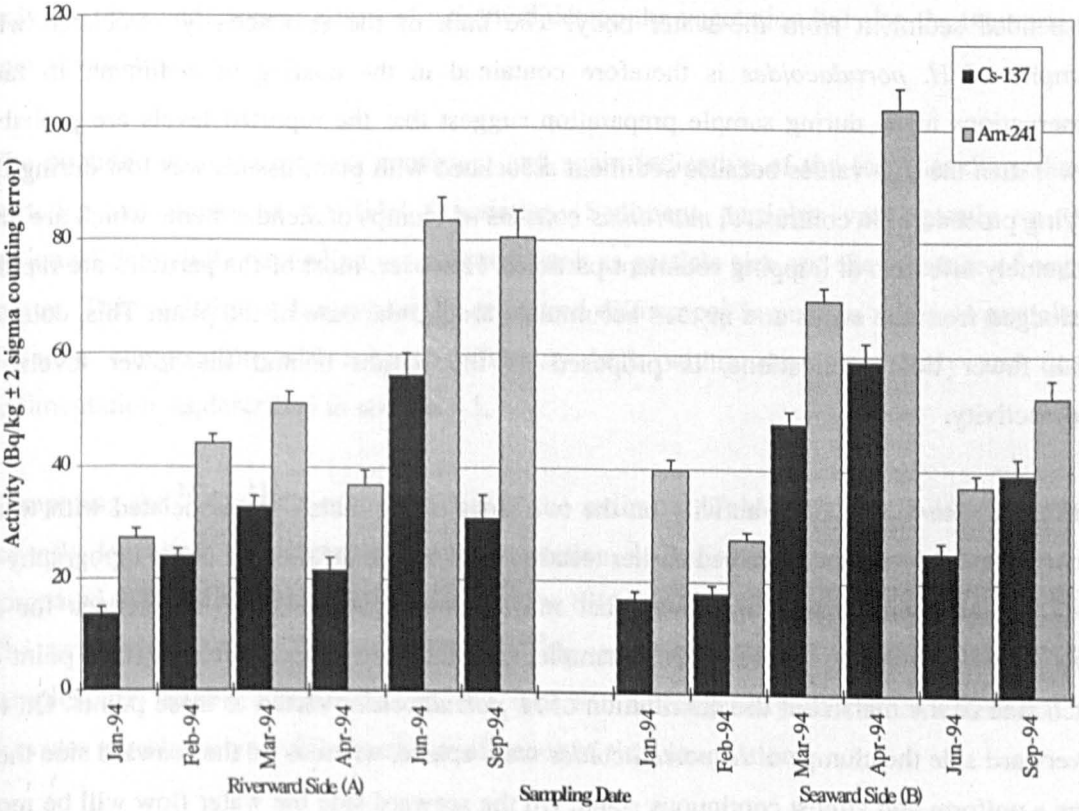


Figure 4.34: ^{137}Cs and ^{241}Am activities in samples of *Juncus maritimus* collected from the River Esk salt marsh.



occurs throughout the main area of both sides of the marsh and its distribution is indicated in Figure 4.1, and Plates 4.1 and 4.2. *H. portulacoides* was sampled approximately 30 m nearer than the front transect (with regard to the river) and comprised both the leaves and woody stem tissue.

In every case, whether sampling *J. maritimus* or *H. portulacoides*, a coating of grey sediment was observed on the vegetation. This is reflected in the higher activities of ^{137}Cs and ^{241}Am recorded for these samples compared to *B. mollis* and *F. rubra*. This was particularly true for *H. portulacoides* which ranges in activity from 200 Bq kg^{-1} to 430 Bq kg^{-1} for ^{137}Cs on the riverward side. This compares to 200 to 610 Bq kg^{-1} for ^{241}Am on the same side and ranges of 650 to 1,240 Bq kg^{-1} and 390 to 850 Bq kg^{-1} for ^{137}Cs and ^{241}Am respectively for the seaward side. There is a significant difference in the activity levels for the two sides in respect of *H. portulacoides* and for both radionuclides ($p<0.01$). In contrast, *J. maritimus*, like *B. mollis* and *F. rubra*, exhibits no significant difference between the two sides of the marsh ($p<0.05$) and has a much lower specific activity, closely resembling *B. mollis* and *F. rubra*. Activity levels in *J. maritimus* vary from 17 to 58 Bq kg^{-1} for ^{137}Cs and 27 to 82 Bq kg^{-1} for ^{241}Am on the riverward side, with a very similar distribution for the seaward side.

The differences between *H. portulacoides* and *J. maritimus* derive from their relative distribution on the marsh and the physical structure of the two species. Thus, the secondary thickening of *H. portulacoides* persists throughout the year and established plants are heavily branched. The leaves are small and form a dense structure that is effective at removing suspended sediment from the water body. The bulk of the radioactivity associated with samples of *H. portulacoides* is therefore contained in the coating of sediment. In fact, observations made during sample preparation suggest that the reported levels are probably lower than the true values because sediment associated with plant tissues was lost during the drying procedure. In contrast, *J. maritimus* consists of clumps of slender stems which are also extremely effective at trapping sediment particles. However, most of the particles are rapidly dislodged from the stems and instead accumulate around the base of the plant. This, coupled with fewer tidal inundations, is proposed as the reason behind the lower levels of radioactivity.

Differences in radionuclide activity on the two sides of the marsh are associated with water flow around the viaduct described earlier (section 4.4). Subtle differences in the topography of the marsh on either side of the viaduct may account for the clear differences for *H. portulacoides* from the two sides. For example, samples were collected from a fixed point on each side of the marsh but the distribution of *H. portulacoides* varied at these points. On the riverward side the clumps of *H. portulacoides* were sparse, whereas on the seaward side there was a uniform and almost continuous stand. On the seaward side the water flow will be more

effectively attenuated leading to higher deposition of the suspended sediment. It was notable that the sediment content of samples collected from the seaward side was visibly greater.

Analysis of the temporal data for *H. portulacoides* shows little evidence of fluctuations throughout the sampling period. Although the data from the riverward side showed significant differences in time for both ^{137}Cs and ^{241}Am ($p<0.05$), this is related to the peak in activity in June, 1994. On the seaward side, the standard deviation in the data masks any otherwise significant differences ($p>0.05$).

4.6.7 Isotopic and Nuclide Ratios

Table 4.17 presents data on the isotopic and nuclide ratios for *B. mollis*, *F. rubra* and *H. portulacoides* collected from the River Esk site. These ratios are averages produced for different months of the study and have been calculated for the strand line transects on both sides of the marsh.

For the isotopic ratios reported, ^{137}Cs:^{134}Cs and ^{238}Pu:$^{239+240}Pu$, the results are encouraging. The similarity in the ratios between *B. mollis* and *F. rubra*, and between the live and senescent tissues of *B. mollis,* provides strong evidence that uptake by plant roots is of minimal importance for this site. The fact that different species exhibit similar ratios strongly suggests that it is sediment particles adhered to plant foliage that override any contribution from internal radionuclides. Consequently, it is thought that tidal inundation is responsible for not only the observed concentrations of radionuclides on the vegetation but also the temporal and spatial variation.

The nuclide ratio data are less consistent and again indicative of the fact that the sediment particles are deposited by tidal inundation. Sediment particles vary greatly in their radionuclide activity depending upon factors such as particle size and the presence of organic matter. This variation will result in the measured differences in nuclide ratios as particles of different composition will behave differently both within the water body and during sedimentation, as described in section 4.1.

Comparing the ^{137}Cs:^{134}Cs ratio with that of the sediment (Table 4.7), it is evident that more recently deposited ^{134}Cs is present on the vegetation. As indicated in section 4.4.1, this change compared to the sediment ratios is related to the differential half-life of ^{134}Cs as against ^{137}Cs. The two ratios, ^{137}Cs:^{134}Cs and ^{238}Pu:$^{239+240}Pu$, measured for the vegetation and flotsam samples are notably similar. Given the inherent heterogeneity in the environment, this provides corroborative evidence of the influence of tidal inundation.

Table 4.17: **Isotopic and nuclide ratios from samples of *B. mollis*, *F. rubra* and *H. portulacoides* collected from the River Esk salt marsh.**

Month	^{137}Cs:^{134}Cs	^{238}Pu:$^{239+240}Pu$	^{137}Cs:$^{239+240}Pu$	^{137}Cs:^{241}Am
RIVERWARD		*B. mollis (live tissue)*		
August '93	15.8	0.32	23.79	7.19
June '94	23.9	0.36	2.07	0.88
September '94	45.8	0.98	26.86	1.32
		B. mollis (senescent tissue)		
January '94	23.51	0.28	2.71	0.94
March '94	79.26	0.27	9.08	0.74
		F. rubra (live tissue)		
August '93	72.7	0.25	6.35	3.17
January '94	140.2	0.22	0.88	0.68
June '94	61.1	0.22	1.65	0.83
September '94	125.6	0.25	1.26	0.65
		H. portulacoides		
January '94	232.5	0.24	1.79	0.70
April '94	168.2	-----	-----	0.99
September '94	90.9	-----	-----	0.61
SEAWARD		*B. mollis (live tissue)*		
August '93*	31.14	0.21	45.54	4.30
June '94	17.27	0.36	28.36	3.46
September '94*	14.59	0.60	24.48	3.10
		B. mollis (senescent tissue)		
January '94	15.1	0.73	1.49	1.08
March '94	12.4	0.42	8.27	1.94
		F. rubra (live tissue)		
August '93	155.7	0.25	3.73	1.23
January '94	189.3	0.25	2.22	0.58
June '94	51.2	0.23	1.99	1.18
September '94	12.2	0.23	2.90	0.80
		H. portulacoides		
January '94	109.0	0.24	2.65	1.46
April '94	127.3	-----	-----	1.41
September '94	97.4	-----	-----	1.41

* Front transect data used for the calculations.
----- = sample either not collected or not analysed.
Calculations made from strand line transects.

The ^{137}Cs:$^{239+240}Pu$ ratios calculated for live *B. mollis* are unexpectedly higher than both measurements made for *F. rubra*, strand line material and sediments. Also, because the ratios are consistently higher over time and on both sides of the marsh, this may be is a real difference in radionuclide deposition and not just an artefact of sampling. Possible explanations for this elevation are that ^{137}Cs is preferentially taken up by *B. mollis* or the

surface structure of *B. mollis* somehow influences the sediment deposition. More detailed analysis is required to confirm which of these is the more plausible.

4.6.8 Concentration Ratios for *Bromus mollis* and *Festuca rubra*

Table 4.18 presents the concentration ratios calculated using the formula: vegetation activity (Bq kg^{-1} dry weight)/activity in sediment (Bq kg^{-1} dry weight). The values show fluctuations over time for both species which may reflect changes in surface contamination of the plant foliage as determined by the time elapsed since the last tidal inundation. It is notable that although the levels are temporally similar for the plant species on both sides of the marsh, there is an indication that *F. rubra* has a higher concentration ratio. However, for the present data set this is insignificant ($p>0.05$). Spatial differences in the concentrations ratios analysed were also insignificant ($p>0.05$). This was expected given that the radionuclide activities in the vegetation and sediment samples both declined towards the back of the marsh.

The concentration ratios for *B. mollis* and *F. rubra* fall within the range quoted by Bunzl and Kracke (1987) for cereal crops grown in soil contaminated by fall-out from global weapons testing where root uptake dominated the transfer mechanism (section 3.6). However, for the

Table 4.18: Concentration ratios for samples of *B. mollis*, *F. rubra* collected from the River Esk salt marsh.

Month	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
RIVERWARD		*B. mollis (live tissue)*		
August '93	9.86E-04	2.30E-04	1.68E-04	2.51E-04
June '94	3.49E-03	1.08E-02	6.21E-03	7.99E-03
September '94	1.99E-03	3.61E-03	2.13E-03	3.03E-03
		F. rubra (live tissue)		
August '93	4.80E-03	7.89E-03	7.48E-03	8.71E-03
January '94	3.11E-02	1.26E-01	1.28E-01	8.20E-02
June '94	1.54E-02	3.43E-02	3.39E-02	3.36E-02
September '94	1.38E-02	6.49E-02	4.67E-02	4.26E-02
SEAWARD		*B. mollis (live tissue)*		
August '93*	2.33E-03	1.36E-03	2.42E-03	1.67E-03
June '94	1.33E-03	1.25E-03	7.74E-04	1.11E-03
September '94*	8.10E-04	4.49E-04	1.52E-04	3.86E-04
		F. rubra (live tissue)		
August '93	2.60E-02	6.25E-02	6.04E-02	8.46E-02
January '94	2.17E-02	6.48E-02	5.84E-02	8.89E-02
June '94	6.89E-03	1.70E-02	1.49E-02	2.01E-02
September '94	4.42E-04	1.16E-02	3.64E-03	4.38E-04

* Front transect used for the calculations.
Calculations made from strand line transects.

River Esk salt marsh a very low root uptake rate was expected because of the strong affinity between radionuclides and sediment particles (section 1.4.1). In addition, the quantity of foliar-associated sediment will vary temporally being dependent upon the time elapsed since the last inundation. Originally, it was anticipated that the foliar-associated sediment would artificially raise the concentration ratios. The results indicate that this is less important than expected though the ratios for the four radionuclides, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am are all very similar. This suggests that the bioavailability of ^{137}Cs is lower than in agricultural soils and is a further indication of the strong association with the sediment particles. Moreover, the similarity of the concentration ratios of the actinides with those of ^{137}Cs provides collateral evidence of the differences in the behaviour of ^{137}Cs and the actinides in a water body.

These concentration ratios are higher than those given for the vegetation from the upper zone of this salt marsh as measured by Livens *et al.* (1994) who reported values of 0.00015 and 0.00017 for $^{239+240}Pu$ and ^{241}Am respectively. However, these were based on samples of vegetation as they were grown in isolation on collected sediment material that never experienced a tidal inundation. Livens *et al.* (1994) also provide further evidence of the low plant uptake of actinides expected from the sediment, by reporting a concentration range between 0.38 and 2.29 and 1.09 to 3.32 Bq kg^{-1} for $^{239+240}Pu$ and ^{241}Am respectively. This compares to the levels of 30 to 40 Bq kg^{-1} in the present study, again elevated by the presence of foliar-associated sediment.

4.6.9 Comparison of Vegetation Collected at Reference Sites to those from the River Esk

Table 4.19 compares data for vegetation samples collected from two reference sites, Ness Gardens, Wirral and Llandridian Marsh, Wales (section 4.2), with results from the River Esk during August, 1993 and September, 1994. Simple visual inspection of the data indicates that there are differences between the River Esk and reference sites. This was confirmed by One-way ANOVA, the results of which are summarised in Table 4.20. These, and the calculated LSD values, show that the differences are significant ($p<0.01$) with the exception of *B. mollis* samples collected from the seaward side.

The results from the back transects are similar to the reference site levels (Table 4.19) and are accounted for by their location, approximately 10 m above the normal limit of tidal reach. Although the sediment samples along the back transects contain considerable activity, limited root uptake and infrequent tidal inundation give rise to the low levels of radionuclides recorded in the vegetation samples. It is notable that the actinide data for *F. rubra* differ from *B. mollis*, in that they are generally between 5 and 10 times greater. This could arise from

Table 4.19: **^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am mean activities (Bq kg^{-1} ± s.d.) in vegetation samples collected from two reference sites compared to the River Esk.**

	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
River Esk (A)		*F. rubra*		
August 1993 Middle	27.3 ± 22.9	2.7 ± 3.5	11.8 ± 15.2	27.4 ± 36.0
September 1994 Middle	94.87 ± 6.4	19.0 ± 3.4	79.1 ± 26.7	146.3 ± 4.2
		B. mollis		
August 1993 Front	15.5 ± 7.6	1.0 ± 0.4	3.9 ± 1.7	7.2 ± 2.9
August 1993 Middle	5.6 ± 0.4	0.1 ± 0.03	0.3 ± 0.1	0.8 ± 0.1
August 1993 Back	16.2 ± 2.2	0.01 ± 0.01	0.3 ± 0.1	0.4 ± 0.1
September 1994 Front	28.3 ± 5.5	1.6 ± 0.1	6.6 ± 0.2	29.1 ± 9.2
September 1994 Middle	13.7 ± 3.5	1.1 ± 0.7	3.6 ± 4.7	10.4 ± 3.0
September 1994 Back	8.5 ± 1.0	0.2 ± 0.2	0.4 ± 0.6	2.7 ± 0.5
River Esk (B)		*F. rubra*		
August 1993 Front	41.7 ± 1.8	3.2 ± 1.9	12.9 ± 6.9	36.8 ± 15.0
August 1993 Middle	199.3 ± 4.5	20.5 ± 0.5	87.7 ± 1.0	227.4 ± 4.3
August 1993 Back	8.4 ± 0.9	0.4 ± 0.1	1.8 ± 0.2	0.4 ± 0.1
September 1994 Front	75.0 ± 17.3	6.0 ± 0.2	25.9 ± 0.4	94.0 ± 26.7
September 1994 Middle	4.4 ± 1.3	4.4 ± 4.7	7.2 ± 4.7	1.6 ± 0.5
September 1994 Back	6.9 ± 0.5	1.4 ± 0.8	4.3 ± 4.6	0.8 ± 0.1
		B. mollis		
August 1993 Front	11.2 ± 8.4	0.5 ± 0.5	1.8 ± 1.8	2.8 ± 2.3
August 1993 Middle	-----	-----	-----	-----
August 1993 Back	10.3 ± 7.9	0.1 ± 0.1	0.2 ± 0.1	4.4 ± 5.5
September 1994 Front	4.2 ± 1.1	0.2 ± 0.0	0.3 ± 0.1	1.5 ± 1.1
September 1994 Middle	4.1 ± 5.7	-----	-----	0.7 ± 1.0
September 1994 Back	9.2 ± 6.7	2.9 ± 1.7	4.3 ± 4.6	1.7 ± 0.6
Ness Gardens				
August 1993	<0.97	0.09 ± 0.05	0.04 ± 0.00	0.10 ± 0.08
July 1994	<0.38	0.05 ± 0.01	0.07 ± 0.02	<0.38
Llandridian Marsh				
August 1994	2.83 ± 3.42	0.05 ± 0.00	0.05 ± 0.01	0.07 ± 0.02

----- Sample not collected.

differences in the uptake of the actinides, their sedimentation and subsequent retention on plant foliage. It is speculated that the latter is more important because the two grass species differ in their structure and growth form. *F. rubra*, for example, is fine leaved and grows in a tight sward which entraps any deposited sediment whereas *B. mollis* is taller and grows more sparsely.

Table 4.20: **Summary of ANOVA tests which compare vegetation samples from the River Esk and the Cheshire and Welsh reference sites.**

	Riverward Side (A)			*Seaward Side (B)*		
	F ratio	*p*	*5% LSD*	*F ratio*	*p*	*5% LSD*
F. rubra						
^{137}Cs	27.71	0.001***	15.72	129.16	0.000***	7.84
^{238}Pu	36.33	0.000***	2.76	15.18	0.000***	2.24
$^{239+240}Pu$	15.49	0.003**	17.76	83.37	0.000***	4.19
^{241}Am	38.12	0.000***	20.92	61.57	0.000***	13.38
B. mollis						
^{137}Cs	15.16	0.000***	3.60	1.60	0.262	
^{238}Pu	7.33	0.004**	0.34	5.90	0.013*	0.73
$^{239+240}Pu$	4.04	0.026*	1.90	1.54	0.278	
^{241}Am	17.96	0.000***	3.37	1.02	0.484	

4.6.10 Summary of Vegetation Results

Activity levels in vegetation showed significant temporal and spatial variation within and between the three sampling transects on the lower, middle and upper marsh. This mainly reflected the intensity and periodicity of tidal inundation. Thus, live plant material, mainly *B. mollis*, ranged from 10 to 80 Bq kg^{-1} (^{137}Cs), 2 to 21 Bq kg^{-1} ($^{239+240}Pu$) and 3 to 54 Bq kg^{-1} (^{241}Am). Senescent material showed higher and even more variable datasets. Vegetation and sediment activity patterns showed a close relationship, with all four radionuclides behaving in a similar manner.

Activity transfer from sediment to vegetation was low, due to strong adsorption co-efficients and consequently low plant uptake. In the complete absence or presence of only occasional tidal inundations, for example within the rearward sections of the marsh, activity levels in vegetation were very low.

F. rubra showed considerable temporal variation in activity levels, but with consistently low values at <100 Bq kg^{-1} for ^{137}Cs, $^{239+240}Pu$ and ^{241}Am. Values were higher in winter than in summer, probably due to tidal patterns and the impact of growth dilution. Predictable peaks in activity occurred along the frontal and strand line transects where, for example, levels of ^{137}Cs were, at 96 and 78 Bq kg^{-1} respectively, an order of magnitude higher than for the landward (rear) transect. ^{137}Cs levels in senescent samples collected in late autumn and winter were, at 40 to 120 Bq kg^{-1} and 40 to 75 Bq kg^{-1} for the seaward and riverward sectors of the marsh, much higher than values for live tissue, at 18 to 41 and 8 to 18 Bq kg^{-1}, for the seaward and riverward sectors respectively.

Live samples of *J. maritimus* and *H. portulacoides* showed consistently higher activities than *B. mollis* and *F. rubra*, a reflection of their habitat and the light coating of grey sediment associated with more frequent inundation. Further inter-specific differences occurred in the same community between *J. maritimus* and *H. portulacoides*, with the latter species showing higher activity levels due to its densely branched and compact structure which acts as a more effective physical scavenger of sediment than the upright slender leaves of the rush species.

Isotopic and nuclide ratios gave strong evidence in support of minimal root absorption as a component of vegetation activity levels, with sediment deposition being of over-riding importance. Concentration ratios for the four nuclides of greatest interest suggest a lower bioavailability of sediment borne ^{137}Cs than in the case of agricultural soils.

4.7 INVERTEBRATE RESULTS

Table 4.21 lists the major invertebrate orders and principal species caught using pitfall traps set either side of the railway viaduct over the River Esk salt marsh. Table 4.22 shows that the same taxonomic groups were caught either side of the marsh and provides information on the seasonal abundance and biomass of each group/species. The groupings used for analysis purposes are indicated in the two tables.

The seasonal abundance of invertebrates will be reflected in the diet of the field mouse, *Apodemus sylvaticus*, and the common shrew, *Sorex araneus* (section 1.4.4.5). Consideration of the seasonal changes in radionuclide activities in invertebrates and in the mammalian diet is therefore required. It has already been indicated (section 4.3) that the low biomass obtained from the pitfall traps necessitated the pooling of samples from the pitfalls over a four month period, and for longer over the winter. The 18 pitfalls exposed on each side of the railway viaduct were pooled to produce the sample mass required for radionuclide determination. The sample groupings are shown in Table 4.22. In addition, the pitfalls exposed at the River Esk site were located around the strand line (Figure 4.1) and the contents were frequently lost because of unexpected tidal inundation. Using tide prediction tables (Laver, 1993 to 1995) the pitfalls were removed and reset around the high tides. However, local climatic conditions influence the tidal heights and they could not be predicted with accuracy.

By observation, invertebrates, particularly *Orchestia* spp., were most active amongst the strand line material which forms a significant component of the salt marsh ecosystem (section 4.5). This bio-activity was reflected in the pitfall capture rates, with *Orchestia* spp. forming between 55 and 70% of the total biomass collected over the four sampling periods. *Orchestia* spp. are omnivorous scavengers that are particularly active at night (Fish and Fish, 1989). These probably form the bulk of the *S. araneus* diet since they are so abundant throughout the year.

Table 4.21: The major invertebrate orders and species caught in pitfall traps at the River Esk salt marsh.

CLASS/ SUBCLASS	*ORDER*	*FAMILY*	*GENUS/SPECIES*
Arachnida	**Araneida**		
	Opiliones*		
Crustacea	Amphipoda		***Orchestia* spp.**
	Decapoda*		*Carcinus maenas*
	Isopoda		*Oniscus asellus*
			Philoscia muscorum
Diplopoda*			
Chilopoda*			
Gastropoda*			
Insecta	Collembola*		
	Coleoptera	**Carabidae**	*Amara aulica*
			Carabus violaceous
			Feronia nigrita
			Leistus spinibarbis
		Curculionidae	
		Elateridae	
		Scarabaeidae	
		Staphylinidae	
	Diptera		
	Dermaptera		*Forficula auricularia**
	Hymenoptera	**Formicidae**	
Oligochaeta*			*Lumbricus spp.*

Those groups highlighted in bold represent pooled units used for analysis; where no species are listed or are not highlighted, then the species were bulked into the higher Family or Order.
Adult and larval forms of Coleoptera were analysed separately.
* Very small masses (<0.2 g dry weight) were collected and not analysed.

Table 4.22: Seasonal abundance and biomass (g dry weight) of the invertebrates analysed from River Esk.

Sample Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August to December '94*
Araneida	6.50	0.74	1.22	1.06
Orchestia spp.	86.41	35.59	26.12	21.75
Isopoda	13.26	8.44	2.53	2.52
Coleoptera (adults)	8.94	5.92	2.23	-----
Coleoptera (larvae)	0.85	0.13	0.07	0.88
Carabidae	17.03	0.88	3.21	1.57
Amara aulica	9.60	0.68	0.36	3.75
Feronia nigrita	5.66	1.99	-----	-----
Diptera*	0.20	0.09	0.25	0.02
Formicidae*	0.67	0.09	0.01	0.02

* Not all samples analysed.
Sample masses combined from both sides.

Of the other invertebrate groups, the Isopoda, Araneida and Carabidae were present throughout the year in sufficient numbers for analysis, although the biomass tended to decline during the winter. The diversity of Diptera was disappointing. The low biomass collected is surprising given the number of adult flies frequenting the decomposing strand material and the assumed presence of the active larval stages. The low mass prevented successful radionuclide determination of all four samples.

4.7.1 ^{137}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am Activity in Invertebrates

Radionuclide activities were recorded on a whole body basis although invertebrates were cleaned of external particulates prior to separation into taxonomic groups. This rinsing also removed the external formalin from body surfaces. The results for ^{137}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am in invertebrates are presented in Tables 4.23 to 4.26. It was not possible to detect and quantify ^{134}Cs within the counting time allocated. Only those samples which were caught frequently over the four sampling periods were analysed for plutonium isotopes.

There is considerable variation between the different taxonomic groups. For example, during the first four month period ^{137}Cs values ranged from 8.8 Bq kg^{-1} in *Feronia nigrita* to 197.4 Bq kg^{-1} in Isopoda, in samples taken from the riverward side. This variation was also recorded for the actinides; 3.1 Bq kg^{-1} to 134 Bq kg^{-1} for ^{238}Pu (Carabidae to Formicidae), 8.8 to 78 Bq kg^{-1} for $^{239+240}$Pu (Carabidae to Isopoda) and 4.1 to 93.8 Bq kg^{-1} for ^{241}Am (*F. nigrita* to Isopoda).

The detritivorous fauna were represented by two groups, Isopoda and *Orchestia* spp. It can be seen that these two groups, Isopoda particularly, exhibit some of the highest radionuclide activities although there are seasonal changes, such as for in ^{137}Cs which reflect radionuclide bioavailability at different times of the year. Figure 4.35 presents the temporal changes in Isopoda and *Orchestia* spp Both Isopoda and *Orchestia* spp. were abundant within the strand line material wherein lies the principal source of food. The strand line material is an important link in the ecosystem and is the primary pathway for the entry of radionuclides into the food chain. It is notable that the actinide data in detritivores remained reasonably consistent throughout the four sampling periods.

Of the predatory invertebrates, Araneida exhibited the highest concentrations of each radionuclide, especially when compared to the predatory ground beetles, Carabidae. This is similar to the results obtained from Lady Wood and is related to the different feeding strategies described in section 3.7.1.

Table 4.23: ^{137}Cs activity (Bq kg^{-1} ± 2σ counting error) in invertebrates from River Esk salt marsh.

Sample Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August to December '94*
		Riverward Side (A)		
Araneida	95.2 ± 19.3	111.9 ± 47.9	44.8 ± 13.0	9.1 ± 3.7
Orchestia spp.	54.1 ± 3.8	104.4 ± 4.8	26.6 ± 3.4	83.3 ± 4.9
Isopoda	197.4 ± 9.6	58.8 ± 10.1	108.0 ± 6.4	32.2 ± 14.2
Coleoptera (adults)	38.8 ± 5.9	21.0 ± 3.7	-----	-----
Coleoptera (larvae)	173.6 ± 31.4	295.7 ± 62.3	-----	-----
Carabidae	26.5 ± 3.2	25.3 ± 8.4	20.6 ± 5.7	-----
Amara aulica	-----	22.1 ± 10.4	-----	-----
Feronia nigrita	8.8 ± 1.7	-----	-----	-----
Diptera	-----	-----	-----	-----
Formicidae	26.6 ± 12.8	-----	-----	-----
		Seaward Side (B)		
Araneida	54.3 ± 8.9	35.7 ± 6.0	142.7 ± 22.4	52.1 ± 14.2
Orchestia spp.	41.5 ± 4.5	93.3 ± 9.4	29.0 ± 4.6	31.4 ± 4.3
Isopoda	39.1 ± 3.2	47.0 ± 9.6	142.0 ± 9.5	44.8 ± 8.5
Coleoptera (adults)	15.2 ± 3.7	38.4 ± 4.6	25.8 ± 8.6	-----
Coleoptera (larvae)	36.3 ± 10.7	-----	-----	56.5 ± 11.3
Carabidae	15.5 ± 2.9	-----	34.5 ± 8.0	23.6 ± 10.7
Amara aulica	3.0 ± 2.0	-----	72.0 ± 16.4	13.9 ± 2.3
Feronia nigrita	-----	-----	-----	-----
Diptera*	201.3 ± 65.4	**597.4 ± 202.5**	169.1 ± 44.8	-----
Formicidae*	-----	**615.7 ± 246.2**	-----	-----

----- Sample not collected.
* Data in bold has been calculated from samples of small mass (<0.3 g dry weight).

Beetles feed upon prey as a whole. This is particularly important for ^{137}Cs which accumulates within soft tissues such as muscle and organs but it does not explain the elevated actinide levels observed within the data set. Of the Carabidae, *A. aulica* and *F. nigrita* were abundant across the salt marsh; but they were not always caught in sufficient numbers to permit analysis for each of the four sampling periods. The Araneida were 3 to 5 times higher in ^{137}Cs than the Carabidae. The temporal variation for these groups is presented in Figures 4.36 and 4.37. The data show that the actinide concentrations remained reasonably constant over time, whereas the ^{137}Cs values fluctuated. This probably relates indirectly to fluctuations in ^{137}Cs concentrations in the strand material. For both Araneida and Carabidae, it is probable that the principal prey consists of *Orchestia* spp. and Isopoda simply on the basis of abundance. However, Carabidae will also feed on smaller Coleoptera and other invertebrates (Chinery, 1993; section 3.7.1). Given that *Orchestia* spp. and Isopoda form the basis of the invertebrate food chains, elevated concentrations of both caesium and the actinides would be expected in the predators given bio-concentration in the predator-prey relationship.

Table 4.24: **^{238}Pu activity (Bq kg^{-1} ± 2σ counting error) in invertebrates from River Esk salt marsh.**

Sample Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August to December '94*
		Riverward Side (A)		
Araneida	6.8 ± 1.0	31.9 ± 4.1	10.1 ± 2.0	2.0 ± 0.5
Orchestia spp.	10.9 ± 0.2	28.5 ± 6.9	4.9 ± 0.3	16.7 ± 0.5
Isopoda	16.2 ± 0.6	16.5 ± 0.7	19.1 ± 1.2	6.2 ± 1.0
Coleoptera (adults)	32.4 ± 1.3	5.9 ± 1.1	-----	-----
Coleoptera (larvae)	16.0 ± 2.5	n/a	-----	-----
Carabidae	3.1 ± 0.3	1.3 ± 0.3	0.9 ± 0.2	-----
Amara aulica	-----	<0.6	-----	-----
Feronia nigrita	n/a	-----	-----	-----
Diptera	-----	-----	-----	-----
Formicidae	138.8 ± 7.6	-----	-----	-----
		Seaward Side (B)		
Araneida	2.7 ± 0.4	15.7 ± 2.4	10.4 ± 2.1	8.5 ± 1.2
Orchestia spp.	3.9 ± 0.2	8.4 ± 0.4	5.0 ± 0.3	3.6 ± 0.2
Isopoda	7.3 ± 0.3	5.1 ± 0.4	3.9 ± 0.8	5.4 ± 0.9
Coleoptera (adults)	1.8 ± 0.2	4.5 ± 0.5	4.1 ± 0.5	-----
Coleoptera (larvae)	13.7 ± 2.0	-----	-----	n/a
Carabidae	1.8 ± 0.2	-----	5.3 ± 0.7	3.9 ± 0.5
Amara aulica	0.8 ± 0.1	-----	12.0 ± 1.5	2.2 ± 0.3
Feronia nigrita	-----	-----	-----	-----
Diptera	n/a	n/a	n/a	-----
Formicidae	-----	n/a	-----	-----

----- Sample not collected.
n/a Sample not analysed

The Coleopteran larvae consisted of predatory larvae belonging to the Carabidae. These actively seek and consume detritivores such as Isopoda. They do not burrow, and forage almost exclusively within the strand material. This would explain the high levels of both caesium and actinides in larvae when compared to the adult Carabids.

Coleopteran larvae were only caught in two of the sampling periods, but it is notable that the activity levels increased during the winter period of year one. This is consistent with the greatest increase in the deposition of radionuclides via both tidal inundation and strand line material.

A notable point is that with the exception of *Orchestia* spp., the invertebrate taxonomic groups are typical of terrestrial habitats and do not include species representative of aquatic environments or the interface between the two. This is surprising given that tidal inundations over the marsh do occur, albeit infrequently. It has been reported that only 25% of fauna in the

Table 4.25: $^{239+240}$Pu activity (Bq kg^{-1} ± 2σ counting error) in invertebrates from River Esk salt marsh.

Sample Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August to December '94*
		Riverward Side (A)		
Araneida	32.0 ± 2.3	22.6 ± 3.5	17.0 ± 2.6	4.4 ± 0.8
Orchestia spp.	51.7 ± 0.5	46.4 ± 8.8	21.7 ± 0.7	55.5 ± 0.8
Isopoda	78.0 ± 1.3	72.3 ± 1.4	47.7 ± 2.0	30.7 ± 2.3
Coleoptera (adults)	11.7 ± 0.8	6.2 ± 1.2	-----	-----
Coleoptera (larvae)	30.3 ± 3.4	n/a	-----	-----
Carabidae	8.8 ± 0.6	5.1 ± 0.6	3.3 ± 0.4	-----
Amara aulica	-----	4.6 ± 0.6	-----	-----
Feronia nigrita	n/a	-----	-----	-----
Diptera	-----	-----	-----	-----
Formicidae	21.1 ± 3.0	-----	-----	-----
		Seaward Side (B)		
Araneida	14.3 ± 0.9	11.3 ± 2.0	32.6 ± 3.7	6.5 ± 1.1
Orchestia spp.	15.2 ± 0.4	37.4 ± 0.9	18.5 ± 0.5	15.3 ± 0.3
Isopoda	28.9 ± 0.6	25.3 ± 0.8	10.5 ± 1.3	24.4 ± 1.9
Coleoptera (adults)	4.0 ± 0.3	12.3 ± 0.7	4.1 ± 0.5	-----
Coleoptera (larvae)	7.7 ± 1.5	-----	-----	n/a
Carabidae	5.1 ± 0.3	-----	4.0 ± 0.6	4.5 ± 0.6
Amara aulica	1.5 ± 0.1	-----	4.1 ± 0.9	4.2 ± 0.4
Feronia nigrita	-----	-----	-----	-----
Diptera	n/a	n/a	n/a	-----
Formicidae	-----	n/a	-----	-----

----- Sample not collected.
n/a Sample not analysed

upper salt marsh zone occurs only in to such habitats and that many terrestrial arthropods are able to survive immersion by seawater (Heydemann, 1979).

The Coleopteran adults consisted of herbivorous weevils and beetles which are typical of grassland communities. The results indicate similar levels of caesium and actinides ($^{239+240}$Pu and ^{241}Am) in the two groups, concentrations being between 20 Bq kg^{-1} and 40 Bq kg^{-1} respectively. Levels of ^{238}Pu were approximately half this. These values are greater than for the predatory beetles and again reflect differences in feeding strategy.

The results for Diptera and Formicidae are sparse and of marginal importance. However, the Diptera activity levels are similar or greater than those for the detritivores. For example, ^{137}Cs levels range from 200 Bq kg^{-1} to nearly 600 Bq kg^{-1}, and the ^{241}Am value is also around 600 Bq kg^{-1}. As indicated in Tables 4.22 and 4.23 to 4.26, the biomass for radionuclide determination was very small. Caution should therefore be exercised when interpreting the results. The high levels reported are nevertheless surprising and unexpected. Individuals from

Table 4.26: **^{241}Am activity (Bq kg^{-1} ± 2σ counting error) in invertebrates from River Esk salt marsh.**

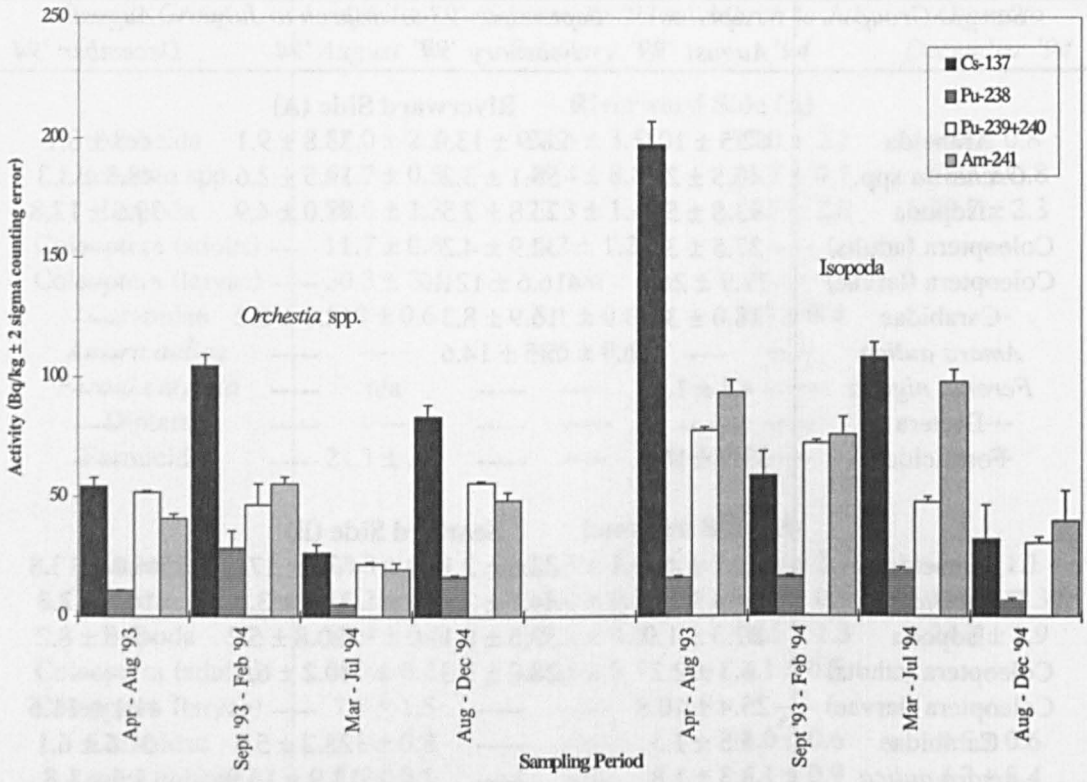
Sample Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August to December '94*
		Riverward Side (A)		
Araneida	62.5 ± 10.9	26.9 ± 13.0	33.8 ± 9.1	6.3 ± 3.1
Orchestia spp.	40.5 ± 2.2	55.1 ± 3.2	19.5 ± 2.6	48.3 ± 3.3
Isopoda	93.8 ± 5.2	75.8 ± 7.5	98.0 ± 4.9	39.5 ± 12.8
Coleoptera (adults)	27.5 ± 3.7	32.9 ± 4.2	-----	-----
Coleoptera (larvae)	77.9 ± 26.2	416.6 ± 121.0	-----	-----
Carabidae	18.0 ± 3.0	16.9 ± 8.3	12.0 ± 5.2	-----
Amara aulica	-----	5.5 ± 14.6	-----	-----
Feronia nigrita	4.1 ± 1.9	-----	-----	-----
Diptera	-----	-----	-----	-----
Formicidae	33.7 ± 10.3	-----	-----	-----
		Seaward Side (B)		
Araneida	22.7 ± 4.4	22.6 ± 2.1	47.4 ± 17.5	46.0 ± 13.8
Orchestia spp.	24.4 ± 3.2	44.7 ± 3.3	24.4 ± 3.8	16.0 ± 2.8
Isopoda	27.3 ± 1.9	39.5 ± 8.1	120.8 ± 5.2	40.5 ± 8.2
Coleoptera (adults)	6.3 ± 2.2	28.0 ± 5.3	10.2 ± 6.9	-----
Coleoptera (larvae)	25.4 ± 10.8	-----	-----	41.1 ± 11.6
Carabidae	5.5 ± 1.5	-----	28.2 ± 5.0	31.5 ± 6.1
Amara aulica	3.3 ± 1.8	-----	18.9 ± 16.9	5.5 ± 1.8
Feronia nigrita	-----	-----	-----	-----
Diptera*	n/a	**595.3 ± 118.0**	n/a	-----
Formicidae	-----	n/a	-----	-----

----- Sample not collected.
* Data in bold has been calculated from samples of small mass (<0.3 g dry weight).
n/a Sample not analysed.

the Diptera were not identified to species level, but it is possible that those present feed on the decomposing strand material. It was noted that large numbers of flying insects were present during late summer and early autumn and that these were most abundant over the strand material. The similarity in the radionuclide levels for Isopoda, *Orchestia* spp. and Diptera may therefore be connected with the decomposition of strand line material.

A notable feature of the data for all the taxonomic groups is that samples collected along the seaward side generally had lower concentrations of radionuclides than those from the riverward side. Given that there are significant differences in the activity levels for some radionuclides in the flotsam, vegetation and sediment samples collected from the two sides, this is not altogether surprising but tends to be more noticeable for the detritivorous and herbivorous groups.

Figure 4.35: Temporal variation in ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in *Orchestia* spp. and Isopoda.



Data taken from the Riverward side.

Figure 4.36: Temporal variation in ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in Araneida.

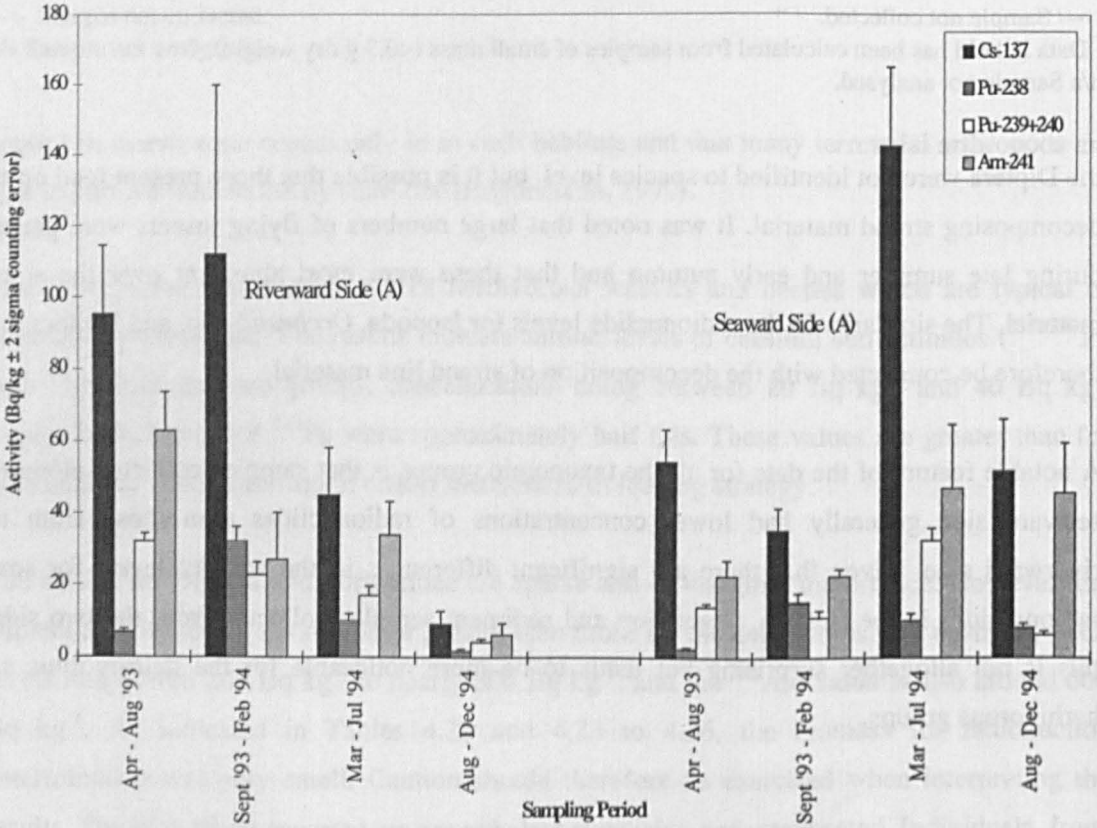
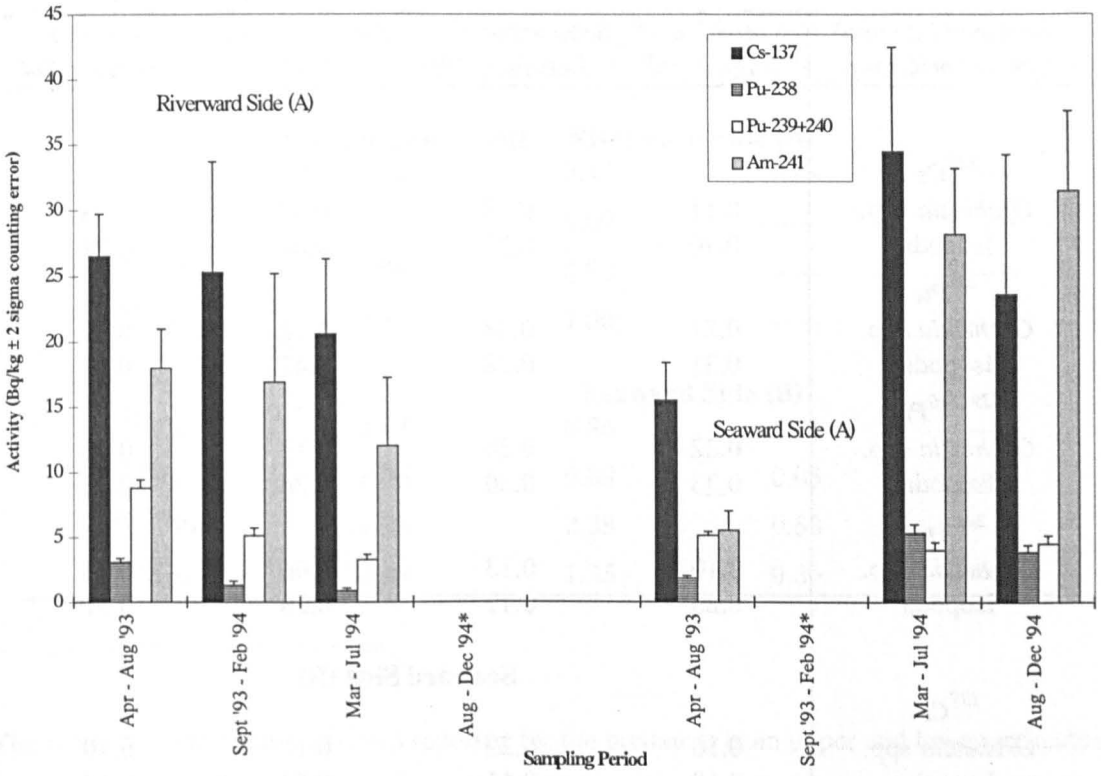


Figure 4.37: Temporal variation in ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in adult Carabids.



* no sample collected during this sampling period.

4.7.2 Radionuclide Concentration Factors in Invertebrates

Concentration factors were determined for each radionuclide for the detritivore, herbivore and predatory invertebrate groups from both sides of the River Esk. The results are presented in Tables 4.27 to 4.29. For the detritivores, the average level of each radionuclide in strand material during the relevant sampling period was used. The vegetation samples were used likewise for the herbivores, while a range of factors was determined against prey items for the predator groups.

The concentration factors for detritivores show little variation temporally or spatially on either side of the viaduct. Isopoda and *Orchestia* spp. both show very similar concentration factors, typically in the range of 0.1 to 0.3 for all four radionuclides (^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am). This suggests that whilst high levels of radionuclides are present in the strand material, transfer into the food chain is limited.

Low concentration factors were expected for the herbivorous beetles based on previous work (Rudge, 1989). However, as Table 4.28 indicates there is a wide range of concentration factors, including some greater than unity. This may be an artificial feature caused by the method employed to calculate the concentration factors. For instance, the herbivorous beetles

Table 4.27: Detritivore dietary concentration factors determined against average radionuclide activities in strand material.

Radionuclide and Species or Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August to December '94*
		Riverward Side (A)		
^{137}Cs				
Orchestia spp.	0.11	0.13	0.34	0.08
Isopoda	0.40	0.23	0.08	0.21
^{238}Pu				
Orchestia spp.	0.21	0.54	0.12	0.32
Isopoda	0.31	0.32	0.47	0.12
$^{239+240}Pu$				
Orchestia spp.	0.22	0.20	0.12	0.23
Isopoda	0.33	0.30	0.26	0.13
^{241}Am				
Orchestia spp.	0.10	0.13	0.06	0.13
Isopoda	0.23	0.17	0.28	0.11
		Seaward Side (B)		
^{137}Cs				
Orchestia spp.	0.10	0.22	0.11	0.10
Isopoda	0.10	0.11	0.54	0.14
^{238}Pu				
Orchestia spp.	0.09	0.21	0.17	0.12
Isopoda	0.17	0.13	0.13	0.18
$^{239+240}Pu$				
Orchestia spp.	0.08	0.21	0.14	0.11
Isopoda	0.15	0.14	0.08	0.18
^{241}Am				
Orchestia spp.	0.08	0.13	0.04	0.05
Isopoda	0.09	0.12	0.18	0.13

will feed across the grassland area and may also feed on the salt marsh vegetation. However, no account of activity levels in *H. portulacoides* were taken during the calculations. Furthermore, 'average' vegetation activities were determined by combining the radionuclide measurements from the three transects on the assumption that invertebrates forage across the whole area. In practice, the animals probably foraged locally to the pitfall traps that were centred around the strand line. 'Average' vegetation activity will therefore be underestimated, artificially elevating the concentration factors reported.

Table 4.28: Dietary concentration factors for the herbivorous Coleoptera from River Esk salt marshes.

Radionuclide and Species or Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August to December '94*
		Riverward Side (A)		
^{137}Cs	1.31	0.45	-----	-----
^{238}Pu	1.27	0.09	-----	-----
$^{239+240}Pu$	9.08	0.83	-----	-----
^{241}Am	2.33	1.00	-----	-----
		Seaward Side (B)		
^{137}Cs	0.43	0.86	0.33	-----
^{238}Pu	0.06	0.08	0.05	-----
$^{239+240}Pu$	1.33	2.38	0.53	-----
^{241}Am	0.48	1.35	0.30	-----

----- invertebrate samples not caught.

The range of concentration factors reported for the predators is an upper and lower estimate of the true value. Factors were estimated by assuming consumption of a single prey species/group although this is extremely unlikely in the field. However, it is suggested the bulk of the diet may well comprise *Orchestia* spp. and Isopoda, given their significant abundance within the strand material. The factors obtained are consistent over the sampling periods, perhaps indicating that the diet contains similar species over time. This would agree with observations on the abundance of *Orchestia* spp. over the study period (section 4.7).

^{137}Cs concentration factors agree with those determined previously (Crossley, 1969; Rudge, 1989) and specifically with those from Lady Wood (section 3.7.2). However, it is surprising that the actinides have similar concentration factors to ^{137}Cs. This may reflect external contamination of the predator species as a consequence of tidal inundation, although preparatory methods for the 'clean-up' of samples were targeted at this. These differences are uniform in the Carabidae which have lower concentration factors for all radionuclides, regardless of time and spatial position, when compared to Araneida and Coleopteran larvae. This provides collateral evidence for the significance of differences in diet and feeding mechanisms in the transfer of radionuclides through the invertebrate food chain.

Table 4.29: Dietary concentration factors for the predators (against a range of prey species) from River Esk salt marshes.

Radionuclide and Species or Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August to December '94*
		Riverward Side (A)		
^{137}Cs				
Araneida	0.48 - 3.59	0.38 - 5.33	0.41 - 1.68	0.11 - 0.28
Carabidae	0.13 - 0.68	0.24 - 1.20	0.19 - 0.77	-----
Coleopteran larvae	0.88 - 4.47	2.83 - 14.08	-----	-----
^{238}Pu				
Araneida	0.21 - 2.19	1.12 - 5.41	0.53 - 11.22	0.12 - 0.32
Carabidae	0.10 - 0.28	0.05 - 0.22	0.05 - 0.18	-----
Coleopteran larvae	0.49 - 1.47	n/a	-----	-----
$^{239+240}Pu$				
Araneida	0.41 - 3.64	0.31 - 4.43	0.36 - 5.15	0.08 - 0.14
Carabidae	0.11 - 0.75	0.07 - 0.82	0.12 - 0.62	-----
Coleopteran larvae	0.39 - 2.59	n/a	-----	-----
^{241}Am				
Araneida	0.67 - 3.47	0.06 - 1.59	0.34 - 2.82	0.13 - 0.16
Carabidae	0.19 - 0.65	0.22 - 0.51	0.12 - 0.62	-----
Coleopteran larvae	0.83 - 2.83	5.50 - 12.66	-----	-----
		Seaward Side (B)		
^{137}Cs				
Araneida	1.31 - 3.57	0.38 - 0.93	1.00 - 5.53	0.92 - 2.21
Carabidae	0.37 - 1.02	-----	0.24 - 1.34	0.53 - 0.75
Coleopteran larvae	0.87 - 2.39	-----	-----	1.26 - 1.80
^{238}Pu				
Araneida	0.20 - 1.50	1.87 - 3.49	1.96 - 2.67	1.51 - 2.36
Carabidae	0.25 - 1.00	-----	1.06 - 1.36	0.72 - 1.08
Coleopteran larvae	1.88 - 7.61	-----	-----	n/a
$^{239+240}Pu$				
Araneida	0.49 - 3.58	0.30 - 0.92	1.76 - 8.15	0.27 - 1.44
Carabidae	0.18 - 1.28	-----	0.22 - 0.98	0.18 - 0.29
Coleopteran larvae	0.27 - 1.93	-----	-----	n/a
^{241}Am				
Araneida	0.41 - 3.60	0.51 - 0.81	0.39 - 4.65	1.12 - 2.88
Carabidae	2.03 - 8.81	-----	0.23 - 2.76	0.78 - 1.97
Coleopteran larvae	0.93 - 4.03	-----	-----	1.01 - 2.51

----- Sample not collected.
n/a Sample not analysed.

4.7.3 Isotopic and Nuclide Ratios in Invertebrates

Table 4.30 presents isotopic and nuclide ratio data for invertebrates caught on the riverward side (A). Data are only presented for selected invertebrate groups; Araneida, *Orchestia* spp.,

Isopoda, Carabidae, Coleoptera (adults). Because ^{134}Cs was not determined successfully only the ^{238}Pu:$^{239+240}Pu$ isotopic ratio is presented.

The ratios for *Orchestia* spp. and Isopoda are very similar to those recorded for strand line material (Table 4.12). This provides further evidence about the dietary composition of these groups. The predatory invertebrates also have similar ratios to those for *Orchestia* spp. and Isopoda, believed to be the principal dietary components. Temporally, the ratios vary within the bounds of their associated error terms.

Table 4.30: Isotopic and nuclide ratios for selected invertebrate groups.

Radionuclide and Species or Group	*April to August '93*	*September '93 to February '94*	*March to July '94*	*August to December '94*
Araneida				
^{238}Pu:$^{239+240}Pu$	0.21	1.41	0.59	0.45
^{137}Cs:$^{239+240}Pu$	2.98	4.95	2.64	2.07
^{137}Cs:^{241}Am	1.52	4.16	1.33	1.44
***Orchestia* spp.**				
^{238}Pu:$^{239+240}Pu$	0.21	0.61	0.23	0.30
^{137}Cs:$^{239+240}Pu$	1.05	2.25	1.23	1.50
^{137}Cs:^{241}Am	1.34	1.89	1.36	1.72
Isopoda				
^{238}Pu:$^{239+240}Pu$	0.21	0.23	0.40	0.20
^{137}Cs:$^{239+240}Pu$	2.53	0.81	2.26	1.05
^{137}Cs:^{241}Am	2.10	0.77	1.10	0.82
Carabidae				
^{238}Pu:$^{239+240}Pu$	0.35	0.25	0.27	-----
^{137}Cs:$^{239+240}Pu$	3.01	4.96	6.24	-----
^{137}Cs:^{241}Am	1.47	1.50	1.72	-----
Coleoptera (adults)				
^{238}Pu:$^{239+240}Pu$	2.77	0.95	-----	-----
^{137}Cs:$^{239+240}Pu$	3.32	3.39	-----	-----
^{137}Cs:^{241}Am	1.41	0.64	-----	-----

----- Sample not collected.

Nuclide ratios for the adult Coleoptera are lower than expected, given the reported ratios for vegetation (Table 4.17). The ^{238}Pu:$^{239+240}Pu$ ratio is similar to that of the vegetation at around 0.2 to 0.4. The difference between the invertebrate and vegetation ratios provides further evidence that the plant foliage is covered with attached sediment particles, and that these particles vary in their radionuclide content and generate the variable nuclide ratios in Table

4.17. In addition, this provides further evidence that the beetles are feeding on a different food stuff as indicated by the concentration factors (section 4.7.3, Table 4.28).

4.7.4 Comparison with Cheshire Invertebrates

Comparing the invertebrate data with the Cheshire reference sites is not ideal given the different ecosystems. However, attempts to collect sufficient biomass from Llandridian Marsh were unsuccessful due to a combination of weather conditions and unconnected equipment problems. However, the species composition from the pitfalls at the River Esk is not totally dissimilar to that of a grassland community so basic comparisons can be drawn.

Invertebrate data for samples collected from Cheshire are presented in Table 4.31. It is evident that there are substantial differences in the activity levels of each radionuclide from the two field sites. These are thought to be biologically significant, but could not be subjected to statistical analysis with the limitations upon the composite data set.

Table 4.31: Radionuclide activity (Bq kg^{-1} ± 2σ counting error) in composite invertebrates collected from Cheshire.

Sample Dates	*Composite Invertebrate samples*			
	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
October 1993	<1.32	<0.19	<0.08	<0.38
March 1994	<0.57	<0.34	0.74 ± 0.09	<0.84
July 1994	2.51 ± 1.7	0.27 ± 0.04	0.23 ± 0.04	<0.73

<: values obtained were less than the limit of detection, the LOD value is reported.

4.7.5 Summary of Invertebrate Results

Levels of activity in ground-living invertebrates varied considerably between taxonomic groups, and were generally higher on the riverward sector of the salt marsh than on the seaward side. Extremes of the activity range for ^{137}Cs were 9 Bq kg^{-1} for adult Carabid beetles and 195 Bq kg^{-1} for the detritivorous Isopoda. Similar broad inter-taxonomic variation was recorded for the actinides: 3 to 134 Bq kg^{-1} for ^{238}Pu (Carabidae to Formicidae), 9 to 78 Bq kg^{-1} for $^{239+240}Pu$ (Carabidae to Isopoda) and 4 to 94 Bq kg^{-1} for ^{241}Am (Carabidae to Isopoda).

Detritivores, in general, exhibited the highest levels of activity, particularly within the strand line community. Of the predatory species, the Araneida (spiders) showed the highest activities, especially for ^{137}Cs. This is attributed to their exoenzyme feeding strategy in which the soft tissue of their prey, in which ^{137}Cs predominates, is pre-digested prior to

consumption. Coleopteran larvae, voracious predators upon the detritivorous adult Isopoda, also showed elevated levels of all four radionuclides. Feeding strategy is clearly a dominant factor in determining activity burdens in ground-living invertebrates.

4.8 SMALL MAMMAL RESULTS

Table 4.32 presents the general details of animals caught during each trapping session. Tables 4.33 to 4.35 present the radionuclide data for each small mammal analysed. Four trapping campaigns were undertaken, with the animals from the first three being analysed for whole body burdens. The fourth campaign focused on collecting field mice, *Apodemus sylvaticus*, for dissection and excision of the gut, body hair, lungs, muscle, organs (kidneys, liver, spleen etc.) and skeleton. Full details are given in section 3.8.3.

Animals were trapped on either side of the railway viaduct on the assumption that this structure would discourage regular movement between the two sides of the marsh. On the fourth trapping session, traps were only set on the riverward side. Attempts were made to collect a minimum of six animals (three of each sex) of each species (*A. sylvaticus*, *M. agrestis* and *S. araneus*). This was not always possible, as indicated in Table 4.32. The capture rate of *S. araneus*, in particular, was very low except for session one. This was observed in the Cheshire trapping sessions as well and is believed to have been caused by a coincident cyclic decline in shrew populations throughout parts of Britain.

Large numbers of juvenile mice and voles were caught during March and July 1994. In addition, the bank vole, *Clethrionomys glareolus*, was caught (typically between 10 and 30 individuals each session). Both of these factors reduced the trapping efficiency for the three target species.

It was originally intended to assess differences in radionuclide burdens between the sexes, but the data indicated that there were no significant differences ($p>0.05$, Mann-Whitney U test). Consequently, the results for females and males were pooled to generate arithmetic means and standard errors (when sufficient numbers were caught). These, and results for individual animals, are presented in Tables 4.33 to 4.35. Following the pilot radiochemical separation applied to mammals from the September 1993 trap session in Lady Wood (section 3.8), it was decided to combine the samples to increase the mass. This procedure was followed for the animals caught at this site. The sample pairings are listed in Tables 4.33 to 4.35.

It is evident from the data that the actinide concentrations are very similar to those recorded for the Cheshire reference site. This is surprising given the quantities of ^{238}Pu, $^{239+240}Pu$ and ^{241}Am within the salt marsh ecosystem, although most is immobilised in the sediment and

Table 4.32: Small mammals caught at the River Esk salt marsh.

Trapping Session	*Number Caught*	*Sex Distribution*	*Wet Weights (g)*
		Riverward Side (A)	
Apodemus sylvaticus			
September 1993	19 (6)	5M, 1F	31.9, 26.1, 27.7, 19.5, 18.7, 24.3
March 1994	23 (8)	3M, 3F	21.7, 18.5, 22.8, 28.6, 24.4, 21.8
July 1994	29 (4)	3M, 1F	22.8, 25.5, 17.4, 25.1
May 1995		see section 3.8.3	
Microtus agrestis			
September 1993	4 (4)	2M, 2F	30.1, 34.9, 24.1, 35.9
March 1994	1 (1)	1M	26.3
July 1994	11 (6)	3M, 3F	15.6, 30.4, 16.6, 15.8, 28.0, 27.9
May 1995	0	-----	-----
Sorex araneus			
September 1993	8 (6)	*	7.3, 7.8, 6.9, 4.2, 4.7, 6.6
March 1994	3 (2)	*	7.5, 6.0
July 1994	0	-----	-----
May 1995	0	-----	-----
		Seaward Side (B)	
Apodemus sylvaticus			
September 1993	5 (4)	3M, 1F	30.2, 23.3, 24.9, 18.6
March 1994	25 (6)	3M, 3F	24.6, 24.9, 28.2, 23.7, 25.7, 21.9
July 1994	0	-----	-----
May 1995	0	-----	-----
Microtus agrestis			
September 1993	7 (5)	3M, 2F	27.9, 36.4, 27.1, 26.7, 18.7
March 1994	0	-----	-----
July 1994	21 (6)	3M, 3F	26.7, 30.7, 33.1, 28.3, 22.1, 35.0
May 1995	0	-----	-----
Sorex araneus			
September 1993	6 (5)	*	3.0, 7.2, 6.7, 7.5, 6.4
March 1994	1 (1)	*	3.7
July 1994	0	-----	-----
May 1995	0	-----	-----

* The sex of *S. araneus* was not determined.

----- No animals were caught. Value in brackets are the number killed. Large numbers of juveniles were caught and released.

Table 4.33: Radionuclide activities in each field mouse, *A. sylvaticus*, taken from the River Esk salt marsh and Cheshire reference site.

Sample Code	^{137}Cs	*Combined*	^{238}Pu	$^{239+240}Pu$	^{241}Am
	Riverward Side				
17A	9.0 ± 3.0	18A	0.47 ± 0.08	0.35 ± 0.06	0.34 ± 0.06
18A	4.2 ± 2.4	-----	-----	-----	-----
19A	4.3 ± 2.0	25A	0.14 ± 0.03	0.30 ± 0.04	0.23 ± 0.05
25A	6.3 ± 3.8	-----	-----	-----	-----
21A	8.5 ± 4.3	38A	0.02 ± 0.01	0.16 ± 0.03	0.28 ± 0.04
38A	9.0 ± 4.4	-----	-----	-----	-----
Mean (A)	6.9 ± 0.9	-----	0.21 ± 0.13	0.27 ± 0.06	0.28 ± 0.03
3E	4.0 ± 2.2	25E	0.86 ± 0.07	0.28 ± 0.04	0.17 ± 0.03
25E	7.0 ± 2.8	-----	-----	-----	-----
8E	19.9 ± 6.2	31E	0.11 ± 0.02	0.34 ± 0.04	0.27 ± 0.04
31E	22.3 ± 5.0	-----	-----	-----	-----
11E	5.9 ± 1.9	21E	0.52 ± 0.05	0.19 ± 0.03	0.09 ± 0.02
21E	3.1 ± 1.6	-----	-----	-----	-----
Mean (E)	10.4 ± 3.5	-----	0.50 ± 0.22	0.27 ± 0.04	0.18 ± 0.05
40F	6.7 ± 2.4	46F	0.09 ± 0.02	0.21 ± 0.03	0.27 ± 0.05
46F	10.1 ± 2.4	-----	-----	-----	-----
11F	15.9 ± 5.0	50F	0.12 ± 0.03	0.43 ± 0.05	0.63 ± 0.12
50F	48.0 ± 13.0	-----	-----	-----	-----
Mean (F)	20.2 ± 9.5	-----	0.11 ± 0.02[1]	0.32 ± 0.16[1]	0.45 ± 0.25[1]
	Seaward Side				
11A	15.2 ± 4.1	42A	0.08 ± 0.02	0.14 ± 0.02	0.23 ± 0.04
42A	1.3 ± 0.3	-----	-----	-----	-----
13A	8.1 ± 2.3	43A	0.12 ± 0.03	0.32 ± 0.04	0.31 ± 0.04
43A	13.4 ± 2.6	-----	-----	-----	-----
Mean (A)	9.5 ± 3.1	-----	0.10 ± 0.03[1]	0.23 ± 0.13[1]	0.27 ± 0.06[1]
12E	27.3 ± 5.2	12E	0.04 ± 0.01	0.14 ± 0.02	0.28 ± 0.05
24E	4.9 ± 3.0	-----	-----	-----	-----
22E	19.3 ± 4.9	27E	<0.03	0.19 ± 0.02	0.21 ± 0.05
27E	3.1 ± 1.5	-----	-----	-----	-----
29E	16.8 ± 3.4	33E	0.03 ± 0.01	0.07 ± 0.01	0.11 ± 0.02
33E	2.5 ± 0.9	-----	-----	-----	-----
Mean (E)	12.3 ± 4.2	-----	<0.03	0.13 ± 0.03	0.20 ± 0.05
	Cheshire Reference Site				
11B	<0.86	17B	1.03 ± 0.07	0.15 ± 0.03	0.06 ± 0.02
14B	<0.12	35B	0.17 ± 0.03	0.07 ± 0.02	0.04 ± 0.00
17B	<2.90	-----	-----	-----	-----
18B	<0.46	19B	0.07 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
19B	<0.74	-----	-----	-----	-----
35B	<0.08	-----	-----	-----	-----
Mean (B)	<0.86	-----	0.42 ± 0.02	0.10 ± 0.003	0.05 ± 0.01

A = September 1993, E = March 1994, and F = July 1994. B = August 1993 trap in Cheshire.
All values in Bq kg^{-1} (± 2 σ counting error) except means which are ± standard error except [1] where standard deviation is reported.

Table 4.34: **Radionuclide activities in the field vole, *M. agrestis*, taken from the River Esk salt marsh and Cheshire reference site.**

Sample Code	^{137}Cs	*Combined with*	^{238}Pu	$^{239+240}Pu$	^{241}Am
	Riverward Side				
4A	8.5 ± 2.5	22A	1.90 ± 0.81	2.12 ± 0.91	4.60 ± 0.30
22A	15.7 ± 2.7	-----	-----	-----	-----
26A	22.2 ± 9.2	45A	0.36 ± 0.06	1.76 ± 0.14	2.33 ± 0.18
45A	9.2 ± 1.7	-----	-----	-----	-----
Mean (A)	13.9 ± 3.2	-----	1.13 ± 1.09[1]	1.94 ± 0.25[1]	3.47 ± 1.61[1]
10E	4.6 ± 2.2	-----	-----	-----	-----
16F	8.4 ± 4.8	41F	0.45 ± 0.09	1.33 ± 0.16	2.32 ± 0.16
41F	8.5 ± 3.2	-----	-----	-----	-----
21F	9.5 ± 4.3	22F	<0.06	0.20 ± 0.03	0.31 ± 0.06
22F	8.2 ± 4.2	-----	-----	-----	-----
45F	11.2 ± 3.4	-----	-----	-----	-----
Mean (F)	9.2 ± 0.6	-----	<0.26[1]	0.77 ± 0.80[1]	1.32 ± 1.42[1]
	Seaward Side				
5A	20.1 ± 2.5	12A+39A	0.34 ± 0.06	1.45 ± 0.12	2.54 ± 0.12
12A	12.2 ± 2.3	-----	-----	-----	-----
39A	7.7 ± 3.7	-----	-----	-----	-----
8A	14.4 ± 3.8	10A	0.20 ± 0.03	0.91 ± 0.07	1.60 ± 0.11
10A	8.5 ± 3.0	-----	-----	-----	-----
Mean (A)	12.6 ± 2.2	-----	0.27 ± 0.10[1]	1.18 ± 0.38[1]	2.07 ± 0.66[1]
1F	4.4 ± 1.9	31F	<0.05	0.23 ± 0.03	0.10 ± 0.02
31F	2.8 ± 2.2	-----	-----	-----	-----
2F	10.3 ± 3.0	14F	0.05 ± 0.01	0.19 ± 0.03	0.31 ± 0.05
5F	12.6 ± 3.4	24F	0.05 ± 0.01	0.18 ± 0.03	0.23 ± 0.05
24F	8.5 ± 4.0	-----	-----	-----	-----
Mean (F)	7.7 ± 1.8	-----	<0.05	0.20 ± 0.02	0.21 ± 0.06
	Cheshire Reference Site				
43B[2]	<1.7	-----	0.20 ± 0.04	0.34 ± 0.04	0.55 ± 0.05
24B[2]	<1.4	-----	0.17 ± 0.03	0.07 ± 0.02	<0.05
45B	<4.6	-----	-----	-----	-----
Mean (B)	<2.6	-----	0.19 ± 0.02[1]	0.21 ± 0.19[1]	<0.30[1]

A = September 1993 and F = July 1994. B = August 1993 trap in Cheshire.
All values in Bq kg^{-1} (± 2 σ counting error) except means which are ± standard error except [1] which is reported as standard deviation.
[2] Several mammal samples pooled to produce these values.

Table 4.35: Radionuclide activities in the common shrew, *S. araneus*, taken from the River Esk salt marsh and Cheshire reference site.

Sample Code	^{137}Cs	*Combined with*	^{238}Pu	$^{239+240}Pu$	^{241}Am
	Riverward Side				
3A	12.9 ± 9.1	7A	0.45 ± 0.10	0.89 ± 0.14	1.05 ± 0.19
7A	24.9 ± 2.2	-----	-----	-----	-----
6A	17.3 ± 7.8	30A	0.23 ± 0.06	0.72 ± 0.10	0.49 ± 0.09
30A	30.2 ± 6.6	-----	-----	-----	-----
15A	51.7 ± 18.8	37A	0.39 ± 0.08	1.10 ± 0.13	1.52 ± 0.18
37A	45.9 ± 15.6	-----	-----	-----	-----
Mean (A)	30.5 ± 6.3	-----	0.36 ± 0.07	0.90 ± 0.11	1.02 ± 0.30
5E	6.6 ± 1.8	-----	0.55 ± 0.10	0.98 ± 0.15	0.85 ± 0.16
19E	10.9 ± 6.4	-----	0.41 ± 0.08	0.43 ± 0.08	0.39 ± 0.07
Mean (E)	8.8 ± 3.0[1]	-----	0.48 ± 0.10[1]	0.71 ± 0.39[1]	0.62 ± 0.33[1]
	Seaward Side				
16A	43.4 ± 18.8	27A+29A	0.19 ± 0.05	0.58 ± 0.08	0.76 ± 0.13
27A	43.4 ± 15.7	-----	-----	-----	-----
29A	81.0 ± 25.3	-----	-----	-----	-----
28A	16.7 ± 2.1	46A	0.28 ± 0.05	0.70 ± 0.09	1.08 ± 0.19
46A	75.4 ± 30.7	-----	-----	-----	-----
Mean (A)	52.0 ± 11.8	-----	0.24 ± 0.06[1]	0.64 ± 0.08[1]	0.92 ± 0.23[1]
34E	15.9 ± 4.2	-----	0.83 ± 0.17	0.97 ± 0.19	1.54 ± 0.30
	Cheshire Reference Site				
21B	<5.2	5B	0.14 ± 0.04	0.13 ± 0.04	0.34 ± 0.07
5B	<1.5	-----	-----	-----	-----
6B	<0.5	-----	0.07 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
Mean (B)	<0.2	-----	0.11 ± 0.05[1]	0.10 ± 0.04[1]	0.20 ± 0.20[1]

A = September 1993 and E = March 1994. B = August 1993 trap in Cheshire.
All values in Bq kg^{-1} (± 2 σ counting error) except means which are ± standard error and [1] which are reported as standard deviation.

therefore biologically unavailable. For example, the mean concentrations for ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in *A. sylvaticus* are 0.21, 0.27 and 0.28 Bq kg^{-1} respectively for the September 1993 campaign. This compares with average reference site levels of 0.42, 0.10 and 0.05 Bq kg^{-1} for ^{238}Pu, $^{239+240}Pu$ and ^{241}Am, respectively. These are, as mentioned in section 3.8, high for reference site values. Using Mann-Whitney U tests, the differences in the data for the three trapping campaigns at the River Esk and the reference site are insignificant for all three actinides ($p>0.05$), with few exceptions. The *A. sylvaticus* data set for $^{239+240}Pu$ and ^{241}Am does exhibit significant differences between the River Esk and reference sites ($p<0.05$), but

the evidence suggests that the concentration of actinides within the mammals is mostly low and insignificant.

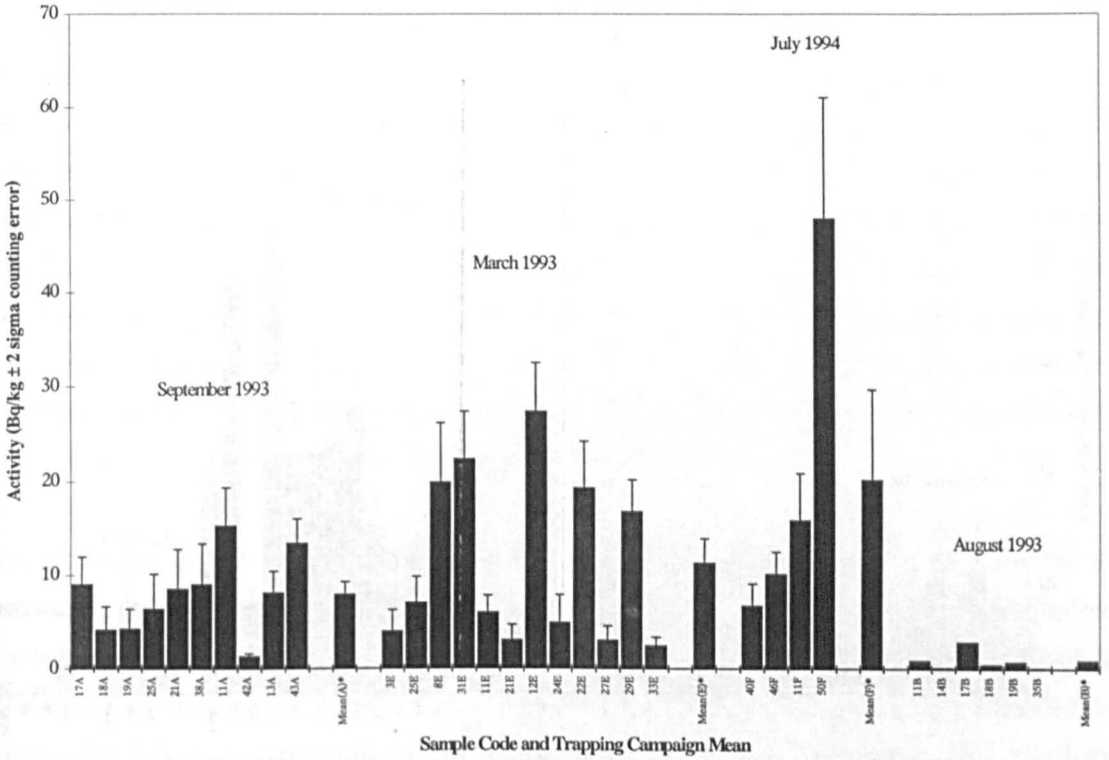
In contrast to the actinides, ^{137}Cs shows markedly higher concentrations in all three small mammal species. The ^{137}Cs levels for the three species range from 6.6 to 81.0 Bq kg^{-1} for *S. araneus* and 4.2 to 48.0 Bq kg^{-1} for *A. sylvaticus* and *M. agrestis*. ^{137}Cs levels are significantly different from those of the Cheshire reference site ($p<0.01$) for all three species even though the reference site data were probably over-estimated as limit of detection values, i.e. were upper values of the true ^{137}Cs body burden. The output from these statistical tests is presented in Table B7, Appendix B. Since caesium is more biologically mobile than the actinides, these results are not surprising. Assuming that the River Esk small mammals are in equilibrium with environmental levels of radionuclides, the principal factor determining the significant difference between the ^{137}Cs and actinide concentrations in the animal body is likely to be the low transfer coefficient of actinides across the gastro-intestinal tract. The ^{137}Cs concentration of all three small mammal species was lower than expected. This is surprising given that the ^{137}Cs levels in sediment at 6,000 to 12,000 Bq kg^{-1}, are over 4 times the concentration measured in Lady Wood soils (Table 3.3), where mean ^{137}Cs levels in *A. sylvaticus* of 55.0, 11.3 and 28.8 Bq kg^{-1} were recorded over three trapping campaigns. This compares with 6.9, 10.4 and 20.2 Bq kg^{-1} for the riverward side of the River Esk. It was expected that the concentrations of ^{137}Cs in River Esk *A. sylvaticus* would be significantly higher than those from Lady Wood, whereas in fact the data indicate the opposite.

The data indicate that the concentrations of both ^{137}Cs and the actinides are not significantly different for animals caught on the two sides of the marsh (Mann-Whitney U test, $p>0.05$). Although the levels of all four radionuclides varied in the sediment, flotsam and vegetation samples, this was mostly insignificant statistically so the similarity in the small mammal data sets was expected. Principally, this is because the small mammals are exposed to very similar concentrations of radionuclides in their diet on both sides of the marsh.

4.8.1 Temporal Variation in Radionuclide Body Burdens

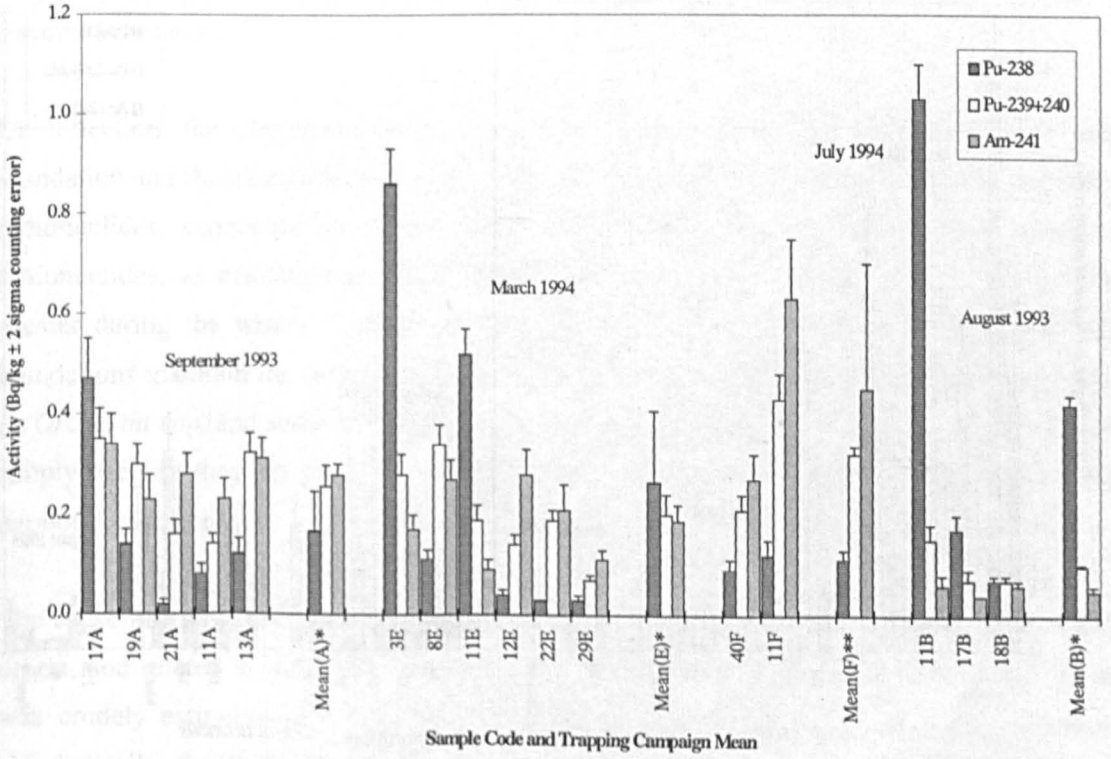
Figures 4.38 to 4.39 and 4.40 to 4.41 present data for both sides of the marsh for *A. sylvaticus*, *M. agrestis* and *S. araneus* respectively. Since the Mann-Whitney U test showed no significant differences between the activity levels for animals caught on either side of the marsh, the data were pooled to produce Figures 4.38 to 4.41. The data indicate little temporal variation, particularly for the actinides. The figures present the individual body burdens and the associated mean activity for each trapping campaign. For comparison, the data from the August 1993 reference trap are also presented.

Figure 4.38: ^{137}Cs in the field mouse, *Apodemus sylvaticus,* on the River Esk salt marsh and in Cheshire.



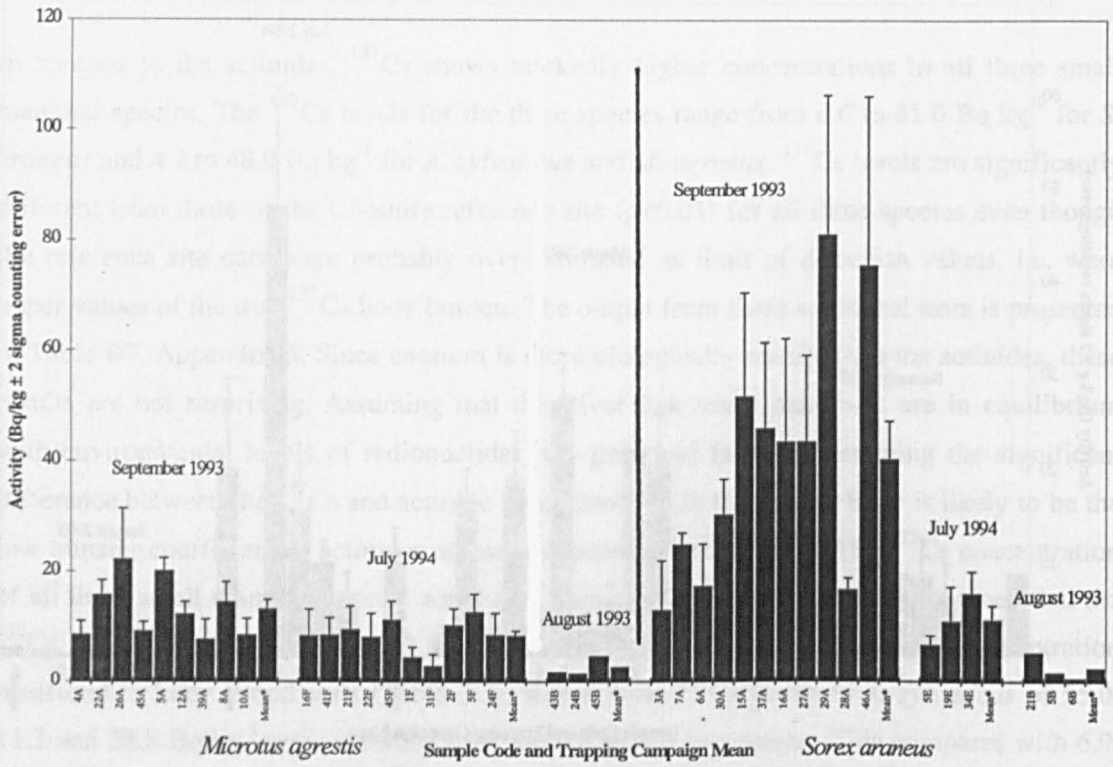
* Mean reported with standard error. Results from the Cheshire animals (August 1993) are < LOD.

Figure 4.39: ^{238}Pu, $^{239+240}$Pu and ^{241}Am in the field mouse, *Apodemus sylvaticus*, on the River Esk salt marsh and in Cheshire.



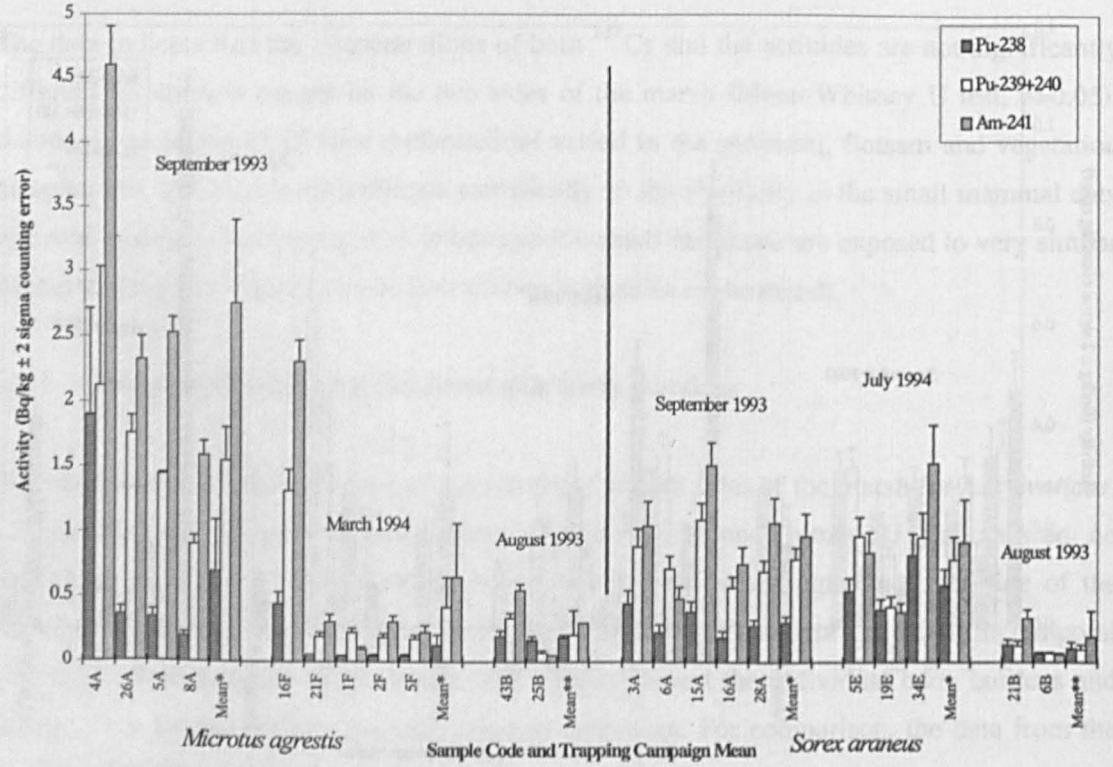
* Mean reported with standard error. ** Mean reported with standard deviation (n=2). Results from the Cheshire animals (August 1993) are < LOD.

Figure 4.40: ^{137}Cs **in the field vole,** ***Microtus agrestis*****, and common shrew,** ***Sorex araneus*****, on the River Esk salt marsh and in Cheshire.**



* Mean reported with standard error. Results from the Cheshire animals (August 1993) are < LOD.

Figure 4.41: ^{238}Pu, $^{239+240}Pu$ **and** ^{241}Am **in the field vole,** ***Microtus agrestis*****, and common shrew,** ***Sorex araneus*****, on the River Esk salt marsh and in Cheshire.**



* Mean reported with standard error. ** Mean reported with standard deviation (n=2). Results from the Cheshire animals (August 1993) are < LOD.

There is considerable variation in the concentration of radionuclides between individual animals from the same trapping session. This is only to be expected and is a function of several factors such as radionuclide concentration of dietary components, age and size of individuals, and time spent in proximity to the contamination. The data for *A. sylvaticus* and *M. agrestis* exhibit little by way of temporal variation over the trapping sessions. It is unfortunate that both *M. agrestis* and *S. araneus* were not caught in each of the three trapping sessions but it is apparent from Figure 4.40 that levels of ^{137}Cs in *S. araneus* differ between September, 1993 and March, 1994. This was not significant when tested using the Mann-Whitney U test ($p>0.05$). Statistical analysis showed conclusively that there was no temporal variation at this site ($p>0.05$). Temporal variation was originally thought probable for *A. sylvaticus* and *M. agrestis* because of changes in radionuclide concentration in the diet with changes in the components thereof throughout the year.

The elements of the diet of each species are described in section 1.4.4. The diet of *A. sylvaticus* in particular varies with seeds, vegetation and invertebrates all making important contributions at different times of the year. It was anticipated therefore that changes related to diet would be observed, and likewise for *M. agrestis*. The latter was expected to respond to factors that influence the concentration of radionuclides in grasses, for example age, condition and perhaps surface features. *S. araneus* was not expected to exhibit significant temporal variation because *Orchestia* spp. was thought to form the bulk of the diet and is abundant throughout the year. Radionuclide availability was thought to be reasonably constant, in contrast with other terrestrial sites where changes in the diet occur as the invertebrate community changes.

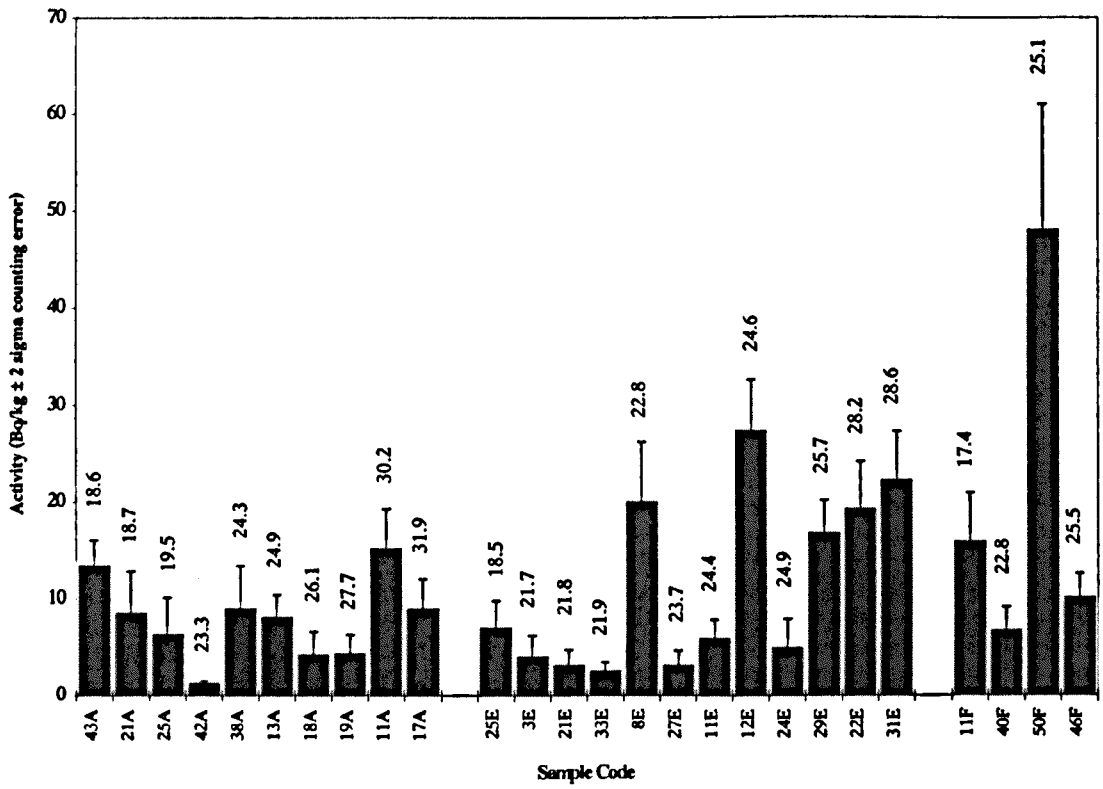
On reflection, the continuous influx of radionuclides to the marsh, from both direct tidal inundation and the deposition of strand material, is surely the dominant source of bioavailable radionuclides, especially as those present in the sediment are immobile. The influx of radionuclides, as measured in strand material samples and on the vegetation, tends to be greater during the winter. It declines during the rest of the year although additional tidal inundations maintain the radionuclide levels. In addition, *A. sylvaticus* will probably also feed on *Orchestia* spp. and some of the other invertebrates which persist through the winter months simply because they are encounter feeders. It is then less surprising that the data show little temporal variation.

The cause of the variation in the activity concentrations of individuals is surely biological in origin, and related to factors such as age, diet and physiological status. The age of animals was crudely estimated using the wet weight data. This is potentially significant because, simplistically, the older the individual, the longer its exposure to radionuclide contamination. Figure 4.42 presents data for ^{137}Cs concentrations in *A. sylvaticus* plotted against individuals,

ranked in order of age using wet weight. There is a trend towards increasing ^{137}Cs concentration with wet weight but this cannot be definitively examined with the current data set. Also, a better indicator of age is required.

The actinide data show neither temporal variation or significant variation in individual body burdens. This is not surprising given the low transfer coefficients for actinides. It is also interesting to note that there are no significant differences in the actinide data between the three species ($p>0.05$). However, comparing the ^{137}Cs data, *A. sylvaticus* and *M. agrestis* have similar concentrations, but these are significantly lower than for *S. araneus,* ($p<0.05$). Higher concentrations of ^{137}Cs in *S. araneus* compared to *A. sylvaticus* and *M. agrestis* have been reported elsewhere following the passage of the Chernobyl plume over Britain (Rudge, 1989). This was accounted for by changes in the activity levels of invertebrate species which form the bulk of the *S. araneus* diet. The body concentrations of ^{137}Cs for *S. araneus* from this site are almost certainly related to prey abundance. The data for *Orchestia* spp. caught in large numbers in the pitfall traps indicate that they contain comparatively high levels of ^{137}Cs and that they are abundant throughout the year. The reported concentrations of ^{137}Cs in *S. araneus* are therefore believed to be related to the consumption of large numbers of *Orchestia* spp.

Figure 4.42: ^{137}Cs Data presented in ascending order of wet weight as an index of age for each trapping session.



Data from both sides of the marsh are combined.

4.8.2 Tissue Distribution of ^{137}Cs

Animals caught during the final trapping session were dissected to provide information about the tissue distribution of ^{137}Cs. The traps were set on the riverward side of the marsh to reduce any influence from spatial variation. Of the eight animals caught, four individuals were combined to produce two replicates for each sample category: body hair, gut (with contents), lungs, muscle, organs (comprising kidneys, liver, spleen, heart and sexual organs) and skeleton. Random number tables were used to select the combination of animals. Table 4.36 presents data for the six body compartments.

The ^{137}Cs levels reported in Table 4.36 are low, as expected from the whole body burden data reported earlier. However, it was expected that the body hair component would be higher than the reported 10 Bq kg^{-1} on the basis that as the animals forage, sediment and detritus particles would become adhered to their body hair. However, it is probable that grooming removes these particles, possibly leading to their ingestion. This behaviour was probably accentuated during the animals' confinement in the trap. In much the same way, the ^{137}Cs activities reported for the gut (plus contents) are lower than expected. In this case, it is probable that gorging of the bait in the traps occurred. This will have diluted the ^{137}Cs concentration and the high ingestion rate will have also caused more material to be defecated. The lost material would have comprised natural food items such as vegetation, invertebrates, and others. Both of these factors will have artificially reduced the radionuclide content of the gut.

Table 4.36: ^{137}Cs in tissues and organs of *A. sylvaticus* from River Esk.

Tissue	*Combined Wet Weight (g)**	*^{137}Cs (rep 1) Bq kg^{-1}***	*^{137}Cs (rep 2) Bq kg^{-1}***	*^{137}Cs Bq kg^{-1} ± Standard Deviation*
Body Hair	2.64	<0.76	11.7 ± 5.2	6.2 ± 7.7
Gut (full)	41.24	10.9 ± 2.1	15.6 ± 1.8	13.2 ± 3.3
Lungs	4.07	79.4 ± 20.0	71.4 ± 21.5	75.4 ± 5.7
Muscle	10.57	31.8 ± 5.9	53.6 ± 24.7	42.7 ± 15.4
Organs	29.87	4.4 ± 0.9	22.3 ± 2.8	13.4 ± 12.7
Skeleton	5.30	<1.6	12.9 ± 2.5	7.2 ± 8.0

* All eight animals. ** Dry weight reported. Replicate values are reported ± 2σ counting error.

Based on the known behaviour of ^{137}Cs within the body, it was thought that the bulk of the activity would be located in soft tissues such as muscle and the various organs extracted. The results support this contention, with levels of 30 to 50 Bq kg^{-1} in muscle and, slightly lower, 4 to 22 Bq kg^{-1} in the organs. In contrast, the ^{137}Cs levels in the skeleton averaged at 7 Bq kg^{-1}. The variation between the replicate samples is high, as indicated by the standard deviation values; but the results still show that ^{137}Cs accumulates more in the soft tissues.

The most notable result is that for the lungs, although being such a small body component the lungs only contribute 1 to 2 Bq kg^{-1} towards the whole body burden. However, the lung tissue analysed exhibited the highest ^{137}Cs activity. This is interesting because resuspension of dry sediment is expected across the marsh and it is anticipated that the resuspended particles would be of respirable size. This is important because uptake of radionuclides will occur across the epithelial lining of the lungs, albeit at a low rate. Furthermore, the ciliated epithelial lining will remove particles from the lungs and these will frequently be ingested and therefore potentially available for uptake from the gastro-intestinal tract. Given the high levels of actinides in sediment from the River Esk, it is likely that alpha emissions in the lungs are also important. In addition, alpha particle emissions during radioactive decay are likely to cause considerable radiation damage to surrounding tissues. Further analysis of these samples is therefore required to confirm the tissue distribution of the actinides, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am within *A. sylvaticus*.

Calculation of the ^{137}Cs levels for the whole body burden for the animals used to determine the activity in the body components listed in Table 4.36 produced estimates of between 5 and 11 Bq kg^{-1}. It should be noted that these are likely to be underestimates because sample material was discarded during dissection and preparation of the skeleton. These estimates are however comparable to the body burdens reported in Table 4.33 and therefore the tissue distribution should be representative of the mammals caught previously.

4.8.3 Isotopic and Nuclide Ratios

Table 4.37 shows calculated isotopic (^{238}Pu:$^{239+240}Pu$) and nuclide (^{137}Cs:$^{239+240}Pu$ and ^{137}Cs:^{241}Am) ratios for the small mammals caught on the River Esk salt marsh. Data for both sides of the marsh and for each trapping session are presented. Analytical difficulties with ^{134}Cs prevented the calculation of the ^{134}Cs:^{137}Cs isotopic ratio.

The data for ^{238}Pu:$^{239+240}Pu$ show limited variation over the trapping sessions and also for the two sides of the marsh. Comparing the levels to those recorded for sediment and vegetation (Tables 4.7 and 4.17 respectively), it is evident that the ratios are higher for the small mammals. This scale of variation was also observed for invertebrates (Table 4.30) and reflects the range of food items in the diet of the different mammal species. It is notable however, that *M. agrestis* values (particularly for those caught on the seaward side) are close to those reported for the two main vegetation species, *B. mollis* and *F. rubra* (Table 4.17), which will form the bulk of the *M. agrestis* diet.

The nuclide ratios reported for the small mammals are much greater than those for the invertebrates, vegetation and sediment. The latter are all low and reflect the surface

contamination caused when tidal inundation occurs. Further evidence of this is provided by the nuclide ratios for *M. agrestis*. These values are lower than those for *A. sylvaticus* and *S. araneus* and, it is believed, reflect the consumption of, and foraging in, the grass swards which are regularly inundated. In doing so, *M. agrestis* is likely to ingest food covered with sediment particles and therefore reflect nuclide ratios closer to those reported for the sediment.

Table 4.37: Isotopic and nuclide ratios for small mammals from River Esk salt marsh.

Trapping Session	$^{238}Pu:^{239+240}Pu$	$^{137}Cs:^{239+240}Pu$	$^{137}Cs:^{241}Am$
		A. sylvaticus	
RIVERWARD SIDE			
September 1993	0.78	25.6	24.6
March 1994	1.85	38.5	57.8
July 1994	0.34	63.1	44.9
SEAWARD SIDE			
September 1993	0.43	41.3	35.2
March 1994	0.23	94.6	61.5
		M. agrestis	
RIVERWARD SIDE			
September 1993	0.58	7.2	4.0
July 1994	0.34	12.0	7.0
SEAWARD SIDE			
September 1993	0.23	10.7	6.1
July 1994	0.25	38.5	36.7
		S. araneus	
RIVERWARD SIDE			
September 1993	0.40	33.9	29.9
July 1994	0.68	12.4	14.2
SEAWARD SIDE			
September 1993	0.38	81.3	56.5
July 1994	0.86	16.4	10.3

4.8.4 Radionuclide Concentration Factors for *M. agrestis* and *S. araneus*

Concentration factors were determined for each radionuclide for *M. agrestis* and *S. araneus*. Factors were not calculated for *A. sylvaticus* because, without specifically analysing for gut contents, it is difficult to assess the dietary components at the time of capture. Table 4.38 presents a range of concentration factors for *M. agrestis* and *S. araneus* for animals caught on both sides of the marsh. *M. agrestis* values were determined against both *B. mollis* and *F. rubra*, which are believed to form the bulk of the diet. Vegetation results from the front and back transects were used to calculate the range presented. Upper and lower concentration factors as determined from the different prey species are presented for *S. araneus*.

It is evident that the concentration factors for *M. agrestis* are very low for the actinides. This is not entirely surprising, although the nuclide ratio data (Table 4.37) indicated the consumption of sediment contaminated foliage so an elevated concentration factor was anticipated. The concentration factors for ^{137}Cs are however, comparable to those obtained by Rudge (1989) who reported a range from 0.10 to 1.49. This is for animals caught on a contaminated grassland site on the BNFL, Drigg site. In addition, Reichle *et al.* (1970) reported a range of concentration factors for a number of herbivorous species from different habitats ranging from 0.3 to 2.0. The results from this study are in agreement with previous work.

S. araneus exhibits a similar range of concentration factors for animals caught on either side of the viaduct. Again, these are very low for the actinides, reflecting the very low mammalian transfer rates. Given the actinide concentrations of the principal invertebrate prey, namely *Orchestia* spp. and *O. asellus* (Tables 4.24 to 4.26), higher concentration factors were anticipated. ^{137}Cs uptake is generally higher although the reported concentration factors are

Table 4.38: Radionuclide concentration factors for *M. agrestis* and *S. araneus* from River Esk.

Trapping Session and Vegetation	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
		M. agrestis		
RIVERWARD SIDE				
September 1993				
B. mollis	0.71 - 1.36	0.06 - 2.13	0.17 - 0.92	0.09 - 4.60
F. rubra	0.61 - 2.10	0.12 - 2.09	0.26 - 1.13	0.03 - 0.84
July 1994				
B. mollis	0.22 - 2.44	0.05 - 1.44	0.04 - 1.12	0.03 - 0.98
F. rubra *	0.11	0.02	0.01	0.01
SEAWARD SIDE				
September 1993				
B. mollis	0.45 - 2.00	0.02 - 0.71	0.02 - 0.53	0.11 - 0.34
F. rubra	0.58 - 1.83	0.02 - 0.69	0.01 - 0.66	0.07 - 1.85
July 1994				
B. mollis	0.35 - 0.84	0.02 - 0.31	0.02 - 0.35	0.01 - 0.23
F. rubra	0.21 - 0.78	0.01 - 0.17	0.01 - 0.23	0.01 - 0.08
		S. araneus		
RIVERWARD SIDE				
September 1993	0.10 - 1.45	0.01 - 0.28	0.01 - 0.20	0.02 - 0.19
March 1994	0.28 - 0.43	0.03 - 0.53	0.01 - 0.22	0.01 - 0.05
SEAWARD SIDE				
September 1993	0.33 - 1.35	0.02 - 0.05	0.02 - 0.06	0.02 - 0.04
March 1994	0.11 - 0.62	0.07 - 0.20	0.03 - 0.24	0.01 - 0.15

Concentration factors for *M. agrestis* were calculated using vegetation samples collected from the front and back transects except * where only the front transect was used.

lower than those observed by Rudge (1989) following the Chernobyl accident. It was expected that *S. araneus* would concentrate radionuclides present in their prey but, as the range of factors reported indicates, much will depend upon the seasonal abundance, and therefore frequency in the diet of *S. araneus*, of the different prey species. The abundance of *Orchestia* spp., for example, is likely to significantly influence towards the concentration of the radionuclides within *S. araneus*. Furthermore, caution is required when interpreting these concentration factors because of the long timescale for the invertebrate collection. It is anticipated that *S. araneus* equilibrates with the bioavailable radionuclides present in its environment very rapidly, perhaps over a few hours to days. However, it is not possible to determine changes in the invertebrate communities over a similar timescale.

4.8.5 Summary of Small Mammal Results

Mean activity levels for ^{137}Cs in common shrews (*S. araneus*) ranges from 6.6 to 81 Bq kg^{-1} compared to 4.2 to 48 Bq kg^{-1} for wood mice (*A. sylvaticus*) and field voles (*M. agrestis*). These values were significantly ($p<0.01$) greater than for the reference site. In contrast, actinide concentrations were low and very similar for the salt marsh ecosystem and the reference site, with mean values for *A. sylvaticus* from the salt marsh at 0.21 Bq kg^{-1} (^{238}Pu), 0.27 Bq kg^{-1} ($^{239+240}Pu$) and 0.28 Bq kg^{-1} (^{241}Am). This compares with 0.42, 0.10 and 0.05 Bq kg^{-1} for these three actinides at the reference site. Differences between the accumulation of radiocaesium and the actinides are attributed to differential mobility of the isotopes.

No differences were found in the body concentrations of actinides or radiocaesium in animals from the riverward and seaward sectors of the marsh. Moreover, and not surprisingly, there was no indication of temporal variation, probably because the age structure of the mammal populations is in balance as regards loss and recruitment. Despite the absence of inter-seasonal differences, there was considerable variation in ^{137}Cs values within each population sample, with age, diet and physiological differences between individuals the dominant factors involved. Concentrations of radiocaesium were notably higher in *S. araneus* than in *A. sylvaticus* and *M. agrestis*, which is surely a reflection of the activity levels in the invertebrates (especially *Orchestia* spp.) comprising the diet of the shrew. Variability within seasonal samples of the populations of all three species was not apparent for the actinides, due to their low transfer coefficients.

Pilot studies into tissue distribution of radionuclides in small mammals showed that the muscle, lungs and pooled viscera (kidneys, liver, spleen, heart and sex organs) were centres of accumulation of ^{137}Cs. The significance of alpha-particle emissions to animal exposure can only be speculated upon in the absence of specific concentration data.

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Chapter Five
A SAND DUNE ECOSYSTEM

5.1 INTRODUCTION

5.1.1 Background - previous studies

Sand dunes are strongly influenced by tides and, like salt marshes, plants have the initial problem of establishing in an unstable soil. Across the foreshore, sand is washed daily by the tides such that no flowering plants have succeeded in colonising it. In addition, when sand is exposed, it soon dries and may be eroded by wind, making establishment difficult. Moreover, unlike the sediment on which salt marshes develop, sand is deficient in plant nutrients and moisture. Consequently, sand dune development begins around the fringes of the tidal reach where the presence of strand litter ameliorates these deficiencies. In much the same way as salt marshes develop, early plant colonisers begin to stabilise the sand which encourages further sand accumulation, beginning dune formation.

The actual rate of dune growth will be determined by a number of factors such as topography which will affect the airflow over the dunes. Sand dunes develop as the sand, being moved by the process of saltation, is trapped and accumulated by clumps of vegetation. In particular, *A. arenaria* is responsible for the formation of sand dunes in many parts of the world (Ranwell, 1972). As the sand accumulates the growth of *A. arenaria* is able to keep pace with the increasing depth and plants therefore avoid becoming buried. Moreover, *Ammophila arenaria* (marram grass) develops extensive horizontal rhizomes and this encourages widescale colonisation of the sand. As the vegetation cover increases so does the rate of dune accretion to a point where the dunes become more stabilised. In this way, dunes gradually develop in size.

Interest in sand dune ecosystems in this study arises primarily because the process of sea to land transfer results in the transport of certain radionuclides which are not normally present in the terrestrial environment, and in quantities which mask the presence of them from fallout from the atmospheric weapons testing of the 1950s and 1960s. There are several radionuclides of radiological significance involved namely, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am.

The discovery of sea to land transfer of radionuclides catalysed a series of studies during the 1980s based around BNFL, Sellafield in Cumbria. In particular, research concentrated on the transfer mechanism involved and the distance inland that enhanced actinide deposition could still be detected (Cambray and Eakins, 1980, 1982; Eakins *et al.*, 1981, 1982; Pattenden *et al*, 1980, 1987; Peirson *et al.*, 1988). The focus was the impact on human health (Garland and McKay, 1992; McHugh and Hetherington, 1987; Stather *et al.*, 1986). However, few studies have investigated the radionuclide uptake into food chains in semi-natural habitats as a result

of contamination by sea to land transfer. In terms of radionuclide exposure of the public, the critical pathway is through external irradiation. This is through gamma rays from material deposited to the vegetation and accumulated in the skeletal soil, and following inhalation of radionuclides resuspended from the sand and associated with sea spray.

5.1.2 Study Objective

The principal objective of this section of work was to investigate the behaviour and transfer of the radionuclides through natural food chains on a remote sand dune ecosystem receiving a continuing input of radionuclides. This was achieved through the analysis of sand, vegetation, invertebrates and small mammals collected over a period of two years. The following discussion presents the results of the spatial and temporal variation in the radionuclides measured. Transfer factors for the radionuclides to the invertebrate and small mammal species are also presented.

5.1.3 Sea to land transfer

The behaviour of radionuclides released to the marine environment from Sellafield has already been described (section 4.1) although principally in the context of deposition across salt marshes as a consequence of tidal inundation. The mechanism of sea to land transfer is now relatively well understood. Cambray and Eakins (1980, 1982) first used sea to land transfer to explain the observed levels of radioactivity in the vicinity of Sellafield, having measured levels of ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in excess of that expected from weapons fallout alone. Further investigations noted that the levels of excess $^{239+240}Pu$ and ^{241}Am decreased with distance inland and that the actinides exhibited an enrichment of 2 to 3 orders of magnitude in sea spray compared to seawater.

The actual mechanism whereby radionuclides in the sea can become airborne is thought to be due to bubbles bursting at the sea surface. As air bubbles rise through seawater, they can scavenge particulate material and transport it to the surface, at which point the bubbles burst. During this latter process, spray droplets are formed which become airborne. Along the shoreline, the surf zone is an area of concentrated bubble production, as air trapped by breaking waves returns to the surface. Eakins *et al.* (1982) showed that this region appears to be the major area for generating radionuclide contaminated sea spray. The size of the spray droplets produced in this manner range from 0.1 to 150 μm (Exton *et al.*, 1985). Sea spray droplets are also formed further out at sea by the shearing of wavecrests by wind (Hickmott, 1981) but although radioactivity may become airborne as a result, it is unlikely to form a significant component of the material returned to the terrestrial environment (Eakins and Lally, 1984). Actinides are strongly associated with particulates and it is these particulates which are

released as the bubbles burst. As a result, significant levels of $^{239+240}Pu$ and ^{241}Am can become airborne and thus return to land. This can account for the observed enrichment of radionuclides in sea spray over seawater and consequently for the elevation in the actinides compared to ^{137}Cs measured in soil within a few kilometres of the coastline.

Several studies have investigated the distance that sea spray travels inland, often through exposure of muslin screens attached to wire frames (Eakins *et al.* 1982; McHugh *et al.*, 1986; Pattenden *et al.*, 1987). Generally, the distance travelled inland varies with climatic and topographic conditions, but the bulk of the sea spray and radioactivity is deposited within the first few hundred metres. For example, Malloch (1972) reported that 90% of sea spray was deposited within the first 500 m, although a proportion of the remaining 10% was recorded 2 km inland.

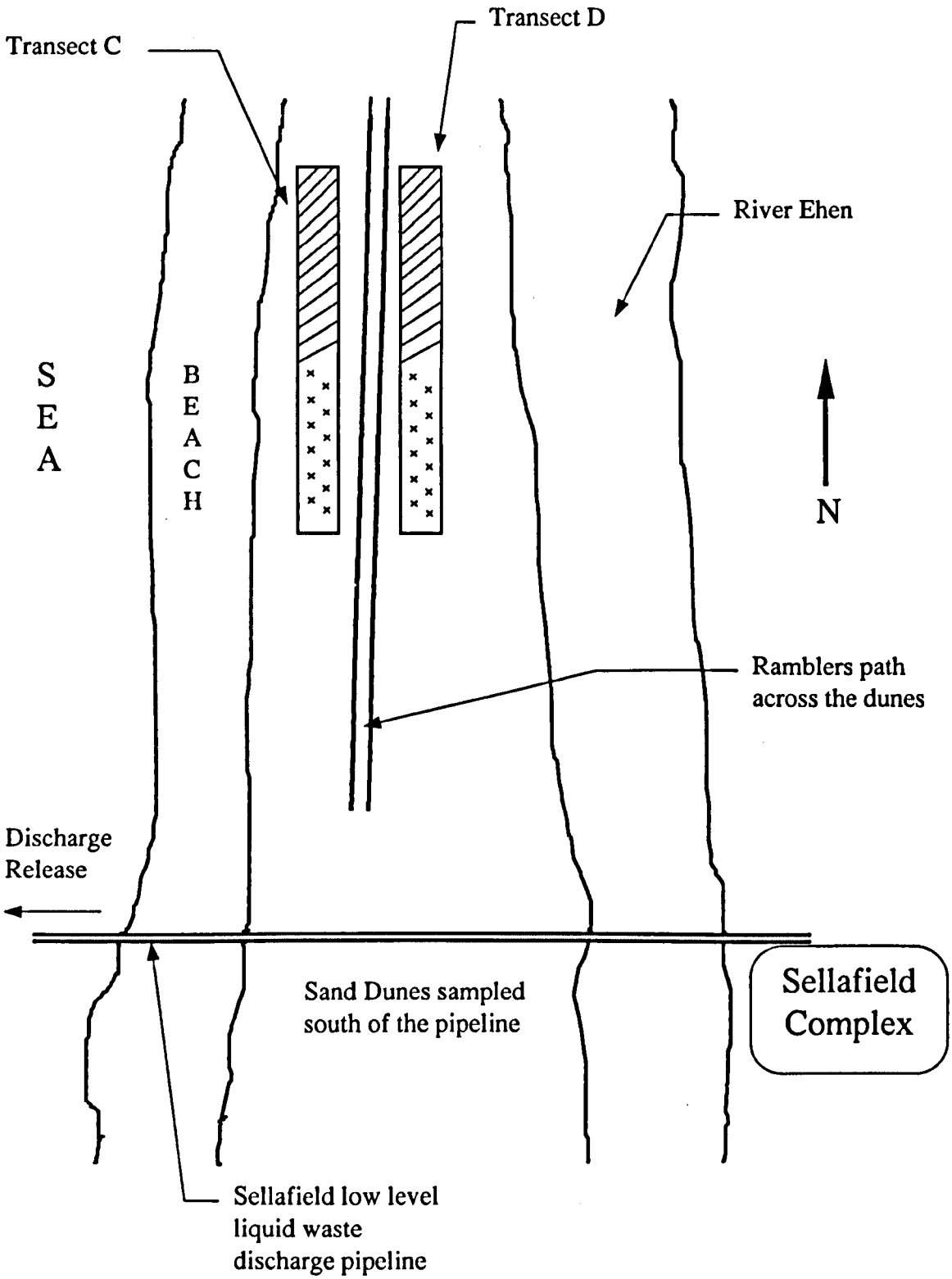
5.2 SITE DESCRIPTION

The sand dunes used in this study lie to the west of Sellafield (OSGR: NY 016037) and are within 500 m of where the low-level liquid waste discharge pipeline enters the sea. The dunes form a narrow corridor, up to 50 m wide, which runs parallel to the coastline for about 2 km. The dunes separate the River Ehen from the sea and, as a consequence, the study area is isolated from nearby agricultural land as shown in Figure 5.1. Any small mammals and ground dwelling invertebrates living on the dunes are therefore confined to living and feeding within the study area. Being within 1 km of the main Sellafield site, it is likely that radionuclides are deposited across the area by aerial emissions as well as sea to land transfer. However, it was expected, based partly upon the dominant wind direction (inland) and the dominance of the sea to land transfer mechanism, that most of the radionuclides deposited across the site will have originated from the low level liquid discharges to the marine environment.

The study site is on land owned by BNFL and has been unaffected by any major land use changes for over forty years. However, about ten years ago the dunes immediately to the south of the pipeline were artificially raised. This has led to a mixing of 'fresh' sand with older contaminated material. In addition, the vegetation cover of the two areas differs, with the raised sand dunes having a greater coverage of red fescue (*Festuca rubra*) than marram grass (*Ammophila arenaria*) compared to the selected field site. This is may be relevant because small mammals could forage in this region.

A study site was selected at random along the dunes. The chosen site was approximately 150 m from the pumping station on the low level liquid discharge pipeline at a point where the dunes were about 30 m wide. To one end of the transects was an area of sea buckthorn

Figure 5.1: **Outline sketch of the sand dunes site indicating sampling locations.**



Not to scale.

x = location of pitfall traps. Shaded area used for small mammal trapping.

(*Hippophae rhamnoides*) which would have provided some cover. The site was conveniently divided by a ramblers path along the highest point of the dunes. One 50 m by 4 m transect was marked out along each side of the path. The two transects were approximately 5 m apart as shown in Figure 5.1. This provided a front edge (transect C) dominated by *A. arenaria* and exposed directly to the sea, and a back edge (transect D) which was in a slightly more sheltered position and dominated by *F. rubra*. The vegetation growing within the site during the study period is given by Table 5.1 but the site was dominated by *A. arenaria* and *F. rubra*, which together covered approximately 95% of the vegetated area. During the summer, annual species appeared and, where possible, samples of these were collected and analysed. A number of the species listed are probably escapes from nearby agricultural areas. Plates 5.1 and 5.2 indicate differences in the vegetation cover along transects C and D, particularly the almost total cover of *A. arenaria* along transect C compared to transect D.

During high tides the sea reaches the base of the sand dunes which are raised by 1 to 2 m above sea level. Vegetation covers more than 90% of the sand dunes, being dominated by *F. rubra* and *A. arenaria*. The topography of the dunes follows a classical formation, having frontal mobile dunes partly stabilised by *A. arenaria* with a slack region to the rear. The soil profile is in a juvenile state and consists mainly of sub-angular and round sand particles in the size range 0.2 to 2 mm (coarse sand). There is little organic matter present, reflecting the recent and slow rate of colonisation and the soil pH is almost neutral at around 7.5 (Table

Table 5.1 **Common species of vegetation found along the sand dunes during the study period.**

Marram Grass	*Ammophila arenaria*
Red Fescue	*Festuca rubra*
Plantain	*Plantago lanceolata*
Sea Bindweed	*Calystegia soldanella*
Bramble	*Rubus fruticosus*
Dandelion	*Taraxacum officinale*
Sea Buckthorn	*Hippophae rhamnoides*
Sea Plantain	*Plantago maritima*
Sea Aster	*Aster tripolium*
Yellow Rattle	*Rhinanthus minor*
Common Storksbill	*Erodium cicutarium*
Bloody Cranesbill	*Geranium sanguineum*
Wintercress	*Barbarea vulgaris*
Corn Spurrey	*Spergula arvensis*
Agrimony	*Agrimonia eupatoria*
Red Hemp Nettle	*Galeopis angustifolia*
Few leaved Hawksweed	*Hieracium murorum*
Fat Hen	*Chenopodium album*
Scentless Mayweed	*Matricaria perforata*

5.6). This is slightly lower than expected, given a normal coastal soil pH level around 8.8 to 9.0 (Putten *et al.*, 1988). The rear dunes are more extensively colonised with a greater proportion of *F. rubra* and have a similar particle size distribution (Table 5.6).

In this study, the dunes have developed along an open coastal site and have stabilised at a fairly low height. The shape of the dunes indicates that the formation has reached its maximum height and that the dune is slowly migrating forward down the beach (although this is tempered by the action of waves). Behind the frontal dune, soil erosion by the action of wind has produced a slack area, classic for this type of sand dune system (Ranwell, 1972).

For comparison purposes, vegetation and soil samples were collected from the University of Liverpool's Botanical Gardens at Ness, Cheshire (OSGR: SJ 304754). The nearest nuclear establishment is BNFL, Capenhurst which releases no radionuclides of interest (section 3.2). However, given the dominance of *A. arenaria* and the sandy soil, some samples were also collected from a sand dune system at Whitford Burrows in Wales (OSGR: SS 444952) which had the same species of vegetation. The Welsh site was selected using data from the MAFF aquatic monitoring report (Camplin, 1993) which showed a decline in the ^{137}Cs activity in filtered seawater from 0.25 Bq kg^{-1} around Sellafield to 0.01 Bq kg^{-1} on the Welsh coastline. Unfortunately, it was only possible to visit the Welsh site on one occasion, during August 1994. The nearest nuclear establishments were Amersham International, located near Cardiff and the Berkeley and Oldbury Power stations operated by Nuclear Electric further up the River Severn. The liquid discharges from these sites were assessed (Table 4.2) and shown not to release the radionuclides of interest in significant quantities. Consequently, the Cheshire site should reflect atmospheric deposition of radionuclides released as a result of historic weapons testing and the Chernobyl accident in 1986. In addition, the Welsh site should reflect the transfer of radionuclides from the sea in a relatively 'clean' environment compared to the environs of Sellafield. More importantly, adoption of the Welsh site allows a direct comparison of similar vegetation species.

5.3 MATERIALS AND METHODS

5.3.1 Soil Core Sampling

Short soil cores were obtained on four occasions between May, 1993 and September, 1994. An area of vegetation was cleared to uncover the sand before a 10 cm diameter soil corer was used to extract cores to a depth of 12 cm. The extracted cores were compared to the remaining hole for any signs of vertical compaction. In this case, there was none. Depending upon the end use of the sample, either two cores were collected at four points along each transect or three cores were collected along the whole of the transect. Each set of cores was combined and any large stones or pieces of vegetation were removed. Wet weights were recorded. The

Plate 5.1: **Photograph of Transect C, the front of the sand dunes indicating the almost complete vegetation cover provided by *A. arenaria*.**

Plate 5.2: **Photograph of Transect D, the back of the sand dunes with the low level radioactive waste disposal pipeline in the background.**

three cores were sectioned carefully in the field into 0 to 4 cm, 4 to 8 cm and 8 to 12 cm slices before being bulked as described for the other cores. Being sand, the cores often collapsed during sectioning. If it was believed that the sections had thus been mixed in the tray, the core was discarded and a replacement extracted. The samples were then prepared as described in section 3.3. Typically, a Marinelli beaker held about 500 g of sand and required count times in the order of 20,000 to 30,000 seconds (5.5 to 8 hours) to obtain data for ^{134}Cs, ^{137}Cs and ^{241}Am. Sub-samples of 5 g were collected for radiochemical separation and analysis for plutonium isotopes. The radiochemistry method is described in Appendix A. Organic matter content, particle size and pH measurements were determined on some of the sand samples. The methods employed are described in section 3.3.

Comparison soil cores were collected from the Cheshire during August 1993 and August 1994 and from Wales during August, 1994. Organic matter and pH measurements were also determined for these samples.

5.3.2 Vegetation Sampling

Vegetation samples in the form of *F. rubra* and *A. arenaria* were collected at intervals from May, 1993 to February, 1995. Initially, samples were collected bi-weekly until March, 1994, then monthly until July, 1994 and finally bi-monthly. Samples were also obtained for herbaceous species which appeared during the year. On most occasions insufficient material of any one species meant that these were formed into a composite sample for the purpose of analysis and therefore comprised a mixture of the species listed in Table 5.1. In all cases the vegetation was collected by clipping to within 2 to 3 cm of the ground. Two samples of *F. rubra* and *A. arenaria* and composite samples were collected per transect. Samples were stored in a cold room (<5°C) until they were prepared for radionuclide determination as described in section 4.4. Wet and dry weights were recorded. A Marinelli held between 40 and 60 g of sample depending upon the plant species. Typically, these required between 40,000 and 85,000 seconds (11 to 24 hours) to obtain data for ^{137}Cs and ^{241}Am. ^{134}Cs was not always determined successfully. A sub-sample of 25 g was taken for radiochemical separation and alpha spectrometry to determine the activity of the plutonium isotopes, ^{238}Pu and $^{239+240}Pu$.

On some sampling occasions it was not possible to collect sufficient live vegetation to constitute a sample. When this occurred, samples of senescent material were collected. During the first two sampling trips sufficient live and senescent material was collected to allow comparisons to be made. Subsequently, where possible, only fresh live material was collected and analysed. In addition, during the first two sampling occasions two further samples were collected along each transect to investigate the spatial variability along the 50 m transects.

During September 1994, additional sand and vegetation samples were collected from the new sand dune region to the south of the discharge pipeline to assess the availability of radionuclides for uptake and transfer through the food chain. The samples were prepared and analysed as described above.

5.3.3 Invertebrate Sampling

Invertebrates were collected using pitfall traps. Full details are given in section 3.3. For this site, two sets of six pitfalls were set along each transect. The pitfalls were first exposed in May 1993 and remained in place until January 1995. They were changed regularly, usually monthly. It was originally intended that each set of six pitfalls would form one replicate. Unfortunately, the low biomass of the different taxonomic groups (section 5.7) necessitated the pooling of all 12 pitfalls in each transect. Samples were also pooled in approximately quarterly intervals. Sample storage and preparation is described in section 3.3. Typically, invertebrate samples were counted in a pot geometry for between 85,000 and 200,000 seconds (24 to 55 hours) to obtain measurements of ^{137}Cs and ^{241}Am. A sub-sample was taken for radiochemical separation for plutonium measurement the quantity being dependent upon the mass of the whole sample.

5.3.4 Small Mammal Sampling

Small mammals were caught using techniques described in section 3.3. Suitable animals were culled in the field, weighed and immediately placed in a cold box before being frozen to prevent tissue degradation. Four trapping campaigns were undertaken, and between 30 and 50 traps were set in lines which traversed the transects in the study area. Trapping success varied with each session as indicated in section 5.8. The animals were prepared for radionuclide determination as described in section 3.3.

5.4 SOIL RESULTS

The reasons behind the analysis of soil samples have already been described in detail (sections 3.4 and 4.4). Once again, the spatial and temporal variation across the site should contribute to an understanding of the transfer of radionuclides through the food chain to small mammals. The fact that the site has been undisturbed for at least the forty years since Sellafield became operational as a nuclear establishment means that the radionuclide inventory within the dunes should reflect past discharges both to the atmosphere and to the marine environment. However, the site was chosen on the basis that the radionuclide inventory contained within the dunes was primarily as a result of marine discharges being relocated to the terrestrial environment by sea to land transfer. The radionuclide inventory in the top 10 to 15 cm of the

dunes is described below as is the relevance of this distribution for plant and animal uptake. Some of the soil parameters which affect the mobility of the radionuclides are also discussed.

5.4.1 Spatial and Temporal Variation

Transects C and D were sited with the intention of providing replicate locations to examine the spatial distribution of radionuclides across the study area. The positioning of the transects parallel to the coastline was an attempt to assess spatial variation in radionuclide levels along the sand dunes, albeit over a relatively small distance. Given the proximity of the transects to the sea, BNFL, Sellafield and each other, little variation in the radionuclide distribution was expected. The results were to form a baseline from which assessments of the transfer of radionuclides to vegetation, invertebrate and small mammal species could be made.

Figures 5.2 and 5.3 present the mean activity (n=4) and standard error for ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am along each transect for samples collected during May- and late June 1993 and September 1994. The data show that the site is dominated by ^{137}Cs, $^{239+240}Pu$ and ^{241}Am, which have similar levels of activity, falling between 200 to 400 Bq kg^{-1}. ^{134}Cs and ^{238}Pu exhibit a much lower activity, by an order of magnitude, at around 30 Bq kg^{-1} for ^{238}Pu and less than 10 Bq kg^{-1} for ^{134}Cs. These lower levels for ^{134}Cs and ^{238}Pu simply reflect the fact that they are discharged to the marine environment from Sellafield at much lower levels than ^{137}Cs, $^{239+240}Pu$ and ^{241}Am (section 1.3.1). Moreover, similar levels for ^{137}Cs, $^{239+240}Pu$, and ^{241}Am were expected because sea to land transfer returns a greater proportion of the actinides than ^{137}Cs, to land (Eakins *et al.*, 1981; 1982; Howorth and Eggleton, 1988).

Table 5.2 shows that the radionuclides were similarly distributed between the two transects. Transect C contained slightly more activity around 55% of the measured concentrations. It was expected that transect C would exhibit a higher activity level than transect D given that there was greater exposure to the sea and that *A. arenaria* was more prevalent along transect C. The combined effect of increased exposure and the greater abundance of *A. arenaria* was expected to lead to a greater interception rate of radionuclides and, consequently, their relocation to the sand dunes. However, the data indicate that the differences are not as large as those anticipated originally. For example, the samples collected during June 1993 show mean values (± standard error) for transects C and D of 170 ± 1 as against 155 ± 2, and 210 ± 3 as against 190 ± 2 Bq kg^{-1} for $^{239+240}Pu$ and ^{241}Am respectively.

Figure 5.2: **^{137}Cs, $^{239+240}Pu$ and ^{241}Am activities in individual soil cores from the Sellafield sand dunes.**

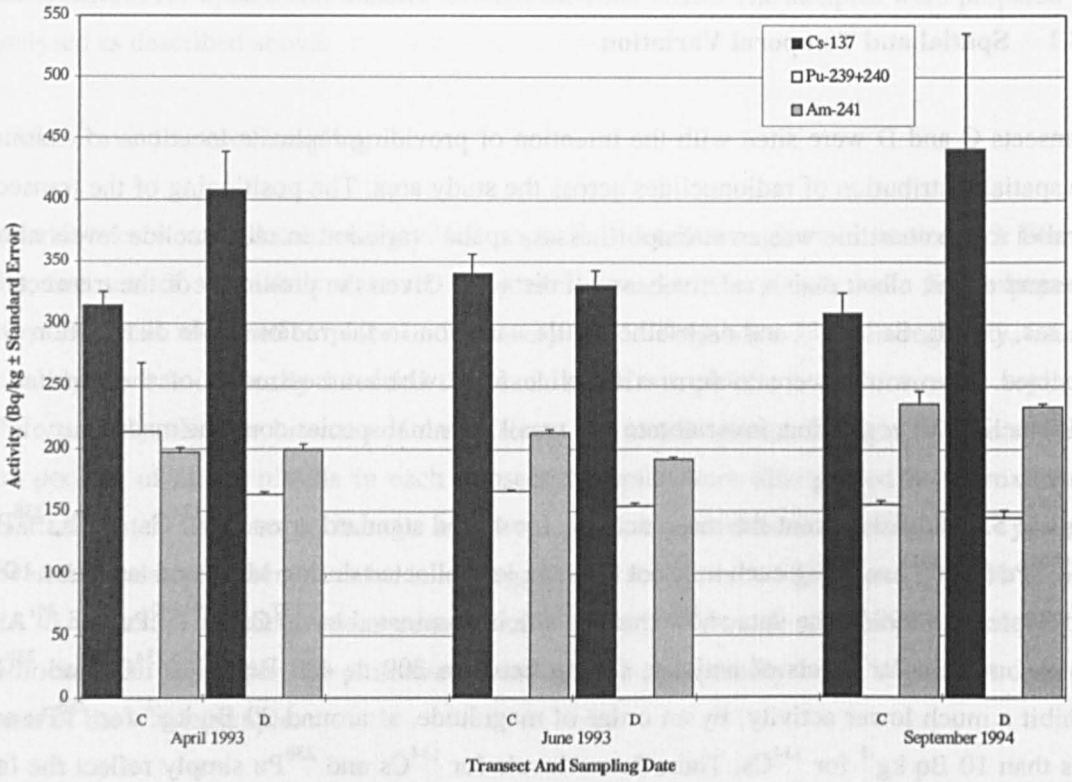


Figure 5.3: **^{134}Cs and ^{238}Pu activities in individual soil cores from the Sellafield sand dunes.**

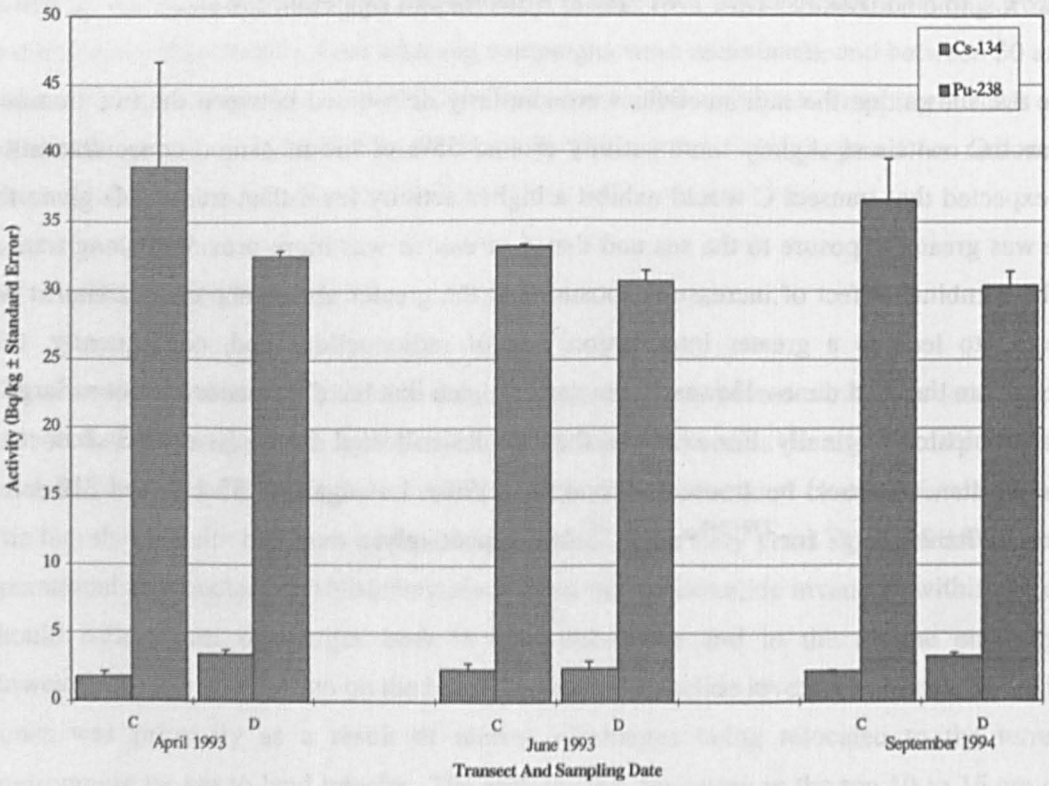


Table 5.2: Mean whole core data (Bq kg^{-1}) for the sand dunes (± s.e.), n=4.

Date	*Transect*	*^{134}Cs*	*^{137}Cs*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*
May 1993	C	1.8 ± 0.4	315 ± 12	40 ± 7.6	215 ± 55	200 ± 3
	D	3.4 ± 0.4	410 ± 31	33 ± 0.4	160 ± 3	200 ± 4
June 1993	C	2.4 ± 0.4	340 ± 16	33 ± 0.2	170 ± 1	210 ± 3
	D	2.4 ± 0.5	330 ± 12	30 ± 0.8	155 ± 2	190 ± 2
September 1994	C	2.2 ± 0.4	440 ± 18	35 ± 3.1	155 ± 3	240 ± 6
	D	1.1 ± 0.9	260 ± 62	31 ± 1.0	150 ± 6	230 ± 11

5.4.1.1 Results from spatial and temporal ANOVA tests

Table 5.3 presents the results of a two factor ANOVA which examined the temporal and spatial differences between seasons and transects. As Figures 5.2 and 5.3 and Table 5.2 have already shown, there is little variation in the data, either temporally or spatially. ^{134}Cs is the only radionuclide which exhibits a significant spatial difference between the two transects. However, LOD values were used during the ANOVA calculations and this may have artificially affected the results. In addition, significant temporal variation was only observed for ^{241}Am. Temporal variation was not expected for sand taken from this site, given that soil cores usually contain radionuclides deposited over a number of years. However, some influence of changes in the structure of the dunes was anticipated because of sand particle movement and entrapment within the dunes under the influence of wind and rain. This was however expected to be of minimal importance because the sand dunes are well stabilised by the mature communities of vegetation. During core extraction in particular a thick root mass was observed which effectively bound up the sand particles. Of the five radionuclides, four do not exhibit temporal variation; ^{241}Am does, and the differences between the samples collected in 1993 and those from 1994 are highly significant ($p<0.001$). There are no obvious reasons why ^{241}Am should behave in this way, especially since the Pu isotopes do not exhibit similar trends.

Table 5.3: **Results from a balanced two factor ANOVA. Test for spatial and temporal changes in whole core ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities respectively.**

^{134}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	2	0.823	0.412	0.39	0.682	
Transect	1	5.782	5.782	5.48	0.031*	1.02
Season*Transect	2	2.615	1.307	1.24	0.313	
Error	18	18.978	1.054			
Total	23	28.198				

^{137}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	2	7211	3605	0.95	0.406	
Transect	1	17184	17184	4.51	0.052	
Season*Transect	2	12255	6128	1.61	0.227	
Error	18	68533	3807			
Total	23	105183				

^{238}Pu:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	2	64.84	32.42	0.73	0.495	
Transect	1	109.53	109.53	2.47	0.133	
Season*Transect	2	16.23	8.11	0.18	0.834	
Error	18	798.30	44.35			
Total	23	988.90				

$^{239+240}Pu$:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	2	5857	2928	1.42	0.269	
Transect	1	2979	2979	1.44	0.246	
Season*Transect	2	2379	1189	0.57	0.573	
Error	18	37234	2069			
Total	23	48448				

^{241}Am:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	2	3176.9	1588.4	11.56	0.001***	15.48
Transect	1	528.8	528.8	3.85	0.065	
Season*Transect	2	546.5	273.3	1.99	0.166	
Error	18	2474.3	137.5			
Total	23	6726.6				

5.4.1.2 Deposition Values, Bq m^{-2}, and contribution of Chernobyl derived radiocaesium

Table 5.4 presents data for the deposition of ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am (Bq m^{-2} to 3 significant figures) for the sand dunes for the three sampling periods. The results are compared with values obtained from grassland sites in two other deposition studies. By comparing the data from Cawse (unpublished) and Jones *et al.* (1996) it is evident that the actinide levels are elevated along the sand dunes. From the latter results, the similarity in the ^{137}Cs, $^{239+240}Pu$ and ^{241}Am data is again clearly seen. Although the ^{137}Cs results are higher than the actinides, it is possible that aerial deposition of all the radionuclides occurs across the sand dunes and that Chernobyl-derived ^{137}Cs is also present. This would elevate and in a sense distort the ^{137}Cs component. The majority of ^{134}Cs released from Sellafield enters the marine environment and a proportion of this will be returned to land through sea to land transfer. However, for the purposes of estimating the contribution of ^{137}Cs from Chernobyl, it is assumed that all of the ^{134}Cs measured was also deposited from Chernobyl. Table 5.5 presents caesium data for whole soil cores collected during May 1993, decay corrected to 2nd May 1986. In addition, data for soils collected at Drigg, Cumbria in June 1986 are presented for comparison (Rudge, 1989).

Table 5.4: Mean deposition values (Bq m^{-2}) from samples collected along the transects on the sand dune site.

Location	*Sample Date*	*^{134}Cs*	*^{137}Cs*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*
C	May '93	159 ± 34	28,800 ± 835	3,590 ± 710	19,600 ± 5130	18,100 ± 635
D	May '93	286 ± 29	34,500 ± 3200	2,740 ± 95	13,800 ± 470	16,800 ± 474
C	June '93	210 ± 38	30,300 ± 1790	2,950 ± 110	14,800 ± 657	18,900 ± 523
D	June '93	179 ± 35	25,000 ± 2550	2,330 ± 250	11,600 ± 1110	14,500 ± 1500
C	Sept. '94	203 ± 41	27,800 ± 3390	1,560 ± 340	14,300 ± 2000	21,400 ± 2850
D	Sept. '94	282 ± 30	38,000 ± 9100	1,400 ± 85	13,000 ± 230	20,000 ± 750
Grassland[1]	August '87		24,670	114	912	2132
Grassland[2]	August '88			146	2061	

----- Sample not collected.
[1] Cawse (unpublished data).
[2] Jones *et al.* (1996).

The decay corrected ^{137}Cs:^{134}Cs ratio shows reasonable agreement with that measured by Rudge (1989) shortly after the Chernobyl deposition, particularly for the back transect. Comparing this ratio to the current values recorded for this site (Table 5.6), it is apparent that the Chernobyl accident contributed significantly to the deposition of caesium. Using ^{134}Cs to estimate the extent of Chernobyl fallout allows the contribution of Chernobyl-derived ^{137}Cs to

be determined. Fulker (1987) reported an average ^{137}Cs:^{134}Cs ratio of 1.6 for the environs of Sellafield immediately after the Chernobyl deposition. Applying this value to Table 5.5, an upper estimate of the measured ^{137}Cs in the whole core data originating from the Chernobyl accident can be derived and equates to a deposition value between 2,300 and 3,500 Bq m^{-2} (approximately 8 to 11% of the ^{137}Cs inventory). This is a similar level of ^{137}Cs activity to that estimated to be from Chernobyl in Lady Wood, around 11% (section 3.4.1).

Table 5.5: Caesium data for whole sand dune cores extracted during May 1993, decay corrected to 2nd May 1986.

Transect	*^{134}Cs (Bq kg^{-1})*	*^{137}Cs (Bq kg^{-1})*	*^{137}Cs:^{134}Cs*
C	18	370	24
D	36	480	14

5.4.1.3 Isotopic and Nuclide Ratios

Table 5.6 presents data for the isotopic and nuclide ratios measured in whole core samples from the dunes. The calculated ratios provide further evidence that there is little spatial or temporal variation in radionuclide activities, within the sand dunes system. Only the ^{137}Cs:^{134}Cs ratio exhibits much by way of variation. Again this is probably artificial and a consequence of the use of LOD values during the calculation for transect D, September 1994. The short count times employed for the quantification of the radionuclides, ^{137}Cs and ^{241}Am, frequently failed to detect or measure ^{134}Cs. The use of LOD values will result in an overestimation of the true ratio, and is the most plausible cause for the discrepancy in September 1994 figures. Furthermore, the temporal variation in the mean ratios for ^{137}Cs:^{134}Cs is insignificant ($p>0.05$) and therefore the observed fluctuations are probably related to sampling.

Table 5.6: Mean isotopic and nuclide ratios for sand dune soils (± standard error) from May 1993 sampling, n=4.

Sample Date	*Transect*	*^{137}Cs:^{134}Cs*	*^{238}Pu:$^{239+240}Pu$*	*^{137}Cs:$^{239+240}Pu$*	*^{137}Cs:^{241}Am*
May '93	C	128 ± 35	0.19 ± 0.01	1.3 ± 0.5	1.3 ± 0.4
	D	122 ± 12	0.20 ± 0.00	2.5 ± 0.2	2.0 ± 0.1
June '93	C	156 ± 23	0.20 ± 0.00	2.1 ± 0.2	1.6 ± 0.1
	D	162 ± 43	0.20 ± 0.00	2.2 ± 0.1	1.7 ± 0.1
September '94	C	140 ± 11	0.22 ± 0.01	1.8 ± 0.1	1.2 ± 0.1
	D*	290 ± 175	0.21 ± 0.00	2.4 ± 0.5	1.6 ± 0.2

* LOD values used during calculations.

It is notable that the ratios for transects C and D are similar in all cases, except for the aforementioned September 1994 ^{137}Cs:^{134}Cs values. This is a reflection of the proximity of the two transects to each other, the sea and Sellafield. Given that the sea is likely to be the principal source of radioactivity, this is perhaps not surprising.

The deposition values and isotopic/nuclide ratios are reasonably consistent with data provided by other authors. For example, ^{238}Pu:$^{239+240}Pu$ ratios varied between 0.10 to 0.13 in soils and 0.18 to 0.23 in air for coastal sites investigated by Pattenden *et al.* (1980), although the ^{137}Cs:$^{239+240}Pu$ ratios for air samples are much higher, at between 15 and 49, compared to the soil data from this study. Toole *et al.* (1990) reported declining activities inland along 5 km transects and values between 0.37 and 4.0 for ^{137}Cs:$^{239+240}Pu$, 0.26 to 0.36 for ^{238}Pu:$^{239+240}Pu$ for samples of beach sand and offshore sediment. These closely resemble those reported in this study.

5.4.2 Radionuclide Distribution in Vertical Soil Core Profiles

Data from the 9 cm sectioned cores are presented in Figures 5.4 and 5.5. The levels of activity are similar to those shown by the whole core samples, with ^{137}Cs, $^{239+240}Pu$ and ^{241}Am exhibiting values in the range 200 to 300 Bq kg^{-1}. ^{134}Cs is again <10 Bq kg^{-1} and ^{238}Pu is between 25 and 30 Bq kg^{-1}. The interesting feature of these data is the increase in activity for each radionuclide with depth. This is not surprising given the very low amount of organic matter and the particle size distribution across the site (Table 5.7). The sand core profiles therefore contradict the expected radionuclide distribution which has been reported to decline with soil depth (Cawse, 1983; Eakins *et al.*, 1981). These data also disagree with the results for the soil cores extracted from Lady Wood (section 3.4.2). Possible reasons are given below.

The 12 cm sectioned cores were collected on two occasions during the study period, in July 1993 and September 1994. Inspection of Figures 5.4 and 5.5 and the raw data suggests that there is little evidence of either temporal or spatial variation. This was expected from the whole core results (section 5.4.1). Lack of cohesion caused problems whilst extracting the 12 cm cores for sectioning, so it was impractical to obtain the longer 50 cm cores described in the previous two chapters. However, based on the 12 cm sectioned core data, it is postulated that radionuclides would be present at depth because of leaching, as exemplified in Figures 5.4 and 5.5.

Figure 5.4: ^{137}Cs, $^{239+240}Pu$ and ^{241}Am activity in sand dune sectioned soil cores.

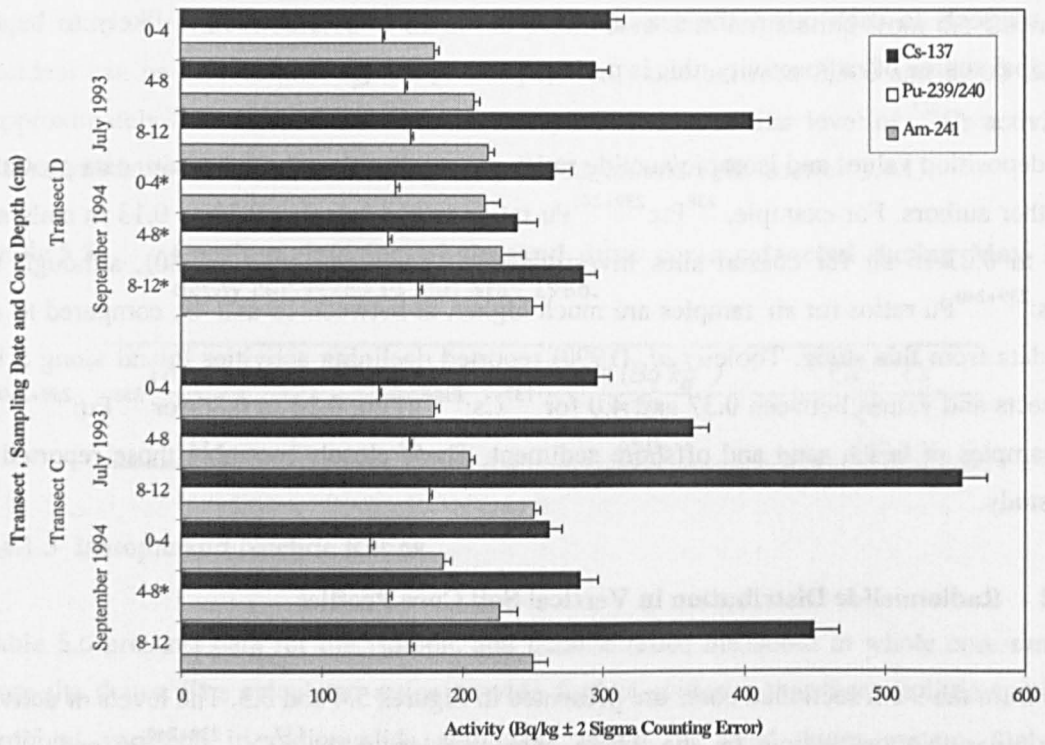
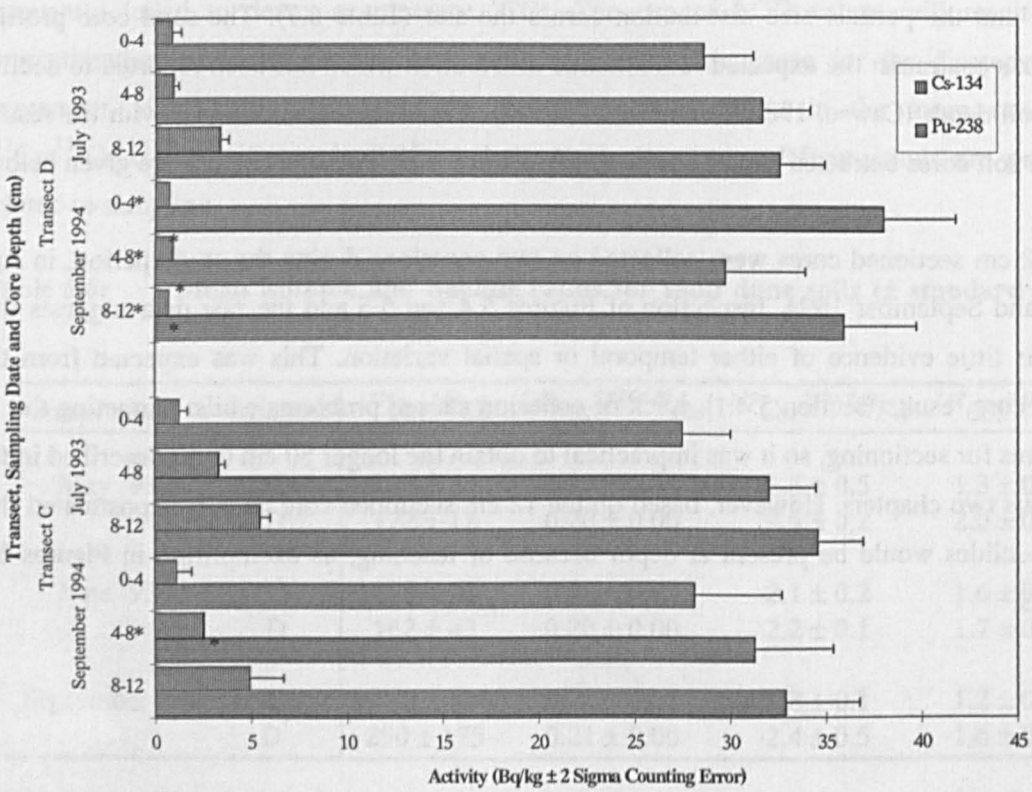


Figure 5.5: ^{134}Cs and ^{238}Pu activity in sand dune sectioned soil cores.



* LOD values used during the calculations.

5.4.2.1 Soil Characterisation

Although the scope of this study precluded detailed examination of the parameters affecting radionuclide mobility in soil, even a limited assessment would be an advantage in explaining the radionuclide distribution. Consequently, Table 5.7 presents data for several of the more important parameters that control radionuclide mobility. These include: organic matter content, particle size distribution and pH (section 1.4.1). These parameters strongly influence the rate of radionuclide migration in a soil profile although their effects will be influenced by climate and bioturbation.

The interaction of radionuclides with soil is complex (section 1.4.1) and there is no one dominant parameter. However, the passage of radionuclides through soil is strongly dependent on the amount of organic matter and the percentage of clay minerals present. Both provide binding sites for radionuclides and there is a strong affinity between them, as exemplified in section 3.4.2 and by a number of studies (Burmann, 1993; Cook *et al.*, 1984; Kuhn *et al.*, 1984). As indicated in Table 5.7, the organic matter and clay content of the sand dunes is low, being less than 2%. This is consistent with how sand dunes form and develop, and with their poor nutrient status. It is well documented that soils with a high clay content produce the greatest fixation of ^{137}Cs (Squire and Middleton, 1966) and therefore low ^{137}Cs levels were expected for the sand dunes (Table 5.2). Furthermore, because >98% of the particles are 200

Table 5.7: Soil parameter results for the sand dunes (± standard deviation), n=2.

Sampling Location			*Particle Size Distribution*		
	pH	*% LOI*	*Sand (%)*	*Silt (%)*	*Clay (%)*
Transect C	7.5	1.4 ± 0.2	98.8 ± 0.2	0.7 ± 0.2	0.5 ± 0.01
Transect D	7.3	1.5 ± 0.4	98.6 ± 0.3	0.9 ± 0.2	0.5 ± 0.02
C 0-3cm	7.7	1.6 ± 0.2	98.9 ± 0.1	0.6 ± 0.1	0.6 ± 0.1
C 3-6cm	7.6	1.4 ± 0.2	98.1 ± 0.7	1.2 ± 0.6	0.7 ± 0.1
C 6-9cm	7.5	1.1 ± 0.3	98.8 ± 0.02	0.6 ± 0.02	0.7 ± 0.01
D 0-3cm	7.6	1.8 ± 0.0	n/a	n/a	n/a
D 3-6cm	7.5	1.7 ± 0.1	n/a	n/a	n/a
D 6-9cm	7.3	1.3 ± 0.3	n/a	n/a	n/a
Whitford Burrows	7.5	2.0 ± 0.2	n/a	n/a	n/a
0-3cm	7.2	2.4*	n/a	n/a	n/a
3-6cm	7.3	2.0*	n/a	n/a	n/a
6-9cm	7.6	1.8*	n/a	n/a	n/a
Ness Gardens	4.3	8.7 ± 1.5	n/a	n/a	n/a
0-3cm	6.4	8.0*	n/a	n/a	n/a
3-6cm	6.6	7.6*	n/a	n/a	n/a
6-9cm	6.6	7.4*	n/a	n/a	n/a

* Only one sample collected.
n/a - Sample not analysed.

μm or larger (coarse sand), rainwater will percolate through the soil quickly, and will leach radionuclides down the soil column. Moreover, rapid water flow through the sand may flush radionuclides, reducing their residence time, and the time available for sorption to clay and organic matter. The soil pH was reasonably constant across the site, at around neutral. From the soil core profiles and the limited soil parameter data, it is anticipated that the plant uptake of radionuclides via the roots is likely to be low.

5.4.4 Comparison of Sand Samples Collected in Wales and Soils Collected from the Cheshire with the Sand Dune Samples

Table 5.8 compares the soil activity data from Ness Gardens, Cheshire with the sand samples collected from Wales and Sellafield. As expected, the data clearly show that the radionuclide contamination of the sand dunes around Sellafield is substantially greater than either of the two reference sites. For example, the ^{137}Cs values are over 300 times higher than the Welsh

Table 5.8: A comparison of the ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities (Bq kg^{-1}) in soil cores extracted from the Sellafield sand dunes and the Cheshire and Welsh reference sites.

Transect/Site	^{134}Cs	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
Whole Cores					
C	2.2 ± 0.1	308 ± 17	33.2 ± 0.2	170 ± 1	240 ± 10
D	3.3 ± 0.3	440 ± 94	30.0 ± 0.8	150 ± 2	230 ± 3
Wales	<0.05	0.9 ± 0.1	0.35 ± 0.13	1.6 ± 0.7	<0.11
Cheshire	<0.15	12.0 ± 1.5	0.02 ± 0.01	0.4 ± 0.1	0.25 ± 0.05
Sectioned Cores					
C 0-3 cm	<0.7	260 ± 13	38 ± 2	150 ± 2	220 ± 10
C 3-6 cm	<0.9	240 ± 14	30 ± 1	150 ± 2	230 ± 16
C 6-9 cm	<0.7	290 ± 10	36 ± 2	170 ± 3	250 ± 7
D 0-3 cm	1.2 ± 0.8	260 ± 9	28 ± 1	135 ± 2	190 ± 5
D 3-6 cm	<2.6	280 ± 13	31 ± 1	150 ± 2	230 ± 13
D 6-9 cm	4.9 ± 1.8	450 ± 18	33 ± 1	160 ± 3	250 ± 11
Wales 0-3 cm	<0.3	1.2 ± 0.3	0.2 ± 0.1	0.9 ± 0.1	0.3 ± 0.3
Wales 3-6 cm	<0.3	0.7 ± 0.2	0.5 ± 0.1	2.3 ± 0.2	<0.3
Wales 6-9 cm	1.1 ± 0.5	2.3 ± 0.2	0.2 ± 0.1	1.0 ± 0.1	0.8 ± 0.4
Cheshire 0-3 cm	<0.1	7.4 ± 0.3	0.4 ± 0.1	6.0 ± 1.0	1.1 ± 0.3
Cheshire 3-6 cm	0.3 ± 0.1	18.0 ± 0.7	4.9 ± 0.4	6.4 ± 0.4	10 ± 0.5
Cheshire 6-9 cm	<0.9	17.0 ± 1.2	n/a	n/a	6.9 ± 1.5

< The reported mean was calculated from LOD values.
Whole core data reported as mean ± standard error, n=4. Sectioned cores reported as mean ± 2σ counting error.
Sand dune data taken from September 1994, Cheshire data from August 1993, and Welsh data from August 1994.
n/a Sample not analysed.

site and over 30 times higher than the Cheshire site. Similar differences can be seen for ^{241}Am (over 150 times higher) and for ^{134}Cs (over 10 times higher). For all the radionuclides, with the exception of ^{134}Cs, the differences between the Sellafield sand dunes and the two reference sites are highly significant ($p<0.001$) using a One-way Analysis of Variance, although the ^{134}Cs values were complicated by the presence of a large number of LOD values. This makes accurate assessment of ^{134}Cs on the three sites difficult. However, it is obvious that the activity levels around Sellafield are much greater than at the two reference sites. This then provides an indication of background levels of anthropogenic radionuclides released in the late 1950s, early 1960s, from the Chernobyl accident in 1986 and from dispersion of Sellafield discharges. In addition, the Welsh site provides measurements that can be used to estimate the reduced levels of radionuclides relocated to the terrestrial environment by the process of sea to land transfer.

5.4.5 Summary of sand dune soils

Core samples from the sand dune ecosystem show evidence of the accumulation of radionuclides principally from sea to land transfer of the aerosol that derives form re-suspension of the sea surface microlayer; but also from limited atmospheric deposition. Accumulated deposits lie within the range 160 to 280 Bq m^{-2} (^{134}Cs), 25,000 to 38,000 Bq m^{-2} (^{137}Cs), 1,400 to 3,600 Bq m^{-2} (^{238}Pu), 11,600 to 19,600 Bq m^{-2} ($^{239+240}Pu$) and 14,500 to 21,400 Bq m^{-2} (^{241}Am). These inventories are consistent with the relative deposition rates for actinides compared to ^{137}Cs for sea to land transfer. A similar spatial distribution was observed for all radionuclides despite differences in the vegetation cover. There was little evidence of any temporal changes over the study period.

The sand dune inventory of plutonium is characterised by ^{238}Pu:$^{239+240}Pu$ ratios in the range 0.19 to 0.21. These are comparable to measurements reported previously for similar ecosystems and to the marine discharge ratio of the two plutonium isotopes which occurred during the 1970s. There is evidence that the ^{137}Cs deposit is influenced by deposition from the Chernobyl accident. Assuming that all the ^{134}Cs present derives from Chernobyl, it is estimated that 2,300 to 3,500 Bq m^{-2} (8 to 11%) of the ^{137}Cs is of this origin.

Data from the sectioned soil cores show greater levels of activity at depth. This is attributed to the leaching of radiocaesium in percolating drainage water accentuated by the coarse texture (>200 μm) and lower organic carbon/ clay mineral content of the sand substrate. The low cation exchange capacity of sand accentuates the downward migration of radionuclides compared to conventional soil. The accretion of sand within the dune formation enhances this process.

5.5 VEGETATION

The role of *A. arenaria* in stabilising sand dunes and its ability to accrete particles around the base of established plants has already been stated. The interest in *A. arenaria* was to determine the level of influence that this species exerts upon the accumulation of radionuclides along the sand dunes. *A. arenaria* has very few natural enemies as the structure of its leaves discourages herbivory. Therefore it is improbable that *A. arenaria* forms part of the diet of the invertebrate and small mammal species inhabiting the dunes. The role of *A. arenaria* in the food chain transfer of radionuclides was assumed to be that of acquiring the contaminants from sea spray. This would either be by direct deposition on the leaf surface or by affecting the airflow such that the sand particles and other fine sediments are deposited from the air directly on to the sand surface or on to other species of vegetation. It was assumed that any radionuclides deposited on to the leaf surface of *A. arenaria* would be washed off under suitable climatic conditions or could transfer to the fur or body surface of fauna. *A. arenaria* was therefore sampled at regular intervals throughout the study period to determine any temporal and spatial differences along the dunes.

In contrast, *F. rubra* was assumed to be a very important component of the diet of the small mammals and herbivorous insects. *F. rubra* was therefore sampled at regular intervals to assess any temporal changes in its radionuclide burden. Since it was not possible to sample all the dietary components of the small mammals, *F. rubra* was used as an indicator for the vegetation component of the diet as prompted by Churchfield and Brown (1987), Evans (1973) and Hansson (1971). During the study it was not always possible to collect live *F. rubra* and so some samples of senescent material were collected. These were analysed separately. In addition, when sufficient quantities of annuals were present, composite samples were collected and analysed. These comprised any of the species listed in Table 5.1 but were dominated by *Plantago lanceolata, Plantago maritima* and *Taraxacum officinale*, all of which are reported to be in the diet of the small mammals.

5.5.1 Spatial and Temporal Variation

Research has shown how radionuclides in the staple diet of herbivorous animals are transferred and, indeed, reflected in the animals themselves (Holm and Rioseco, 1987; Liden and Gustafsson, 1967). In this study, it was anticipated that temporal variation would occur in the radioactivity on the vegetation as the growth cycle advanced during the course of a year. However, and perhaps more significantly, it was also expected that the concentrations of radionuclides adhered to the external surface of the leaves would change with climatic conditions. Given that the underlying sand contains little organic matter and clay minerals to bind radionuclides, their uptake through root absorption processes was not expected to be very

important on this site. Conversely, with the nature of the deposition, it was expected that radionuclides would become adhered to the external leaf surface and therefore available for accidental ingestion.

5.5.2 Results for *F. rubra*

Mean activities (n=2 to 4) and standard errors for live *F. rubra* are presented in Figures 5.6 to 5.9. Because of measurement difficulties, the results for ^{134}Cs have been omitted from the following discussion. ^{137}Cs levels are approximately double those reported for ^{241}Am, with the Pu isotopes considerably lower at approximately one tenth the level of ^{241}Am. For example, ranges of 20 to 70 Bq kg^{-1}, 10 to 30 Bq kg^{-1} and 0.5 to 3.5 Bq kg^{-1} were recorded for ^{137}Cs, ^{241}Am and $^{239+240}Pu$ respectively. ^{238}Pu levels were typically less than 0.5 Bq kg^{-1}. These are significantly lower than those reported for the sand samples.

Figures 5.6 to 5.9 indicate that there is actually very little temporal variation. Furthermore, in all cases where comparable samples of the same species have been obtained along both transects, the activity level along transect D was 5 to 40% lower than transect C. Differences in the region of 10% were most prevalent. This indicated that spatial variation across the site was also low. Low spatial variation was not unexpected given the proximity of the two transects and based upon the soil core results (section 5.4.1).

Table 5.9 presents the results from a balanced two factor ANOVA examining spatial and temporal variation for ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am. The results clearly indicate that there are temporal differences in the data and that for all radionuclides, except ^{238}Pu, there are spatial differences between the two transects. The former was expected given that the radioactivity present on the foliage should be dependent upon previous deposition events. These events will be influenced by climatic conditions such as rainfall, wind direction and speed. It is thought that variation in climatic conditions is the primary cause for temporal change because, based on Figures 5.6 to 5.9, there is little evidence of a cyclic pattern attributable to seasonal changes in plant growth for any radionuclide. Furthermore, comparison of the data with the calculated LSD values suggests that there is significant variation almost on a monthly basis which indicates that climatic conditions immediately antecedent to sample collection were important. A limited investigation to assess the effects of prominent climatic conditions (rainfall, wind direction, wind speed and relative humidity) on the deposition of ^{137}Cs and ^{241}Am to *F. rubra* and *A. arenaria* was attempted using meteorological data obtained from the Sellafield weather station. By taking individual weather components, only limited correlations were possible for the deposition of ^{241}Am on *F. rubra*. However, it is extremely unlikely that the deposition of radionuclides is influenced by a single climatic condition; more probably it is due to a combination of conditions.

Figure 5.6: **^{137}Cs activity in live *F. rubra* collected from the Sellafield sand dunes.**

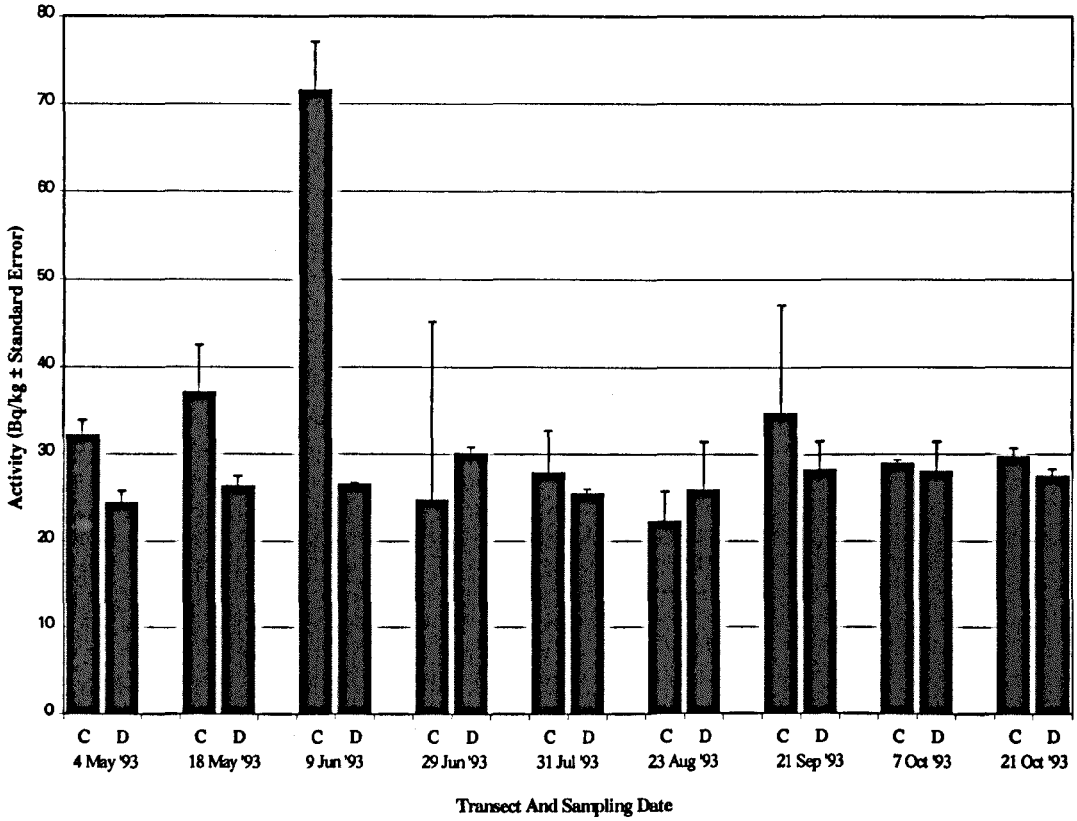


Figure 5.6: **(Continued.)**

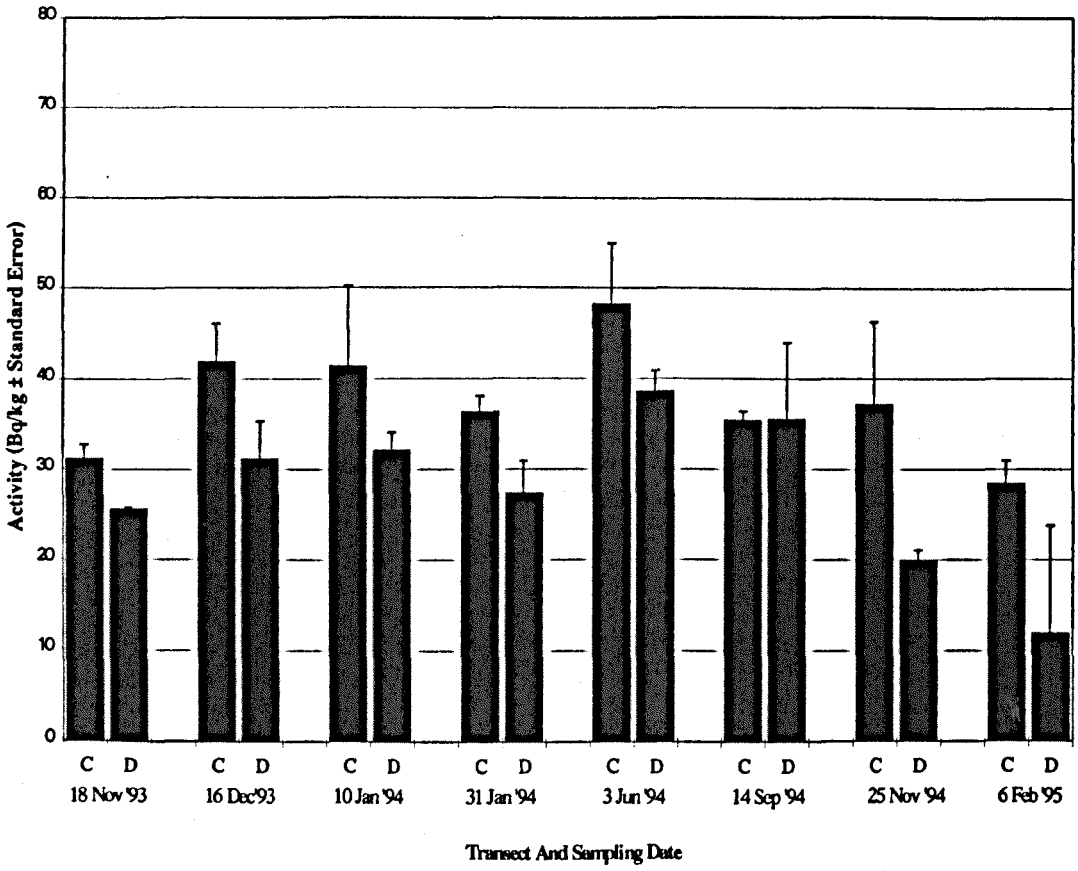


Figure 5.7: ^{238}**Pu activity in live *F. rubra* collected from the Sellafield sand dunes.**

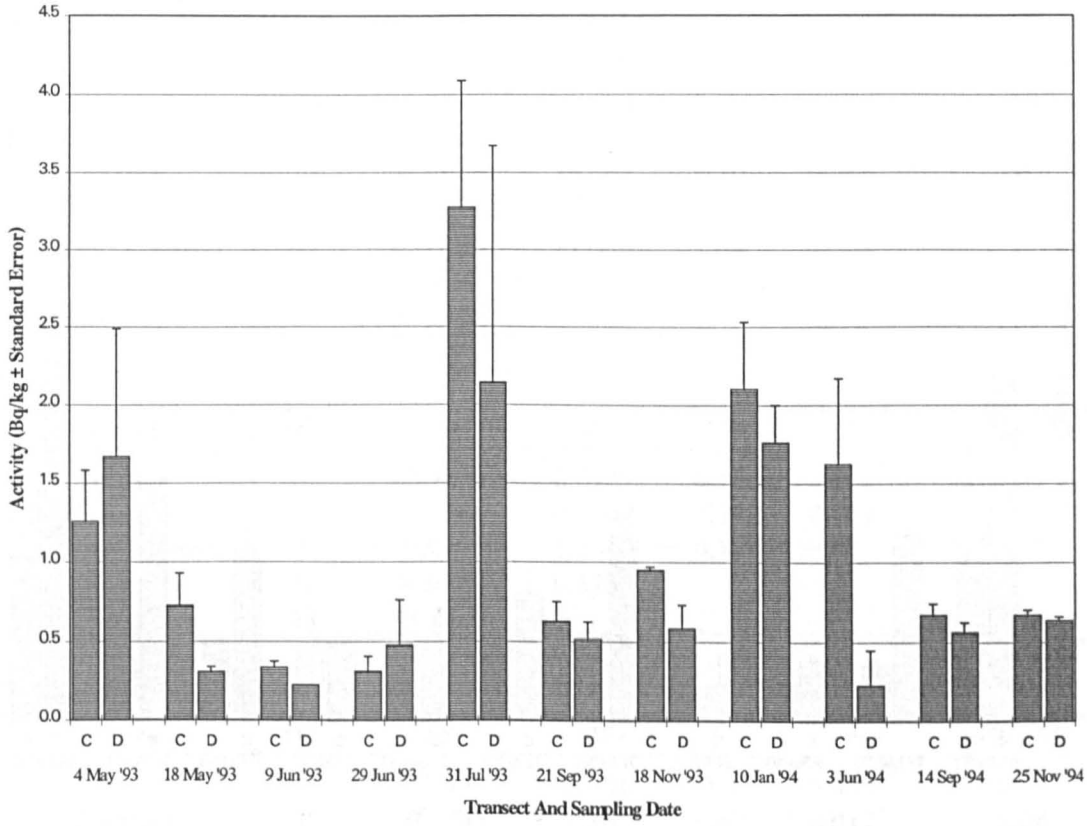


Figure 5.8: $^{239+240}$**Pu activity in live *F. rubra* collected from the Sellafield sand dunes.**

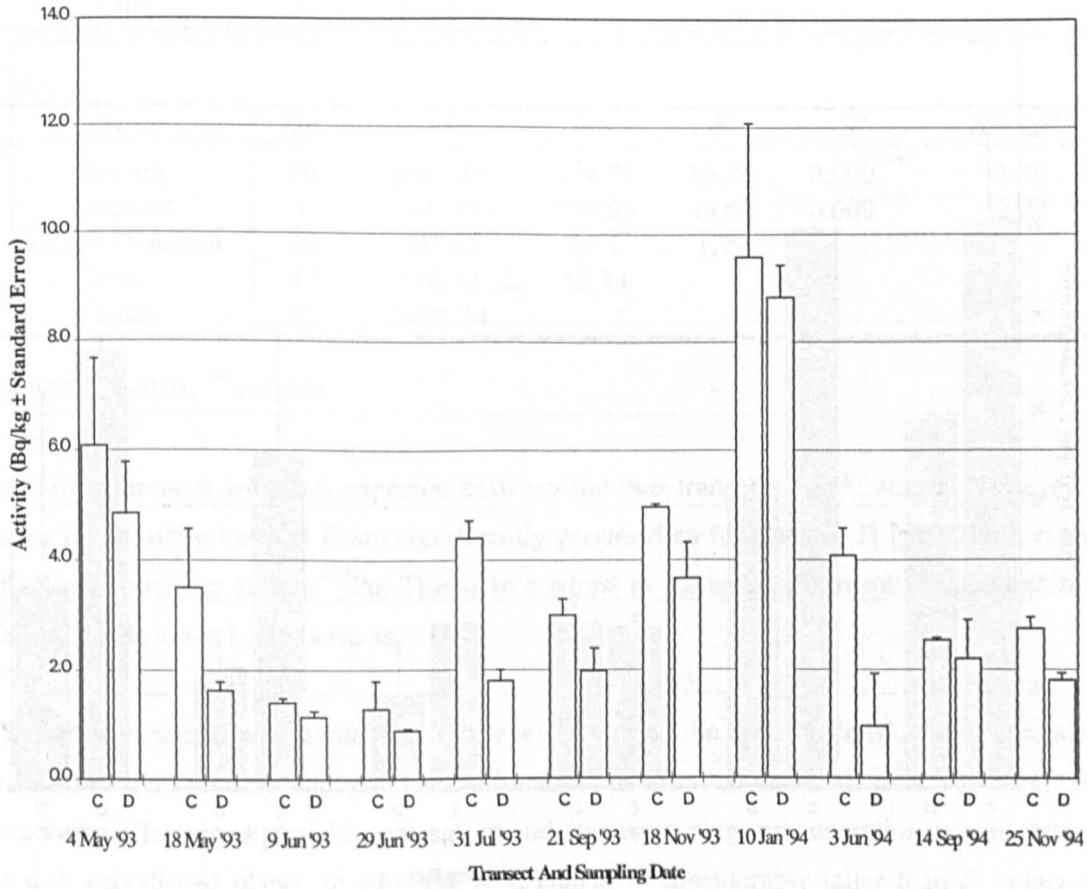


Figure 5.9: ^{241}Am activity in live *F. rubra* collected from the Sellafield sand dunes.

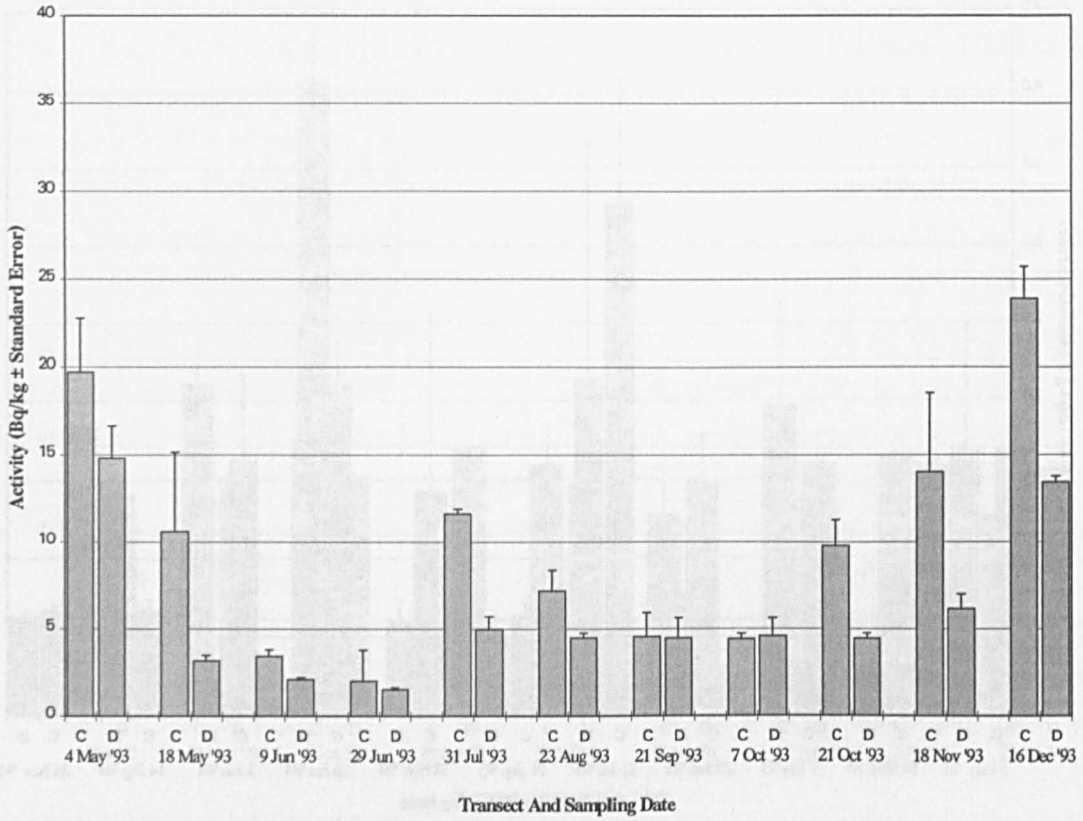


Figure 5.9: (Continued.)

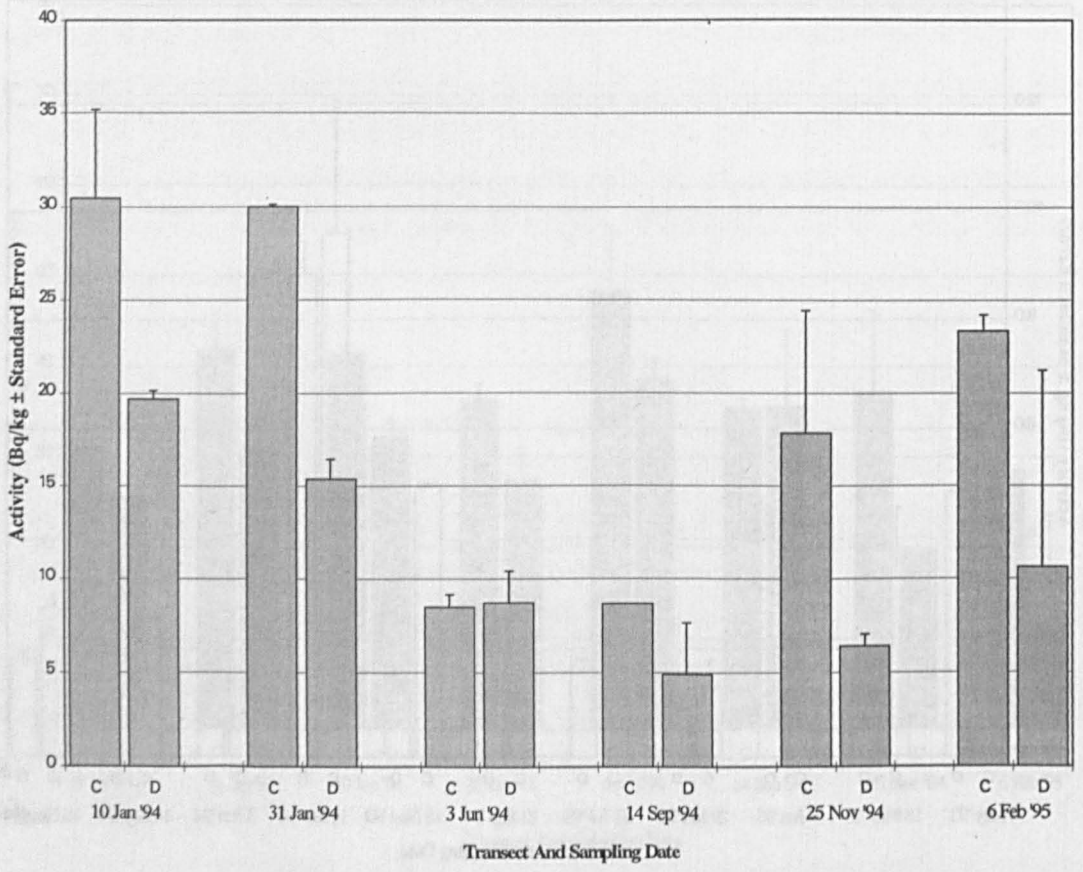


Table 5.9: **Results from a balanced two factor ANOVA - test for spatial and temporal changes in *F. rubra* ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities respectively.**

^{137}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	20	3233.48	161.67	2.50	0.006**	3.67
Transect	1	1123.99	1123.99	17.38	0.000***	5.19
Season*Transect	20	2351.96	117.60	1.82	0.051	
Error	42	2716.97	64.69			
Total	83	9426.39				

^{238}Pu:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	10	22.9286	2.2929	6.11	0.000***	0.47
Transect	1	0.8209	0.8209	2.19	0.153	
Season*Transect	10	1.2340	1.2340	0.12	0.33	
Error	22	8.2606	0.3755			
Total	43	33.2440				

$^{239+240}Pu$:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	10	221.499	22.150	15.32	0.000***	0.80
Transect	1	9.570	9.570	6.62	0.017*	0.84
Season*Transect	10	7.385	0.739	0.74	0.51	
Error	22	31.813	1.446			
Total	43	270.268				

^{241}Am:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	20	3890.33	194.52	15.76	0.000***	1.60
Transect	1	550.55	550.55	44.60	0.000***	2.27
Season*Transect	20	441.95	22.1	1.79		
Error	42	518.42	12.34			
Total	83	5401.24				

* $p<0.05$, ** $p<0.01$, *** $p<0.001$.

Spatial differences were not expected between the two transects for *F. rubra*. However, *F. rubra* levels along transect C are significantly greater than for transect D ($p<0.01$) in respect of all radionuclides except ^{238}Pu. This is in contrast to the sand core results which exhibited similar levels along both transects.

These differences can be explained in two ways, firstly, the abundance of *A. arenaria* along transect C and secondly, topographical differences in the sand dunes. *A. arenaria* is reported to be very effective at particle interception and at encouraging sand accretion around the base of well established plants. In addition, *A. arenaria* is considerably taller than *F. rubra* and

therefore air flowing at a greater height above the ground is influenced by its presence. This effectively means that wind flowing across the dunes interacts with *A. arenaria* before reaching transect D. Furthermore, there is evidence that the deposition rate drops rapidly with distance from the source of sea spray, even over a few hundred metres. Subsequently, under the influence of rain, radionuclides will be washed down from *A. arenaria* on to the lower vegetation, in this case *F. rubra*, and sand.

The influence of *A. arenaria* and the shape of the sand dunes will also cause the air to flow along the beach and be forced up over the front of the dunes, thereby passing over transect D at a greater height and velocity. This could reduce the direct deposition along transect D sufficiently to reduce the levels of radionuclides to those observed. Figures 5.6 to 5.9 show these differences but, with the exception of samples taken on 9 June 1993, they are not large even though the ANOVA indicates that they are highly significant. It is believed that the changes in air flow over the dunes are likely to be sufficient to cause the observed decline in activity along transect D.

Unlike the radionuclides deposited on vegetation through sea to land transfer, sand particles will contain only a proportion of radioactivity arising from this process. The sand originated from the sea and will have been contaminated with radioactivity prior to its deposition by the action of waves and wind. Much of this sand will have been physically relocated by wind. In particular, sand from the front of the dune system will be displaced towards the rear, leading to the classic sand dune formation, with a short and steep windward slope and a relatively long and gentle leeward slope. This means that sand cores taken from either transect will contain material originating from the sea, deposited and transferred across the dunes. Given that material to a depth of 12 cm was analysed, it is thus not surprising to discover similar radionuclide levels throughout the sand.

5.5.3 Results for live and senescent *F. rubra*

Sufficient live and senescent material for a direct comparison in the activity of the vegetation for the radionuclides ^{137}Cs and ^{241}Am was collected on four occasions, two in May 1993, and then in June 1993 and January 1994. On other occasions, only live material was collected because there was insufficient senescent *F. rubra* for analytical purposes. The results for the senescent samples are presented in Figures 5.10 and 5.11. Comparing the activity of these with the live *F. rubra,* given in Figures 5.6 and 5.9 for ^{137}Cs and ^{241}Am respectively, it can be seen that differences occur depending upon the radionuclide. For ^{137}Cs both the live and senescent material contain in the order of 20 to 40 Bq kg^{-1} and there is no apparent difference between the two.

Figure 5.10: ^{137}Cs activity in senescent *Festuca rubra* collected from the Sellafield sand dunes.

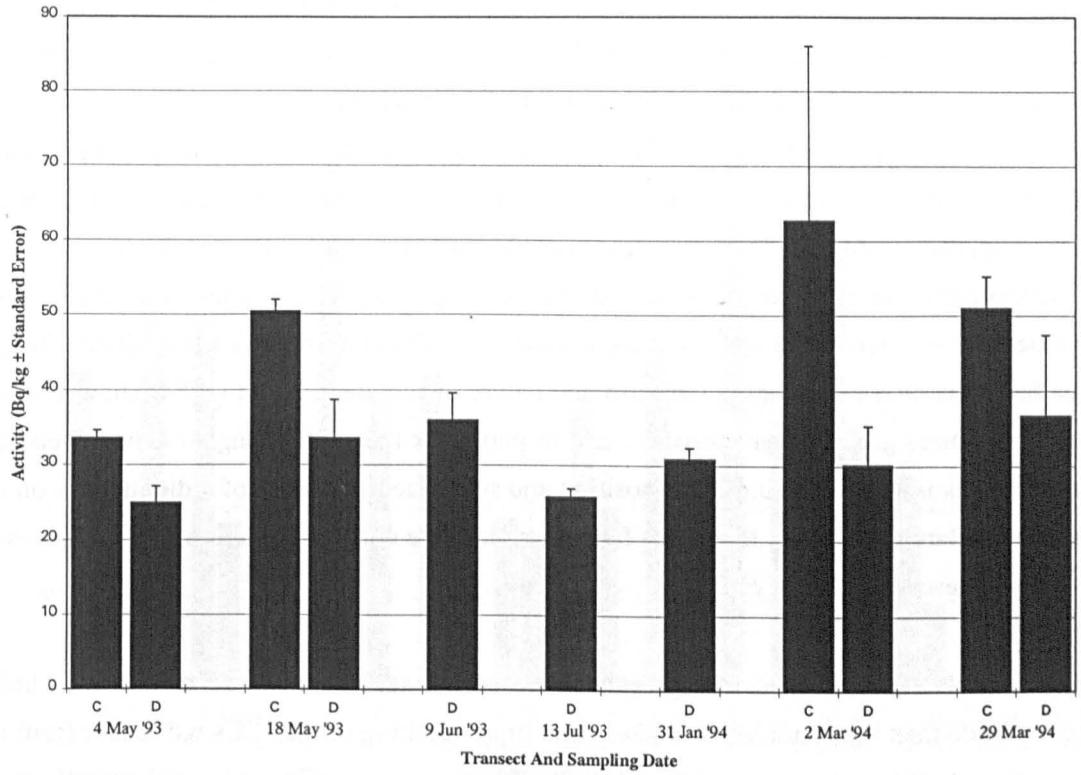
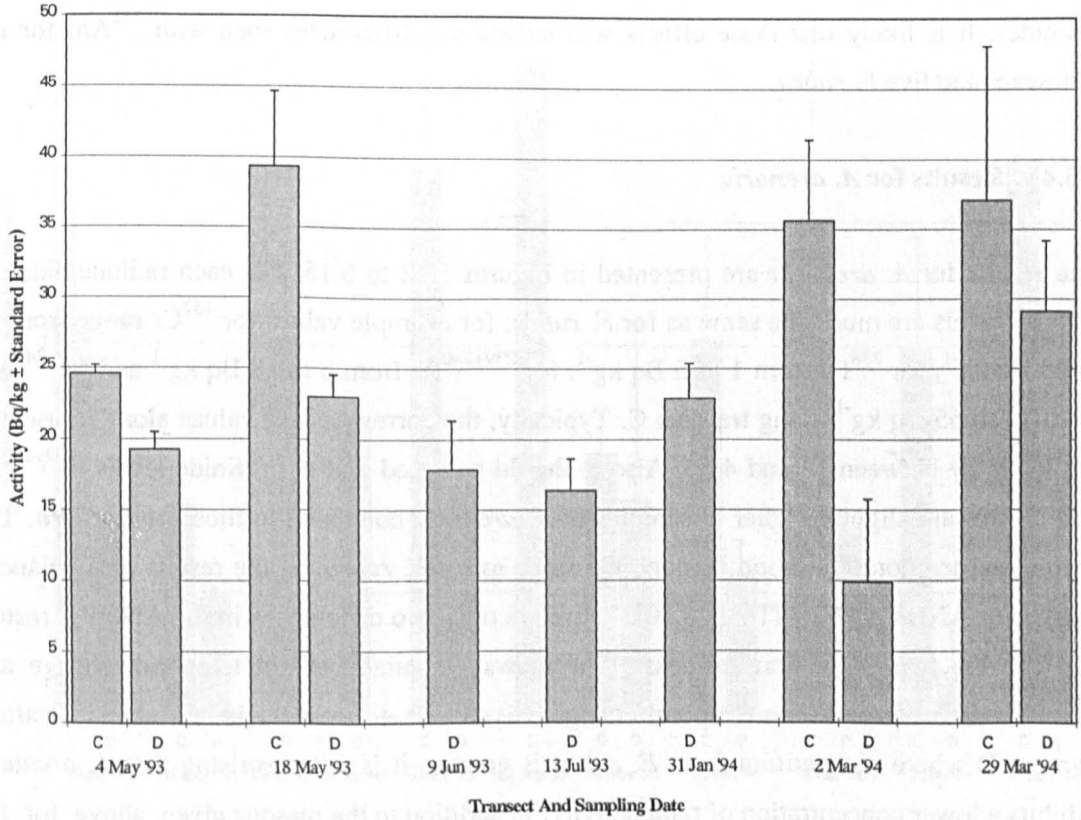


Figure 5.11: ^{241}Am activity in senescent *Festuca rubra* collected from the Sellafield sand dunes.



^{241}Am exhibits a different pattern to ^{137}Cs. For live material, ^{241}Am levels are between 10 and 50% lower than those of senescent tissue. This could be due to the fact that as *F. rubra* senesces ^{241}Am, which is not utilised by plants, could be relocated within the plant to the dying tissues, just as trees and other species of vegetation remove toxins from the healthy living tissues prior to leaf fall. In addition, most of the dead material was collected during the winter and early spring as *F. rubra* died back. During the winter it was anticipated that more radionuclide material would be transferred back to the terrestrial environment due to changes in the climatic conditions. Therefore, it is feasible, and more probable, that the results would be influenced by elevated deposition across the site, especially as the vegetation tends to form an extensive mat across the soil when die-back occurs. This will increase the available surface area for deposition by changing the form and nature of the plant. Allen (1984) showed that a number of plant growth characteristics, and in particular the contact angle of the vegetation, were important in determining the deposition and subsequent retention of radionuclides on the surface of plants. However, the above fails to explain the equal distribution of ^{137}Cs between live and senescent leaves of *F. rubra*.

^{137}Cs will also be deposited to a greater extent on the senescent *F. rubra*; however, being more mobile than the actinides, it is likely that more leaching of the ^{137}Cs will occur from the vegetation. In this case, the greater surface area of the senescent material will actually work against its accumulation and the senescent material will undergo more extensive leaching. In addition, the mobility of the ^{137}Cs and its analogous behaviour to potassium means that a greater proportion will be taken up by the plants via the roots and stomata compared to the actinides. It is likely that these effects will negate the differences seen with ^{241}Am for the senescent and live *F. rubra*.

5.5.4 Results for *A. arenaria*

The results for *A. arenaria* are presented in Figures 5.12 to 5.15. For each radionuclide the activity levels are much the same as for *F. rubra*; for example values for ^{137}Cs range from 18 to 68 Bq kg^{-1}, for ^{238}Pu from 1 to 5 Bq kg^{-1}, for $^{239+240}$Pu from 8 to 18 Bq kg^{-1} and for ^{241}Am from 14 to 65 Bq kg^{-1} along transect C. Typically, the corresponding values along transect D are lower by between 10 and 40%. Also it should be noted that the actinide levels ($^{239+240}$Pu and ^{241}Am) are slightly higher in samples of *A. arenaria* compared to those of *F. rubra*. The spatial and temporal variation is much the same as for *F. rubra*, as the results of a balanced two factor ANOVA show (Table 5.10). There are only two differences in the ANOVA results between the two vegetation species: ^{238}Pu shows a spatial, but not temporal, change and $^{239+240}$Pu does not exhibit a temporal change. Given that *A. arenaria* is less abundant along transect D where competition from *F. rubra* is greater, it is not surprising that *A. arenaria* exhibits a lower concentration of radioactivity. In addition to the reasons given above for the

Figure 5.12: ^{137}Cs activity in *A. arenaria* collected from the Sellafield sand dunes.

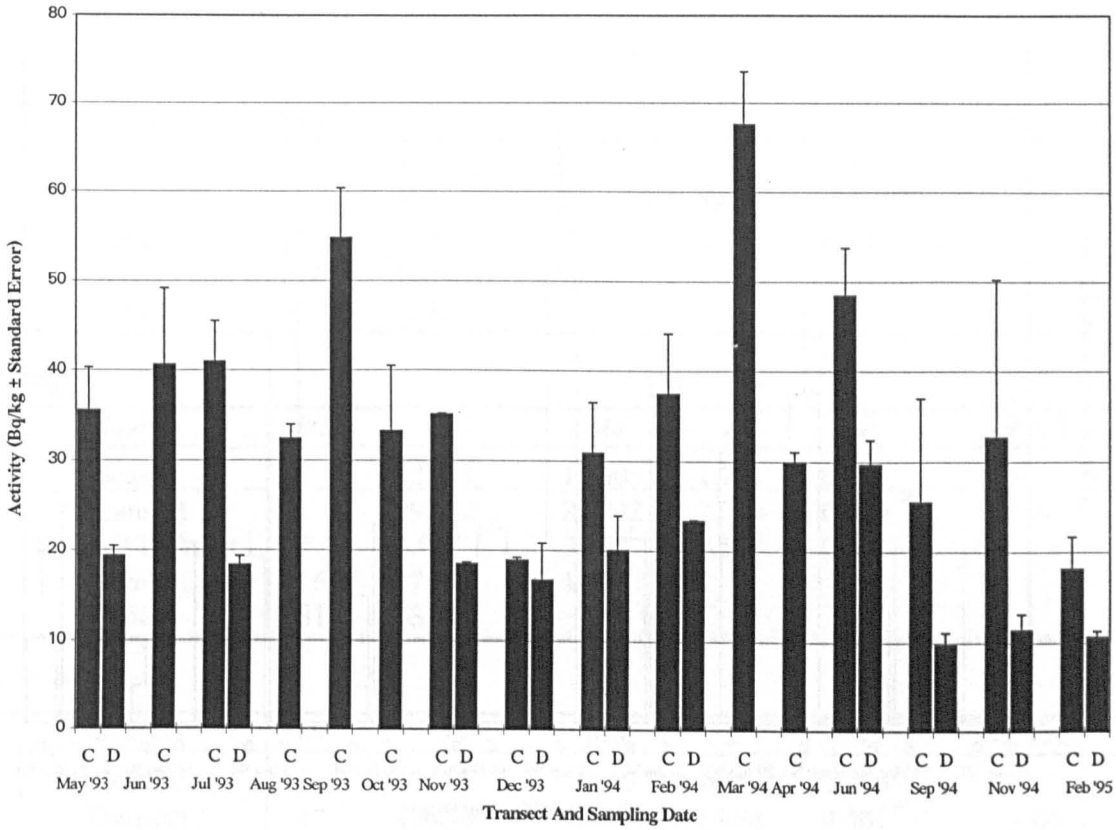


Figure 5.13: ^{238}Pu activity in *A. arenaria* collected from the Sellafield sand dunes.

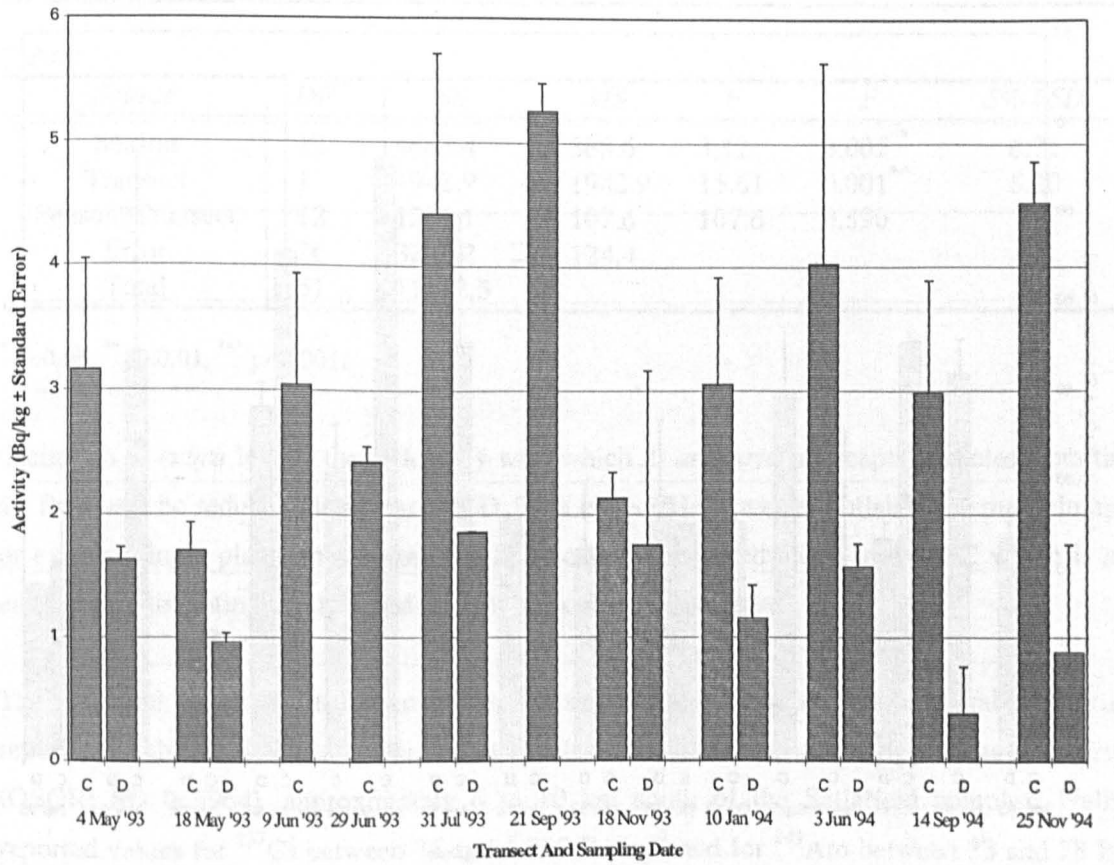


Figure 5.14: $^{239+240}$Pu activity in *A. arenaria* collected from the Sellafield sand dunes.

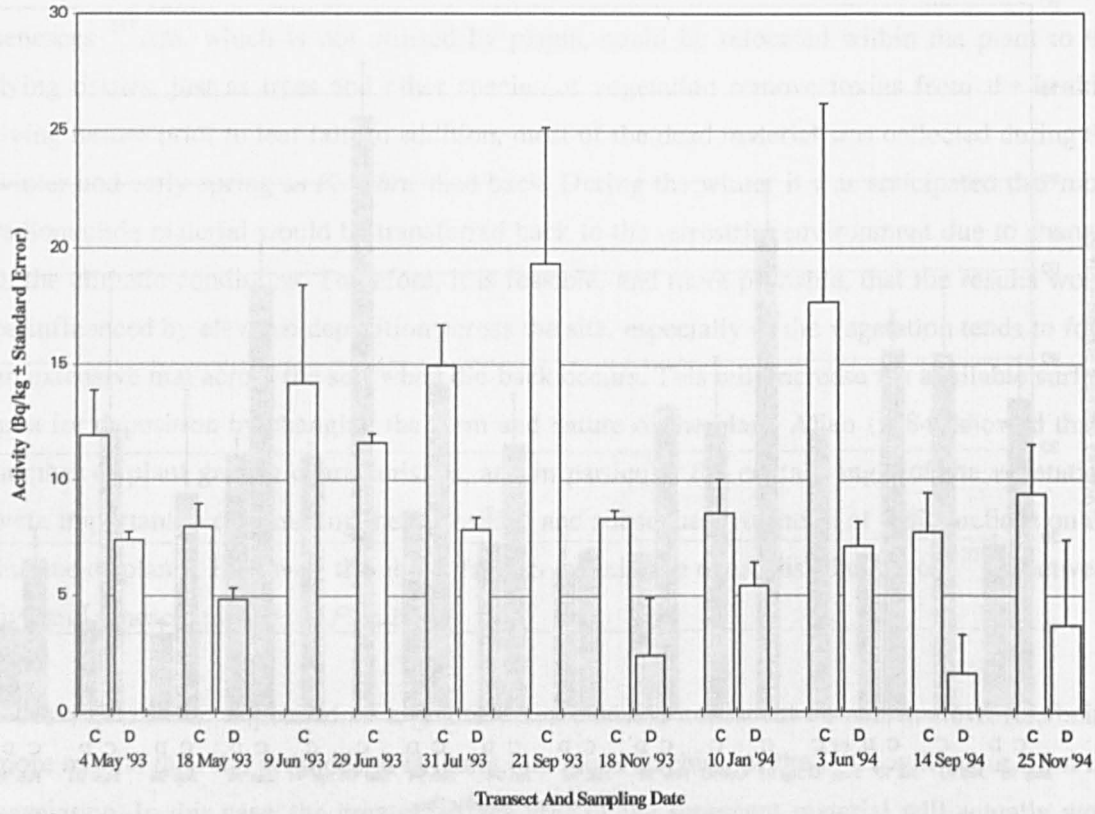


Figure 5.15: ^{241}Am activity in *A. arenaria* collected from the Sellafield sand dunes.

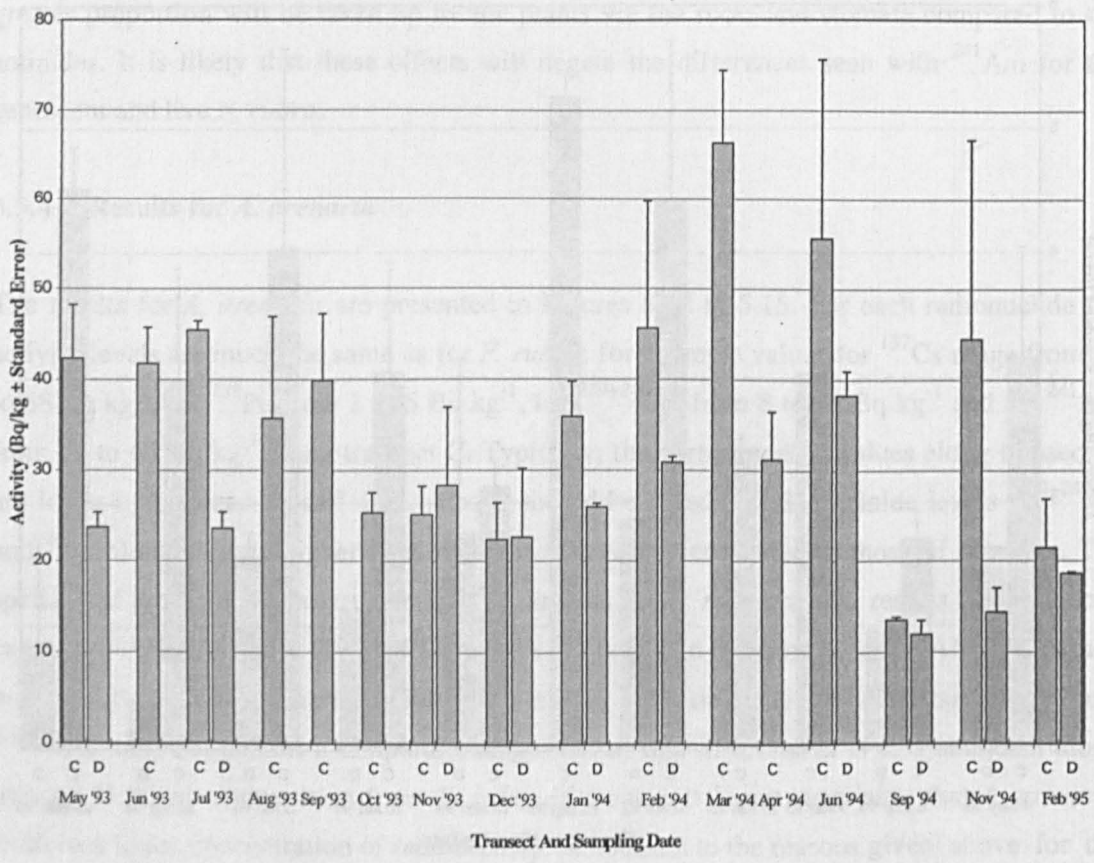


Table 5.10: **Results from a balanced two factor ANOVA. for spatial and temporal changes in *A. arenaria* ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities respectively.**

^{137}Cs:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	12	2758.46	229.87	4.31	0.001**	1.43
Transect	1	2579.6	2579.60	48.42	0.000***	4.82
Season*Transect	12	452.32	37.69	0.71	0.731	
Error	26	1385.16	53.28			
Total	51	7175.53				

^{238}Pu:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	7	12.811	1.830	1.70	0.178	
Transect	1	29.242	29.242	27.24	0.000***	0.64
Season*Transect	7	3.967	3.967	0.567	0.53	
Error	16	17.179	1.074			
Total	31	63.199				

$^{239+240}Pu$:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	7	171.88	24.55	2.21	0.090	
Transect	1	206.76	206.76	18.58	0.001**	2.06
Season*Transect	7	56.31	56.31	8.04	0.72	
Error	16	178.08	11.13			
Total	31	613.03				

^{241}Am:

Source	*DF*	*SS*	*MS*	*F*	*P*	*5% LSD*
Season	12	4663.4	388.6	3.12	0.007**	6.71
Transect	1	1942.9	1942.9	15.61	0.001**	5.20
Season*Transect	12	1291.1	107.6	107.6	0.590	
Error	26	3235.2	124.4			
Total	51	11132.5				

* $p<0.05$, ** $p<0.01$, *** $p<0.001$.

decline in *F. rubra* levels, the efficiency with which *A. arenaria* intercepts particles from the air flow will be reduced along transect D. This is because it was established in small clumps or even as single plants thus losing the thick canopy observed along transect C which is so effective at disrupting air flow and causing deposition of particles.

The ^{137}Cs and ^{241}Am results obtained from the analysis of *A. arenaria* are comparable to those reported by Nellis (1990) for the same species collected from the Drigg Nature Reserve (OSGR: SD 065964), approximately 6 to 10 km south of the Sellafield complex. Nellis reported values for ^{137}Cs between 75 and 1,395 Bq kg^{-1} and for ^{241}Am between 33 and 78 Bq

kg^{-1}. The high ^{137}Cs value of 1,395 Bq kg^{-1} was recorded for a sample collected six months after deposition of Chernobyl-derived ^{137}Cs. Moreover, Nellis (1990) showed a clear decline in activity levels for both ^{137}Cs and ^{241}Am in *A. arenaria* samples with distance inland. For both radionuclides, the reported levels range from 40 to 1,200 Bq kg^{-1} for ^{137}Cs and 40 to 80 Bq kg^{-1} for ^{241}Am, for samples collected at a comparable distance from the high water mark at the Drigg and Sellafield sand dune sites.

It is interesting that temporal variation is less marked for *A. arenaria*, particularly for the plutonium isotopes. It was expected that the temporal distribution would be similar to that of *F. rubra* since both species will receive a similar deposition, although radionuclides adhered to the foliage of *A. arenaria* could be more susceptible to washoff by rainfall perhaps even on to the more dense swards of *F. rubra*. This may be true, but comparing Figures 5.6 to 5.9 with 5.12 to 5.15, it appears that *A. arenaria* exhibits fractionally higher levels of actinides compared to *F. rubra*, and that there is no consistent temporal pattern between the two species. This suggests that *A. arenaria* is an important component within the ecosystem for the accumulation of radionuclides.

5.5.5 Isotopic and Nuclide Ratios

Table 5.11 presents the isotopic and nuclide ratios for *A. arenaria* and *F. rubra* samples collected from the sand dune site. These ratios are averages from samples collected during May 1993, June 1993 and September 1994. These can be compared to the ratios obtained from the soil cores. Results from both transects are reported.

As expected, the ^{137}Cs:^{134}Cs ratios are very variable because of the use of LOD values. By using the LOD values, all the ratios reported in Table 5.11 are upper estimates. There appears to be no consistent temporal or spatial difference in the ratios. The same is true for ^{238}Pu:$^{239+240}Pu$. However, examining the data for the nuclide ratios, ^{137}Cs:$^{239+240}Pu$ and ^{137}Cs:^{241}Am (Table 5.11) there appears to be a potentially significant species difference, with *F. rubra* exhibiting much higher ratios than *A. arenaria*. This suggests that there is a greater concentration of ^{137}Cs either on, or within, *F. rubra*. It is thought that this may be related to either increased ^{137}Cs root or foliar absorption with subsequent translocation within the plant or it may be related to particle size and thus interception. The latter could be important if the deposited ^{137}Cs is associated with a particular particle size such as, for example, clays. Clay particles may be more efficiently entrapped by *F. rubra* which is fine leaved but grows in thick swards. This is in contrast to *A. arenaria* which is known to be effective at accreting larger sand particles.

Table 5.11: Isotopic and nuclide ratios from samples of *A. arenaria* and *F. rubra* collected from the Sand dunes.

Date	*Transect*	^{137}Cs:^{134}Cs	^{238}Pu:$^{239+240}Pu$	^{137}Cs:$^{239+240}Pu$	^{137}Cs:^{241}Am
			A. arenaria		
May '93	C	49 ± 8*	0.26 ± 0.04	3.1 ± 0.2	0.84 ± 0.02
	D	19 ± 4*	0.22 ± 0.01	2.6 ± 0.2	0.83 ± 0.05
June '93	C	72 ± 21*	0.22 ± 0.00	3.5 ± 0.2	0.92 ± 0.11
	D	-----	-----	-----	-----
September '94	C	33 ± 17*	0.44 ± 0.21	3.8 ± 2.3	1.84 ± 0.96
	D	12 ± 3*	n/a	n/a	0.76 ± 0.06
			F. rubra		
May '93	C	40 ± 8*	0.21 ± 0.00	6.4 ± 1.5	1.7 ± 0.3
	D	51 ± 18*	0.31 ± 0.10	5.9 ± 1.5	1.8 ± 0.4
June '93	C	175 ± 21*	0.24 ± 0.05	51.1 ± 0.6	21.0 ± 0.7
	D	43 ± 6*	0.20 ± 0.02	23.5 ± 1.9	12.7 ± 0.5
September '94	C	79 ± 0.5*	0.27 ± 0.03	14.0 ± 0.2	3.8 ± 0.4
	D	27 ± 0.7*	0.28 ± 0.06	19.4 ± 10.1	7.8 ± 2.8

----- Sample not collected.
n/a Sample not analysed.
* ^{134}Cs values used to calculate the ^{137}Cs:^{134}Cs ratio were <LOD.

Comparing the ratios for vegetation reported in Table 5.11 with those for soil cores (Table 5.6) it is notable that the ^{238}Pu:$^{239+240}Pu$ ratio is very similar. Furthermore, the nuclide ratios determined for *A. arenaria* are also similar to the sand. This perhaps lends support to the concept of differential particle size interception for the two species of vegetation. On the whole, the ^{137}Cs:^{134}Cs ratios are much lower for vegetation than for soil. This is thought to reflect the analytical difficulties posed by ^{134}Cs and also because of the presence of Chernobyl-derived ^{137}Cs in the soil cores.

5.5.6 Concentration ratios for *A. arenaria* and *F. rubra*

Table 5.12 presents calculated concentration ratios for *A. arenaria* and *F. rubra* collected from this site. The calculations were carried out using the formula described elsewhere (sections 3.6 and 4.6.3). Over time, the concentration ratios reported in Table 5.12 for each species along the two transects remained reasonably consistent; for example *A. arenaria* values from transect C for ^{238}Pu range from 0.086 to 0.097 and for $^{239+240}Pu$ 0.050 to 0.055. However, the data indicate that there are differences in the concentration ratios between the two species for the actinides, for example along transect C, the ^{238}Pu values decline to between 0.019 and 0.065 for *F. rubra* compared to the *A. arenaria* values reported above.

Table 5.12: Concentration ratios for samples of *A. arenaria* and *F. rubra* collected from the Sellafield sand dunes.

Month/Transect	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
		A. arenaria		
Transect C				
May '93	0.113	0.097	0.055	0.212
January '94	0.091	0.094	0.050	0.171
September '94	0.050	0.086	0.050	0.061
Transect D				
May '93	0.047	0.055	0.046	0.118
January '94	0.061	0.054	0.046	0.136
September '94	0.038	0.025	0.022	0.056
		F. rubra		
Transect C				
May '93	0.102	0.039	0.028	0.099
January '94	0.122	0.065	0.056	0.145
September '94	0.080	0.019	0.016	0.039
Transect D				
May '93	0.060	0.057	0.030	0.074
January '94	0.097	0.060	0.057	0.104
September '94	0.137	0.018	0.015	0.025

This is not consistent throughout all the actinide data but does suggest that *A. arenaria* is more effective at accumulating particles with a higher actinide content.

The concentration ratios are also much higher than expected if root uptake alone was the dominant mechanism for the transfer of radionuclide contamination. This is not surprising given, firstly, the probable low bioavailability in the soil and, secondly, the interception by foliage of particles returning to land by sea to land transfer. It is anticipated that the radionuclide levels adhered to foliage are therefore a significant component in the food chain transfer for this site. The latter would also help to explain the consistency in the calculated ratios (Table 5.12).

5.5.7 Comparison of Vegetation Samples Collected in Cheshire and Wales with those from the Sellafield sand dunes

Table 5.13 compares the activity data from Ness Gardens, Cheshire with the vegetation samples collected from Wales and the Sellafield sand dunes. It is evident that there are highly significant differences in radionuclide contamination between the sand dunes and the two reference sites. For example, ^{137}Cs levels are between 11 and 100 times higher in samples from the Sellafield dunes than either reference site. The data for ^{238}Pu, $^{239+240}Pu$ and ^{241}Am are between 20 and 50, 20 and 165, and 45 and 500 times higher along the Sellafield dunes

Table 5.13: **A comparison of the ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities (Bq kg^{-1}) for vegetation from the Sellafield sand dunes with the Cheshire and Welsh reference sites.**

Transect/Site	*^{137}Cs*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*
		A. arenaria		
C	29.6 ± 3.7	4.4 ± 1.3	14.9 ± 1.8	35.4 ± 10.5
D	20.7 ± 3.7	1.9 ± 0.0	7.9 ± 0.5	23.6 ± 5.9
		F. rubra		
C	22.2 ± 3.6	3.3 ± 0.8	4.4 ± 0.4	7.2 ± 1.2
D	25.92 ± 5.5	3.1 ± 0.7	1.8 ± 0.2	4.5 ± 0.4
		F. rubra		
Cheshire	<0.97	0.09 ± 0.05	0.04 ± 0.00	0.10 ± 0.08
		A. arenaria		
Wales	0.68 ± 0.13	0.05 ± 0.01	0.09 ± 0.01	0.66 ± 0.38
		F. rubra		
Wales	2.83 ± 3.42	0.05 ± 0.00	0.05 ± 0.01	0.07 ± 0.02

Sand dune data taken from May 1993, Cheshire data from August 1993, and Welsh data from August 1994. Data from reference sites ± standard deviation (n=2), from sand dunes ± standard error (n=4).respectively.

The significance of these results was confirmed for ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am with a One-way ANOVA ($p<0.01$), a summary of which is presented in Table 5.14. These results agree with the hypothesis that along the Sellafield dunes the radionuclide inventory is strongly influenced by discharges from the fuel reprocessing plant.

Table 5.14: **Summary of One-way ANOVA examining the differences in radionuclide activity for vegetation samples collected from the Sellafield sand dunes and the Welsh and Cheshire reference sites.**

Radionuclides	*F-ratio*	*P*	*5% LSD*
		A. arenaria	
^{137}Cs	29.29	0.004**	10.12
^{238}Pu	10.30	0.024*	2.46
$^{239+240}Pu$	61.13	0.001***	3.48
^{241}Am	8.51	0.033*	22.87
		F. rubra	
^{137}Cs	16.35	0.010**	12.62
^{238}Pu	13.98	0.014*	1.49
$^{239+240}Pu$	116.72	0.000***	0.72
^{241}Am	26.60	0.004**	2.46

5.5.8 Summary of sand dune vegetation

The sand dunes support mature, stabilising communities of marram grass (*A. arenaria*) and red fescue (*F. rubra*).Samples were analysed for ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am. Measurement difficulties precluded quantification of ^{134}Cs. Radionuclide concentrations were surprisingly similar for the two species, ranging from 20 to 70 Bq kg^{-1} (^{137}Cs), 1 to 5 Bq kg^{-1} (^{238}Pu), 10 to 30 Bq kg^{-1} ($^{239+240}Pu$) and 10 to 65 Bq kg^{-1} (^{241}Am). As with soils, little temporal variation was observed for either species, reflecting both the proximity of the two transects to the sea, and each other, and the dominant influence of deposition by sea to land transfer. The little temporal variation which was observed is thought to result from variations in climatic conditions antecedent to sampling.

In contrast to the soils, both species of vegetation exhibit spatial differences for ^{137}Cs, $^{239+240}Pu$ and ^{241}Am, with higher levels of activity in the areas closest to the sea. The deposition rate from sea to land transfer declines exponentially within a few hundred metres from the shoreline. This factor combined with the influence of accretion around *A. arenaria*, is thought to be the cause of local variation in activity.

Concentration factors for the two species were similar at 0.05 to 0.14 for ^{137}Cs, 0.025 to 0.097 for ^{238}Pu, 0.022 to 0.057 for $^{239+240}Pu$ and 0.025 to 0.212 for ^{241}Am. These are higher than expected if root uptake was the exclusive transfer mechanism and they certainly reflect the general low level of transfer of radionuclides from substrate to plant and the high external contamination of radionuclides adhered to the plant foliage.

5.6 THE SAND DUNES TO THE SOUTH OF THE SELLAFIELD LIQUID DISCHARGE PIPELINE

Modifications to the sand dune system south of the Sellafield discharge pipeline have already been described briefly. These dunes were re-engineered by BNFL as part of the ongoing development of the Sellafield site. During the alterations, the original contaminated sand was diluted by the addition of 'clean' material. The area was then re-seeded with the effect that *F. rubra* became considerably more abundant along the leading edge of the windward slope compared to the principal study area. It was realised that small mammals particularly would transverse and forage in the modified sand dunes. This prompted a limited sampling programme to determine whether there are any significant differences in the overall radionuclide inventory of the dune area and the principal study site.

A number of soil cores and vegetation samples (principally *A. arenaria* and *F. rubra*) were collected during May 1994 (vegetation only) and September 1994. Tables 5.15 and 5.16

present the data for soil and vegetation respectively. Samples were collected from the windward slope and from the apex of the dunes, an area between 10 and 20 m wide. Comparing the results from Table 5.15 with Table 5.2, it is clear that the radionuclide inventory in the 'south' dune sand is approximately one third that of the principal site. However, comparing the data for *A. arenaria* in Table 5.16 with Figures 5.12 to 5.15, and for *F. rubra* in Table 5.16 with Figures 5.6 to 5.9, it is apparent that the levels are generally similar. This indicates that foliar contamination is prevalent throughout the sand dune system which in itself is unsurprising since the dunes will be exposed to the same meteorological conditions. The data indicate that root uptake is minimal and that herbivorous animals will be subjected to similar radionuclide levels in their diet wherever they forage on the dunes. The lower radionuclide levels in the soil will be important, however, when considering the dose received by individual animals. The reduction in contamination due to the mixing of 'clean' sand will have produced an area where external irradiation from the soil will be less than in the principal study area.

Table 5.15: **^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities (Bq kg^{-1} ± s.d., n=2) for soil cores extracted from the modified sand dunes to the south of the Sellafield pipeline.**

Position	*^{137}Cs*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*
SLOPE				
September '94	260 ± 20	7.5 ± 2.1	55 ± 4	70 ± 5
TOP				
September '94	325 ± 16	6.7 ± 0.9	69 ± 6	77 ± 4

Table 5.16: **^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am activities (Bq kg^{-1} ± s.d., n=2) in *A. arenaria* and *F. rubra* collected from the modified sand dunes to the south of the Sellafield pipeline.**

Position	*^{137}Cs*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*
		SLOPE		
May '94				
A. arenaria	37 ± 0.9	1.9 ± 0.7	3.8 ± 0.1	39 ± 0.1
F. rubra	21 ± 5	0.8 ± 0.4	4.6 ± 1.3	10 ± 0.7
September '94				
A. arenaria	10.8 ± 0.5	2.3 ± 2.3	5.9 ± 5.0	13.3 ± 0.9
F. rubra	9.0 ± 2.0	3.5 ± 2.0	4.5 ± 2.2	6.1 ± 1.5
		APEX		
September '94				
A. arenaria	31 ± 11	4.3 ± 0.9	7.3 ± 0.1	40 ± 8
F. rubra	23 ± 2	2.7 ± 1.3	6.6 ± 1.1	6.2 ± 1.4

5.7 INVERTEBRATE RESULTS

Table 5.17 lists the major invertebrate orders and principal species caught using pitfall traps set along transects C and D. Table 5.18 provides information on the seasonal abundance and biomass of each group/species. The data in Table 5.18 were produced by combining the catch from each pitfall set. The taxonomic groups analysed are indicated in the two tables. The low biomass obtained for the major taxonomic groups presented analytical difficulties which were overcome by pooling the samples from the pitfalls over a period of four months. In addition, it was originally intended that the 24 pitfalls exposed would produce four replicates for analysis. However, it became necessary to combine the contents of all 12 pitfalls from each transect thus reducing replication.

There was a much greater diversity of invertebrate species caught in the pitfall traps on the sand dune site compared to that of the woodland and salt marsh sites. However, in most cases the numbers and biomass collected for each taxonomic group were low, as indicated in Table 5.18. For Gastropoda, only a few individual slugs and snails were caught but these contributed significantly to the total biomass because of their relative size. Of the other taxonomic groups,

Table 5.17: The major invertebrate orders and species caught in pitfall traps at the Sellafield sand dunes.

CLASS/ SUBCLASS	*ORDER*	*FAMILY*	*GENUS/SPECIES*
Arachnida	**Araneida**		
	Opiliones		
Crustacea	**Isopoda**		*Oniscus asellus*
			Philoscia muscorum
Diplopoda			
Insecta	**Collembola**		
	Coleoptera	**Carabidae**	*Carabus violaceous*
			Feronia nigrita
		Cryptophagidae	
		Curculionidae	
		Elateridae	
		Scarababeidae	
		Staphylinidae	
	Diptera		
	Hymenoptera*		*Bombus spp.*
		Formicidae	
	Lepidoptera*		
Gastropoda			
Oligochaeta			*Lumbricus spp.*

Those groups highlighted in bold represent pooled units for analysis; where no species are listed or are not highlighted, then species were bulked into the higher Family or Order.
Adult and larvae forms of Coleoptera were analysed separately.
* Insufficient sample for analysis (<0.2 g dry weight).

Table 5.18: Seasonal abundance and biomass (g dry weight) of the invertebrates analysed from the Sellafield sand dunes.

Sample Group	*May to August '93*	*September '93 to February '94*	*March to July '94*	*August '94 to January '95*
Araneida	0.70	0.69	3.14	0.51
Opiliones*	2.25	1.73	0.20	0.97
Isopoda	7.32	3.42	1.42	0.83
Diplopoda	1.85	0.88	1.10	0.60
Collembola*	0.01	-----	1.70	0.98
Coleoptera (adults)	1.45	6.31	0.79	3.65
Coleoptera (larvae)*	-----	-----	0.28	0.26
Carabidae	-----	3.59	1.12	1.37
Diptera*	0.53	0.34	0.10	0.03
Formicidae*	3.75	1.71	0.02	0.48
Gastropoda	2.89	5.48	3.18	9.91
Oligochaeta*	0.61	0.65	1.41	0.30

----- Sample not obtained.
* Not all samples were analysed due to limitations of biomass.

Isopoda was caught in large numbers, and the remainder were caught consistently throughout the study but only a small biomass was acquired. During the winter period, samples were pooled over five months to obtain sufficient biomass for analysis.

5.7.1 ^{137}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am activity

Radionuclide activities were measured on a whole body burden basis although invertebrates were rinsed to remove any external particulates and the formalin solution in which they were stored. The results for ^{137}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am are presented in Tables 5.19 to 5.22 respectively. In the counting time available it was not possible to successfully quantify the activity of ^{134}Cs. Consequently, results for this nuclide have been omitted from the following discussion.

There is considerable variation in the radionuclide activities in the different taxonomic groups, as expected from the results discussed in the previous two chapters. For example, during the first sampling period, ^{137}Cs levels ranged from 3.6 Bq kg^{-1} for Araneida to 144 Bq kg^{-1} for Isopoda caught along transect C. This variation was also recorded for the actinides: 0.8 to 33.4 Bq kg^{-1} for ^{238}Pu, 3.7 to 148.7 Bq kg^{-1} for $^{239+240}$Pu and 4.1 to 165 Bq kg^{-1} for ^{241}Am for the same two taxonomic groups.

The detritivorous fauna is represented mainly by Isopoda (woodlice) and Oligochaeta (earthworms) but also by Collembola (springtails) and Diplopoda (millipedes). Only a very limited number of Oligochaeta and Collembola were caught over the period of study.

Table 5.19: **^{137}Cs activities (Bq kg^{-1} ± 2σ counting error) in invertebrates from the Sellafield sand dunes.**

Sample Group	*May to August '93*	*September '93 to February '94*	*March to July '94*	*August '94 to January '95*
		Transect C		
Araneida	5.6 ± 0.3	36.2 ± 5.2	69.0 ± 18.1	8.6 ± 1.4
Opiliones	54.9 ± 5.1	60.5 ± 7.2	4.6 ± 0.7	15.9 ± 2.9
Isopoda	154.0 ± 13.4	-----	31.3 ± 8.3	8.4 ± 0.6
Diplopoda	22.0 ± 2.1	-----	7.2 ± 1.9	6.3 ± 1.2
Collembola	n/a	-----	75.0 ± 19.3	-----
Coleoptera (adults)	43.4 ± 4.1	-----	5.6 ± 1.4	24.8 ± 4.6
Coleoptera (larvae)	-----	-----	6.2 ± 1.6	-----
Carabidae	-----	35.7 ± 4.2	8.2 ± 2.1	-----
Diptera	6.0 ± 0.6	4.8 ± 0.6	1.2 ± 0.3	n/a
Formicidae	42.2 ± 3.9	17.7 ± 2.1	-----	7.3 ± 1.4
Gastropoda	116.9 ± 10.0	148.1 ± 18.6	125.1 ± 32.8	209.3 ± 38.0
Oligochaeta	47.8 ± 4.5	6.0 ± 0.7	69.1 ± 17.8	9.5 ± 1.8
		Transect D		
Araneida	62.3 ± 4.2	15.7 ± 3.3	29.1 ± 3.2	8.0 ± 0.2
Opiliones	45.1 ± 5.6	21.6 ± 3.8	5.6 ± 0.6	6.5 ± 0.4
Isopoda	196.5 ± 12.7	35.8 ± 6.5	4.7 ± 0.5	7.2 ± 0.5
Diplopoda	26.7 ± 1.7	12.6 ± 2.1	12.9 ± 1.3	0.7 ± 0.1
Collembola	-----	-----	-----	21.2 ± 1.4
Coleoptera (adults)	55.3 ± 3.6	24.8 ± 4.1	3.7 ± 0.4	4.2 ± 0.3
Coleoptera (larvae)	-----	-----	-----	2.8 ± 0.2
Carabidae	-----	-----	-----	4.9 ± 0.3
Diptera	8.7 ± 0.6	1.4 ± 0.2	0.4 ± 0.04	n/a
Formicidae	103.3 ± 6.8	16.6 ± 2.8	-----	3.5 ± 0.2
Gastropoda	102.7 ± 7.4	121.1 ± 20.2	17.0 ± 1.9	41.2 ± 2.0
Oligochaeta	5.5 ± 0.4	45.1 ± 7.0	50.7 ± 5.0	5.4 ± 0.4

----- Sample not obtained.
n/a Sample was not analysed due to limitations with biomass.

Diplopoda were analysed but these detritivorous millipedes are thought to only form a small part of the diet of *S. araneus* (Churchfield, 1982; Rudge, 1968). From section 3.7.1 it was anticipated that any Oligochaeta would contain high levels of radionuclides. However, from Tables 5.19 to 5.22 it is evident that this is not the case. This is probably related to the paucity of clay minerals in the Oligochaete gut. Most of the radioactivity in samples of Oligochaeta has been shown to be associated with the gut content (Rudge, 1989) and this forms the bulk of the body weight for individuals. Moreover, earthworms preferentially ingest organic debris and small particle size fractions of soil, neither of which are common in sand dunes (Table 5.7). The lower radionuclide content for Oligochaeta is then probably due to the much higher content of sand particles in its diet. Further evidence for this is provided by comparisons of the actinide and ^{137}Cs levels for the same sampling period. Typically, the actinide levels are

Table 5.20: **^{238}Pu activities (Bq kg^{-1} ± 2σ counting error) in invertebrates from the Sellafield sand dunes.**

Sample Group	*May to August '93*	*September '93 to February '94*	*March to July '94*	*August '94 to January '95*
		Transect C		
Araneida	0.83 ± 0.07	5.86 ± 0.62	21.48 ± 1.85	1.81 ± 0.29
Opiliones	12.76 ± 1.12	8.16 ± 0.87	0.79± 0.07	3.72 ± 0.59
Isopoda	38.48 ± 2.94	-----	7.90 ± 0.85	1.82 ± 0.13
Diplopoda	5.12 ± 0.45	-----	2.21 ± 0.19	1.51 ± 0.24
Collembola	n/a	-----	23.01 ± 1.99	-----
Coleoptera (adults)	10.10 ± 0.89	-----	1.71 ± 0.15	5.94 ± 0.95
Coleoptera (larvae)	-----	-----	1.90 ± 0.16	-----
Carabidae	-----	4.74 ± 0.50	2.53 ± 0.22	-----
Diptera	1.39 ± 0.12	0.63 ± 0.07	0.36 ± 0.03	n/a
Formicidae	9.81 ± 0.86	2.34 ± 0.25	-----	1.74 ± 0.28
Gastropoda	24.85 ± 2.18	20.97 ± 2.23	38.99 ± 3.36	48.58 ± 7.77
Oligochaeta	11.11 ± 0.98	0.79 ± 0.08	21.21 ± 1.83	2.26 ± 0.36
		Transect D		
Araneida	2.27 ± 0.33	3.63 ± 0.45	4.01 ± 0.69	1.12 ± 0.15
Opiliones	3.00 ± 0.44	4.17 ± 0.52	0.70 ± 0.12	2.41 ± 0.32
Isopoda	7.83 ± 1.00	6.16 ± 0.89	2.58 ± 0.10	4.15 ± 0.40
Diplopoda	0.94 ± 0.14	2.32 ± 0.29	1.61 ± 0.28	0.27 ± 0.03
Collembola	-----	-----	-----	7.87 ± 1.03
Coleoptera (adults)	1.95 ± 0.29	4.57 ± 0.57	0.46 ± 0.08	1.57 ± 0.21
Coleoptera (larvae)	-----	-----	-----	1.04 ± 0.14
Carabidae	-----	-----	-----	1.83 ± 0.24
Diptera	0.31 ± 0.04	0.26 ± 0.03	0.05 ± 0.01	n/a
Formicidae	3.65 ± 0.53	3.06 ± 0.38	-----	1.28 ± 0.17
Gastropoda	3.98 ± 0.58	22.34 ± 2.79	2.37 ± 0.41	11.56 ± 1.51
Oligochaeta	0.19 ± 0.03	7.76 ± 0.97	6.20 ± 1.06	2.01 ± 0.26

----- Sample not obtained.
n/a Sample was not analysed due to limitations with biomass.

close to those of ^{137}Cs and in some cases even exceed them. This indicates that sand particles are ingested.

Collembola and Diplopoda both show high levels of radionuclides when compared to the predatory groups but are significantly lower than the levels reported for Isopoda. Isopoda consistently exhibited high concentrations of radionuclides from all three field sites and this may reflect the physiological role of the hepatopancreas of *Oniscus asellus* which is known to be important in concentrating metals (Nriagu, 1979). The temporal variation in Isopoda levels is indicated in Figure 5.16. The results for all four radionuclides show that the activity levels are somewhat higher during the summer compared to the second winter of the study. The data for transect D show a marked and unexpected decline in radionuclide levels between successive years. There is no obvious explanation for this change, which is consistent for all

Table 5.21: **$^{239+240}$Pu activities (Bq kg^{-1} ± 2σ counting error) in invertebrates from the Sellafield sand dunes.**

Sample Group	*May to August '93*	*September '93 to February '94*	*March to July '94*	*August '94 to January '95*
		Transect C		
Araneida	3.7 ± 0.2	25.6 ± 0.9	101.9 ± 4.0	8.6 ± 0.6
Opiliones	56.7 ± 2.4	35.6 ± 1.3	3.8 ± 0.2	17.7 ± 1.3
Isopoda	148.7 ± 6.2	-----	46.9 ± 1.9	3.9 ± 0.3
Diplopoda	22.7 ± 1.0	-----	10.5 ± 0.4	7.2 ± 0.5
Collembola	n/a	-----	109.1 ± 4.3	-----
Coleoptera (adults)	44.8 ± 1.9	-----	8.1 ± 0.3	28.3 ± 2.1
Coleoptera (larvae)	-----	-----	9.0 ± 0.4	-----
Carabidae	-----	20.7 ± 0.8	12.0 ± 0.5	-----
Diptera	6.2 ± 0.3	2.8 ± 0.1	1.7 ± 0.1	n/a
Formicidae	43.5 ± 1.8	10.2 ± 0.4	-----	8.3 ± 0.6
Gastropoda	110.4 ± 4.6	91.5 ± 3.4	184.9 ± 7.3	231.3 ± 17.0
Oligochaeta	49.3 ± 2.1	3.5 ± 0.1	100.6 ± 4.0	10.8 ± 0.8
		Transect D		
Araneida	7.4 ± 0.6	14.7 ± 0.9	17.4 ± 1.4	4.5 ± 0.3
Opiliones	9.8 ± 0.8	16.9 ± 1.1	3.0 ± 0.3	9.7 ± 0.6
Isopoda	22.3 ± 1.8	29.0 ± 1.8	2.5 ± 0.2	12.3 ± 0.8
Diplopoda	3.1 ± 0.3	9.4 ± 0.6	7.0 ± 0.6	1.1 ± 0.1
Collembola	-----	-----	-----	31.7 ± 2.1
Coleoptera (adults)	6.4 ± 0.5	18.5 ± 1.2	2.0 ± 0.2	6.4 ± 0.4
Coleoptera (larvae)	-----	-----	-----	4.2 ± 0.3
Carabidae	-----	-----	-----	7.4 ± 0.5
Diptera	1.0 ± 0.1	1.1 ± 0.1	0.2 ± 0.0	n/a
Formicidae	11.9 ± 1.0	12.4 ± 0.8	-----	5.2 ± 0.3
Gastropoda	13.0 ± 1.1	90.4 ± 5.6	10.3 ± 0.9	46.6 ± 3.0
Oligochaeta	0.6 ± 0.01	31.4 ± 2.0	27.0 ± 2.2	8.1 ± 0.5

----- Sample not obtained.
n/a Sample was not analysed due to limitations with biomass.

the radionuclides, although less marked for ^{238}Pu and $^{239+240}$Pu (Tables 5.20 and 5.21). The most plausible explanation for the decline in year two is that, for some unknown reason, the activity levels in detritus were lower and therefore reflected in the levels measured for Isopoda. More interestingly, the levels of the actinides are similar to, or even greater than, those of ^{137}Cs which is in contrast to the expected results.

The adult Coleoptera group predominantly comprised herbivorous weevils and beetles. In addition, a number of slugs and snails belonging to the group Gastropoda were caught. Both these groups exhibited high levels of radionuclides. The temporal variation exhibited by Gastropoda is presented in Figure 5.16. It is notable that the ^{241}Am levels are actually greater than those for ^{137}Cs over all the sampling periods. This was unexpected but may reflect enhanced levels of the actinides due to sea to land transfer. The fluctuation in radionuclide

Table 5.22: **^{241}Am activities (Bq kg^{-1} ± 2σ counting error) in invertebrates from the Sellafield sand dunes.**

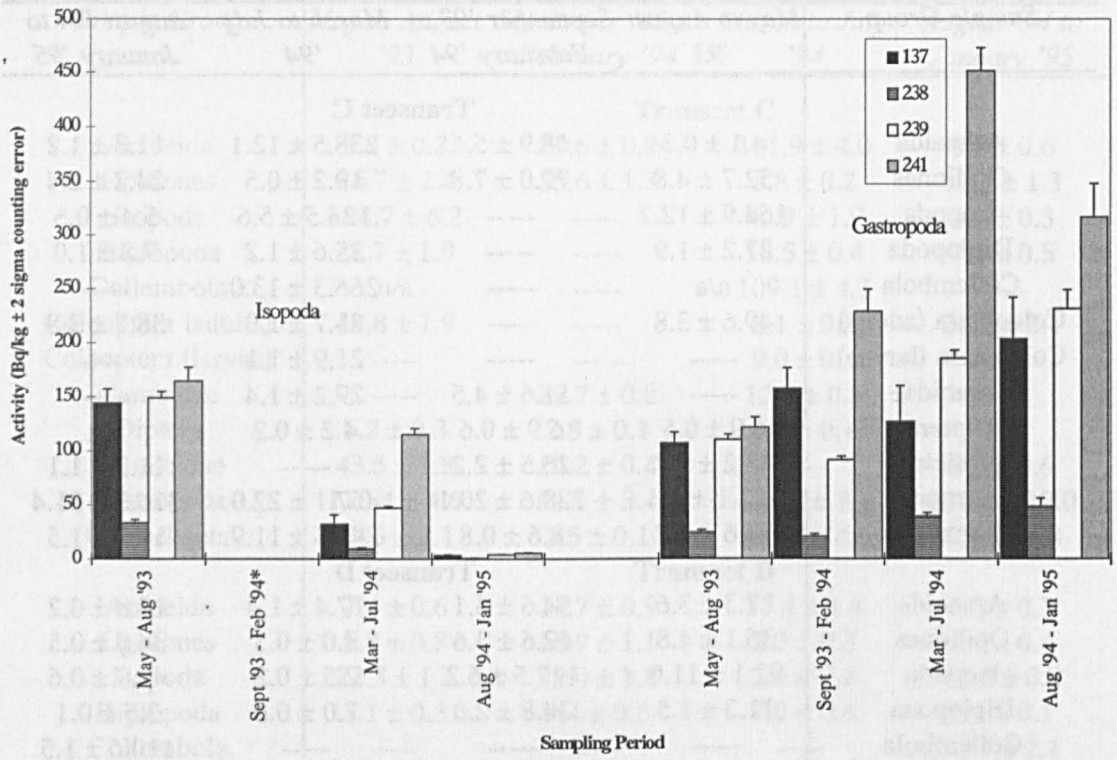
Sample Group	*May to August '93*	*September '93 to February '94*	*March to July '94*	*August '94 to January '95*
		Transect C		
Araneida	4.1 ± 0.3	68.9 ± 5.6	238.5 ± 12.1	11.8 ± 1.2
Opiliones	52.7 ± 4.8	79.0 ± 7.8	19.2 ± 0.5	24.2 ± 2.4
Isopoda	164.9 ± 12.7	-----	124.5 ± 5.6	5.4 ± 0.5
Diplopoda	27.2 ± 1.9	-----	25.6 ± 1.2	9.8 ± 1.0
Collembola	n/a	-----	266.3 ± 13.0	-----
Coleoptera (adults)	49.6 ± 3.8	-----	21.7 ± 1.0	38.7 ± 3.8
Coleoptera (larvae)	-----	-----	21.9 ± 1.1	-----
Carabidae	-----	51.6 ± 4.5	29.2 ± 1.4	-----
Diptera	6.9 ± 0.5	6.9 ± 0.6	4.2 ± 0.2	n/a
Formicidae	48.2 ± 3.7	25.5 ± 2.2	-----	11.3 ± 1.1
Gastropoda	132.1 ± 9.4	228.6 ± 20.1	457.1 ± 22.0	316.3 ± 31.4
Oligochaeta	54.6 ± 4.2	8.6 ± 0.8	245.4 ± 11.9	14.7 ± 1.5
		Transect D		
Araneida	27.3 ± 3.6	54.6 ± 3.1	17.4 ± 1.4	15.8 ± 0.2
Opiliones	36.1 ± 4.8	62.6 ± 3.6	3.0 ± 0.3	34.0 ± 0.5
Isopoda	92.1 ± 11.0	117.5 ± 6.2	2.5 ± 0.2	42.9 ± 0.6
Diplopoda	11.3 ± 1.5	34.8 ± 2.0	7.0 ± 0.6	3.5 ± 0.1
Collembola	-----	-----	-----	110.6 ± 1.5
Coleoptera (adults)	28.4 ± 3.1	68.6 ± 4.0	2.0 ± 0.2	22.1 ± 0.3
Coleoptera (larvae)	-----	-----	-----	14.7 ± 0.2
Carabidae	-----	-----	-----	25.8 ± 0.3
Diptera	3.7 ± 0.5	4.2 ± 0.2	0.2 ± 0.0	n/a
Formicidae	48.8 ± 5.6	46.0 ± 2.7	-----	18.1 ± 0.2
Gastropoda	47.8 ± 6.4	335.6 ± 19.3	10.3 ± 0.9	165.6 ± 2.2
Oligochaeta	4.3 ± 0.3	126.6 ± 6.7	27.0 ± 2.2	28.2 ± 0.4

----- Sample not obtained.
n/a Sample was not analysed due to limitations with biomass.

levels of Gastropoda (Figure 5.16) is likely to be due in part to changes in radionuclide levels on the vegetation but also due to changes in the species composition of the pooled sample.

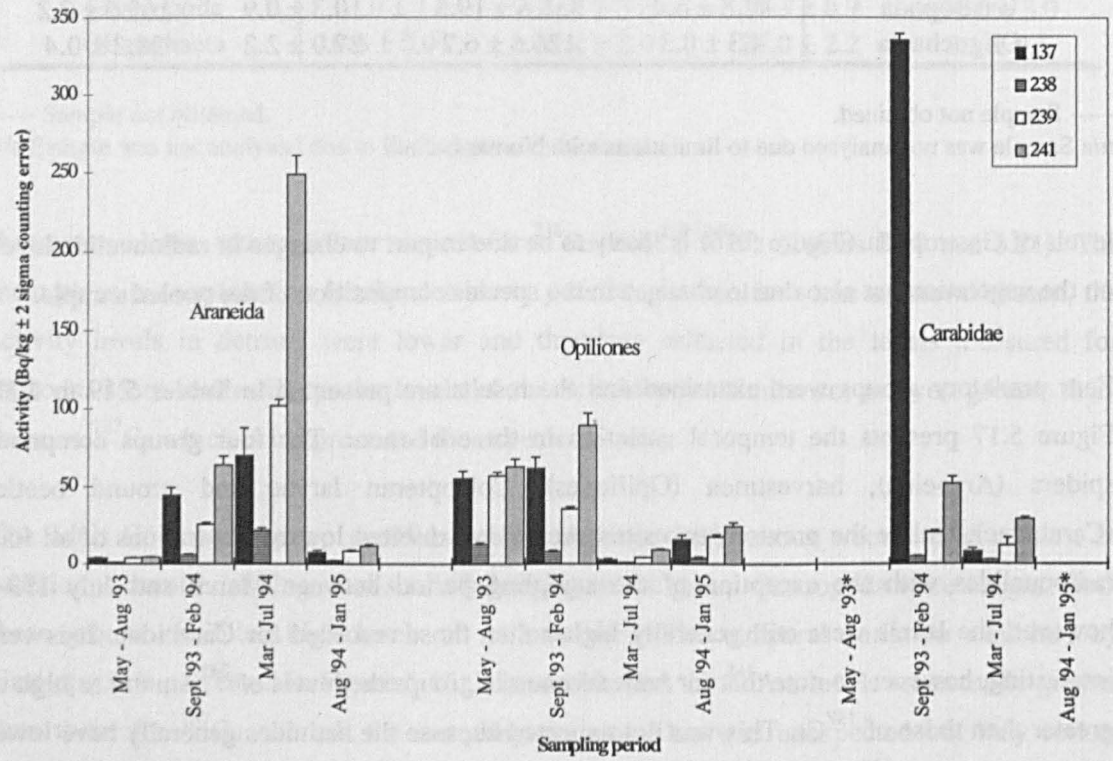
Four predatory groups were examined and the results are presented in Tables 5.19 to 5.22. Figure 5.17 presents the temporal variation in three of them. The four groups comprised spiders (Araneida), harvestmen (Opiliones), Coleopteran larvae and ground beetles (Carabidae). Unlike the previous two sites, Araneida exhibited low concentrations of all four radionuclides with the exception of the sampling period between March and July 1994; however, the levels were still generally higher than those recorded for Carabidae. It is very interesting, however, to note that for both taxonomic groups the levels of ^{241}Am are as high or greater than those of ^{137}Cs. This was not expected because the actinides generally have lower transfer rates than ^{137}Cs across the gastro-intestinal tract. However, this probably reflects the

Figure 5.16: Temporal variation in ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in Gastropoda and Isopoda.



* Sample not collected.

Figure 5.17: Temporal variation in ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in Araneida, Opiliones and adult Carabids.



* Sample not collected.

radionuclide content of food items in their diet which, for example, will include Isopoda and the herbivorous beetles; both of these groups contain high levels of actinides compared to ^{137}Cs.

Of the other taxonomic groups analysed, the ants (Formicidae) contained levels of ^{137}Cs ranging from 4 to 100 Bq kg^{-1} and between 5 to 12 Bq kg^{-1} of $^{239+240}Pu$, with levels of ^{241}Am again high at 18 to 46 Bq kg^{-1}. Large numbers of ants were caught during each trapping session but the total biomass was low. Ants are unlikely to form a major component in the diet of insectivorous small mammals but are interesting in their own right as scavengers and herbivores.

The results for Diptera are low, for example between 1 and 8 Bq kg^{-1} for ^{137}Cs and 0.2 to 6 Bq kg^{-1} for $^{239+240}Pu$. This was surprising given the radionuclide levels for Diptera caught at the River Esk (section 4.7.1) where ^{137}Cs levels ranged between 200 and 600 Bq kg^{-1}. Although there is much less strand material on the beach compared to that sampled at the River Esk site, it was expected that flies would utilise very efficiently what little there was and thus become contaminated. It was noted however that the amount and type of material present along the strand line varied considerably over time and so is not likely to be dependable for routine foraging.

5.7.2 Radionuclide concentration factors in invertebrates

Concentration factors were determined for each radionuclide for the detritivore, herbivore and predatory groups and the results are presented in Tables 5.23 to 5.25. For the detritivores the activities in senescent *F. rubra* had to be used in the calculations because samples of decomposing material were not collected during the study. Live *F. rubra* values were averaged across the two transects during the appropriate invertebrate trapping period before being used to calculate the concentration factors for herbivores. For the predator groups a range of factors was determined against prey items.

The values for detritivores (Table 5.23) were calculated only for ^{137}Cs and ^{241}Am as the samples of senescent *F. rubra* were not analysed for plutonium. For ^{241}Am, the values show little temporal variation for each species analysed and, interestingly, nearly all the values were close to unity or greater. This may reflect the choice of senescent *F. rubra* for the calculations although it is probable that decomposing vegetation is actually the principal foodstuff. The high ^{241}Am values are thought to be caused by the ingestion of sand particles and the presence of these in the gut prior to capture. In contrast, ^{137}Cs concentration factors were generally less than unity, suggesting that the accumulation of ^{137}Cs was limited. Only Oligochaeta exhibited high levels of ^{137}Cs and it is probable that this is due to the presence of sand particles and

Table 5.23: Detritivore dietary concentration factors determined against average radionuclide activities in senescent *F. rubra*.

Radionuclide and Species or Group	*May to August '93*	*September '93 to February '94*	*March to July '94*	*August to January '95*
^{137}Cs				
Isopoda	3.81	-----	0.85	0.22
Diplopoda	0.71	0.33	0.34	0.17
Oligochaeta	1.26	1.11	1.31	0.25
^{241}Am				
Isopoda	3.28	4.30	4.58	1.72
Diplopoda	1.01	1.39	1.02	0.39
Oligochaeta	2.18	0.34	1.08	1.13

----- Sample not obtained.

NB. Senescent *F. rubra* samples were not analysed for plutonium and therefore concentration factors could not be determined.

organic matter in their gut. This supports the contention that sand particle ingestion is important in the concentration of radionuclides within the body. Furthermore, it is theoretically possible that actinides deposited via sea to land transfer are more biologically available because they may be present in a different chemical form to that found in the previous two study sites. More detailed analysis would be required to examine this. However, it is believed that the high actinide levels (compared to ^{137}Cs) in the invertebrate groups are due to the elevated levels of ingested sand particles, adhered either to the foliage of plants or to sand particles ingested accidentally.

Table 5.24 presents concentration factors for the herbivorous invertebrate groups, Gastropoda and adult Coleoptera. Low concentration factors were anticipated from previous work on terrestrial grasslands. However, as the results show, many of the values are greater than unity. The concentration factors were determined against *F. rubra* activities averaged over the two transects as foraging between the two areas could easily occur given the proximity of the transects. Using just *F. rubra* may lead to inaccuracies because it is possible that *A. arenaria*, and particularly the annual species listed in Table 5.1, may also be utilised by herbivorous insects; but it is expected that the bulk of their diet will comprise *F. rubra*. It is probable that the concentration factors reflect the ingestion of particles adhered to the surface of the foliage because, as mentioned previously, a high degree of external contamination arises from the marine deposition mechanism. This will raise the activity levels in invertebrates even though the radionuclides may not actually accumulate within the soft tissues of the individuals.

Table 5.24: Dietary concentration factors for the herbivorous invertebrates caught at the Sellafield sand dunes.

Radionuclide and Species or Group	*May to August '93*	*September '93 to February '94*	*March to July '94*	*August to January '95*
^{137}Cs				
Gastropoda	3.50	3.97	3.22	1.16
Coleoptera	1.42	0.81	0.14	0.93
^{238}Pu				
Gastropoda	3.72	19.24	50.64	17.65
Coleoptera	1.82	4.19	2.22	2.40
$^{239+240}Pu$				
Gastropoda	4.82	16.93	4.21	20.48
Coleoptera	2.37	3.46	3.31	2.81
^{241}Am				
Gastropoda	6.22	29.73	1.34	10.97
Coleoptera	3.04	8.92	2.57	2.62

It is notable that the concentration factors are considerably higher for slugs and snails (Gastropoda) caught on the site. Banded snails, *Cepea* spp., were principally caught and these contributed significantly to the overall invertebrate biomass. They also form part of the diet of insectivorous mammals such as *S. araneus* (Pernetta, 1976). These high factors may be attributable to the feeding mechanism of snails which scrape off the surface of the vegetation. As a consequence of this, particulate material adhered to the vegetation may be ingested. It is also known that marine molluscs accumulate radionuclides, particularly the actinides, within their shell and this may be occurring (McDonald *et al.*, 1993).

Concentration factors were determined for three of the predatory groups and the results are presented in Table 5.25. It can be seen that the concentration factors vary considerably depending upon the abundance and radionuclide levels of the possible prey species at different times of the year, although there is little evidence of temporal variation in the concentration factors. This is not altogether surprising. Although the principal prey will change from season to season due to fluctuating abundance, the activity levels in most prey species were reasonably similar so predators are exposed to a consistent input of radionuclides.

Concentration factors close to unity probably indicate the consumption of smaller animals within their own taxonomic group, particularly in respect of the Carabidae. In addition, it is known that Opiliones, unlike Araneida, are not strictly carnivorous and may consume decaying vegetable material and other foodstuffs. The concentration factors reported are probably, therefore, overestimates. In general, the concentration factors reported in Table 5.25 are in reasonable agreement with those determined in other invertebrate studies which range from 0.3 to 0.92 for ^{137}Cs (Crossley, 1969; Reichle and Crossley, 1969; Rudge, 1989).

Table 5.25: Dietary concentration factors for the predators (against a range of prey species) from the Sellafield sand dunes.

Radionuclide and Species or Group	*May to August '93*	*September '93 to February '94*	*March to July '94*	*August to January '95*
^{137}Cs				
Araneida	0.03 - 0.33	0.51 - 1.14	2.17 - 6.83	0.37 - 2.34
Carabidae	-----	0.92 - 2.01	0.25 - 1.14	0.60 - 1.40
Opiliones	0.38 - 0.44	0.58 - 1.59	0.08 - 1.19	0.79 - 4.59
^{238}Pu				
Araneida	0.02 - 0.29	0.95 - 1.32	0.01 - 2.72	0.99 - 2.04
Carabidae	-----	0.77 - 2.03	0.32 - 1.14	0.44 - 1.43
Opiliones	0.33 - 0.38	0.59 - 0.68	0.31 - 1.55	0.58 - 0.62
$^{239+240}Pu$				
Araneida	0.03 - 0.33	0.51 - 0.88	0.37 - 2.17	0.70 - 1.44
Carabidae	-----	0.71 - 2.03	0.26 - 1.14	0.60 - 0.89
Opiliones	0.17 - 0.44	0.58 - 1.22	1.20 - 1.52	0.37 - 2.49
^{241}Am				
Araneida	0.02 - 0.30	0.46 - 0.59	0.14 - 1.92	0.28 - 0.37
Carabidae	-----	0.46 - 1.12	0.11 - 0.23	0.23 - 0.60
Opiliones	0.32 - 0.39	0.53 - 0.67	0.15 - 1.20	0.56 - 0.79

----- Sample not collected.

5.7.3 Isotopic and Nuclide Ratios in Invertebrates

Table 5.26 presents calculated isotopic (^{238}Pu:$^{239+240}$Pu) and nuclide (^{137}Cs:$^{239+240}$Pu and ^{137}Cs:^{241}Am) ratios for selected taxonomic groups across the four trapping campaigns. Lack of quantification of ^{134}Cs prevented calculation of the ^{134}Cs:^{137}Cs ratio. The first point to note is that the ^{238}Pu:$^{239+240}$Pu is consistent across the five taxonomic groups and trapping campaigns, varying only between 0.17 and 0.47.

This is very similar to the values obtained for soil and vegetation and provides collateral evidence that the uptake of material deposited via sea to land transfer is occurring throughout the food chain, most probably in the form of particle ingestion from material adhered to foliage. This is then available for transfer through the invertebrate food chains. Furthermore, given the elevated levels of actinides compared to ^{137}Cs in material returned to the terrestrial environment, low nuclide ratios were expected for the invertebrates. Table 5.26 indicates this to be the case, with many of the values being less than one. This is even lower than expected based on the nuclide ratios for vegetation (Table 5.11). Why the actinides should exhibit such elevated uptake and concentration is unknown, and is in contrast to the behaviour of ^{137}Cs.

Table 5.26: Isotopic and nuclide ratios for selected invertebrate groups.

Radionuclide and Species or Group	*May to August '93*	*September '93 to February '94*	*March to July '94*	*August to January '95*
Araneida				
$^{238}Pu:^{239+240}Pu$	0.22	0.23	0.21	0.21
$^{137}Cs:^{239+240}Pu$	1.51	1.41	0.68	1.00
$^{137}Cs:^{241}Am$	1.37	0.53	0.29	0.73
Opiliones				
$^{238}Pu:^{239+240}Pu$	0.23	0.23	0.21	0.21
$^{137}Cs:^{239+240}Pu$	0.97	1.70	1.21	0.90
$^{137}Cs:^{241}Am$	1.04	0.77	0.24	0.66
Isopoda				
$^{238}Pu:^{239+240}Pu$	0.26	0.21	0.17	0.47
$^{137}Cs:^{239+240}Pu$	1.04	1.23	0.67	2.15
$^{137}Cs:^{241}Am$	0.93	0.30	0.25	1.56
Carabidae				
$^{238}Pu:^{239+240}Pu$	-----	0.23	0.21	0.25
$^{137}Cs:^{239+240}Pu$	-----	1.72	0.68	0.66
$^{137}Cs:^{241}Am$	-----	0.69	0.28	0.19
Coleoptera (adults)				
$^{238}Pu:^{239+240}Pu$	0.23	0.25	0.21	0.21
$^{137}Cs:^{239+240}Pu$	0.97	1.34	0.69	0.88
$^{137}Cs:^{241}Am$	0.88	0.36	0.26	0.64

----- Sample not collected.

5.7.4 Comparison with Cheshire Invertebrates

Table 5.27 presents data for invertebrates from the reference site in Cheshire. Comparing these data with the sand dune system is not ideal given the different structure and functioning of the ecosystems. However, the invertebrate community of the two sites is not totally dissimilar in comparison so cautious comparisons can be drawn.

It is clear that there is substantial elevation in the activity levels of each radionuclide at the Sellafield sand dunes when compared to the composite invertebrate sample obtained from the reference site. These differences are thought to be significant, but this cannot be confirmed statistically because of limitations of the composite data set.

Table 5.27: Radionuclide activity (Bq kg^{-1} ± 2σ counting error) in composite invertebrates collected from Cheshire.

Sample Dates	*Composite Invertebrate samples*			
	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
October 1993	<1.32	<0.19	<0.08	<0.38
March 1994	<0.57	<0.34	0.74 ± 0.09	<0.84
July 1994	2.51 ± 1.7	0.27 ± 0.04	0.23 ± 0.04	<0.73

< Values were less than the limit of detection which is reported.

5.7.5 Summary of invertebrate results

Whole body burdens of invertebrates varied between species and pooled taxonomic groups. Levels in Araneida and Isopoda ranged from 4 to 145 Bq kg^{-1} (^{137}Cs), 0.8 to 34 Bq kg^{-1} (^{238}Pu), 4 to 150 Bq kg^{-1}, 4 to 150 Bq kg^{-1} ($^{239+240}Pu$) and 4 to 165 Bq kg^{-1} (^{241}Am). The low levels of ^{134}Cs precluded its quantification.

Of the detritivorous fauna, Isopoda contained the highest level of radionuclides. This may be a result of radionuclide accumulation within the hepatopancreas. Low numbers of Oligochaeta were caught, reflecting the sandy properties of the soil profile, and these typically contained low levels of radionuclides. This is thought to reflect the lower quantity of nuclide-rich clay minerals present in their diet.

Herbivorous weevils, beetles, slugs and snails all exhibited high levels of radionuclides, particularly the snails . Using *F. rubra* to calculate concentration factors, snails exhibit very high accumulation rates. This probably reflects their feeding mechanism, which may encourage the ingestion of particulates adhered to the external surfaces of vegetation.

Of the predatory groups, Araneida and Opiliones exhibited higher concentrations than Carabidae. Again, this is probably associated with the feeding strategy of the different groups. In many cases the ^{241}Am levels in invertebrates are equivalent to or greater than ^{137}Cs, possibly reflecting differences in the bioavailability of ^{241}Am attributable to its marine origin.

Radionuclide concentration factors varied across species and seasons, with many factors higher than unity. This was most noticeable for detritivores and herbivores and occurred for all four radionuclides. This is thought to reflect the relatively high levels of actinides and possibly differences in their chemical form.

5.8 SMALL MAMMAL RESULTS

Table 5.28 presents the general details of the animals caught at each trapping session. Tables 5.29 to 5.31 present the radionuclide data (^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am) for each small mammal. Four trapping campaigns were undertaken and the animals were used as described in section 3.8. Following pilot radiochemical separation, it was decided to pool small mammals to increase the mass of sample. The same methods were applied to the animals caught in the sand dunes. The sample pairings are indicated in Tables 5.29 to 5.31.

Animals were trapped along two lines parallel to the coastline, these extending beyond the 50 m transects marked out for the main vegetation study (Figure 5.1). It was expected that the animals would forage some distance because of the shape of the dunes and the physical boundary presented by the River Ehen. A large number of juveniles were caught during the March and July 1994 trapping sessions, reducing the trapping efficiency. Attempts were made to collect an equal number of both sexes; however this was not always possible (Table 5.28). As for the previous two sites, it was intended to assess differences in radionuclide burdens between the sexes. However, when tested, no significant differences were observed ($p>0.05$, Mann-Whitney U test). As before, the results for females and males were then pooled to generate arithmetic means and standard errors (Tables 5.29 to 5.31).

Table 5.28: Small mammals caught at the Sellafield sand dunes.

Trapping Session	*Number Caught**	*Sex Distribution*	*Wet Weights (g)*
Apodemus sylvaticus			
September 1993	29 (6)	5M, 1F	19.3, 21.4, 22.3, 23.1, 23.8, 32.0
March 1994	53 (6)	4M, 2F	18.3, 21.8, 24.0, 25.2, 26.8, 27.9
July 1994	13 (6)	3M, 3F	17.4, 24.2, 25.5, 26.0, 26.6, 28.3
May 1995			See section 5.8.3
Microtus agrestis			
September 1993	0	-----	-----
March 1994	3 (3)	1M, 2F	15.3, 19.6, 24.2
July 1994	40 (9)	6M, 3F	17.4, 19.2, 20.3, 22.2, 22.9, 23.6, 25.1, 26.1, 30.4
May 1995	0	-----	-----
Sorex araneus			
September 1993	4 (4)	**	5.9, 5.9, 6.5, 7.1
March 1994	0	-----	-----
July 1994	4 (4)	**	9.1, 9.7, 9.7, 10.8
May 1995	0	-----	-----

* Number of animals caught, (number) killed. Note large numbers of juveniles were released in some cases.

** The sex of *S. araneus* was not determined.

Table 5.29: Radionuclide activities in individual field mice, *Apodemus sylvaticus*, from the Sellafield sand dunes and Cheshire reference site.

Sample Code	^{137}Cs	*Combined with*	^{238}Pu	$^{239+240}Pu$	^{241}Am
1A	23.2 ± 4.2	2A	0.09 ± 0.01	0.31 ± 0.03	0.22 ± 0.04
2A	24.7 ± 3.3	-----	-----	-----	-----
35A	22.6 ± 4.4	36A	0.06 ± 0.01	0.12 ± 0.02	0.15 ± 0.03
36A	10.0 ± 3.9	-----	-----	-----	-----
40A	24.4 ± 4.6	41A	0.29 ± 0.10	1.30 ± 0.05	0.22 ± 0.04
41A	17.4 ± 3.2	------	-----	-----	-----
Mean (A)	20.4 ± 2.3	-----	0.15 ± 0.07	0.58 ± 0.37	0.20 ± 0.04
1E	7.5 ± 2.0	13E	0.11 ± 0.02	0.19 ± 0.03	0.29 ± 0.05
13E	12.5 ± 2.1	-----	-----	-----	-----
4E	16.2 ± 3.5	23E	0.03 ± 0.01	0.39 ± 0.05	0.34 ± 0.06
23E	18.3 ± 3.3	-----	-----	-----	-----
7E	17.6 ± 3.8	30E	0.15 ± 0.03	0.24 ± 0.04	0.31 ± 0.06
30E	8.1 ± 2.9	-----	-----	-----	-----
Mean (E)	13.4 ± 1.9	-----	0.10 ± 0.04	0.27 ± 0.06	0.31 ± 0.01
17F	14.1 ± 5.0	23F	0.12 ± 0.03	0.22 ± 0.02	0.18 ± 0.04
23F	17.4 ± 4.0	-----	-----	-----	-----
15F	16.5 ± 5.6	25F	0.25 ± 0.06	0.36 ± 0.05	0.30 ± 0.05
25F	<1.6	-----	-----	-----	-----
8F	25.2 ± 3.6	37F	0.11 ± 0.02	0.21 ± 0.03	0.33 ± 0.06
37F	81.6 ± 5.2	-----	-----	-----	-----
Mean (F)	26.1 ± 11.5	-----	0.16 ± 0.05	0.26 ± 0.05	0.27 ± 0.05
	Cheshire Reference Site				
11B	<0.86	17B	1.03 ± 0.07	0.15 ± 0.03	0.06 ± 0.02
14B	<0.12	35B	0.17 ± 0.03	0.07 ± 0.02	0.04 ± 0.00
17B	<2.90	-----	-----	-----	-----
18B	<0.46	19B	0.07 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
19B	<0.74	-----	-----	-----	-----
35B	<0.08	-----	-----	-----	-----
Mean (B)	<0.86	-----	0.42 ± 0.02	0.10 ± 0.00	0.05 ± 0.01

A = September 1993, E = March 1994, and F = July 1994. B = August 1993 trap in Cheshire. All values in Bq kg^{-1} (± 2σ counting error) except means which are ± standard error.

The data show that the actinide concentrations for animals caught on the sand dunes are very similar to those from the Cheshire reference site. Given the apparent biological availability of the actinides in the invertebrates (Tables 5.19 to 5.22), this is surprising. For example, mean ^{238}Pu, $^{239+240}Pu$ and ^{241}Am levels in *A. sylvaticus* are 0.15, 0.58 and 0.20 Bq kg^{-1} respectively, and compare to average levels of 0.42, 0.10 and 0.05 Bq kg^{-1} for the same radionuclides in mice caught in Cheshire. Using Mann-Whitney U tests, the data from the sand dune and reference site animals are not significantly different ($p>0.05$). Only the $^{239+240}Pu$ and ^{241}Am activities for *S. araneus* are significantly different to the reference site. This may reflect the

Table 5.30: Radionuclide activities in individual field voles, *Microtus agrestis*, from the Sellafield sand dunes and Cheshire reference site.

Sample Code	^{137}Cs	*Combined with*	^{238}Pu	$^{239+240}Pu$	^{241}Am
6E	13.2 ± 6.1	17E	0.15 ± 0.03	0.25 ± 0.03	0.37 ± 0.07
17E	15.6 ± 3.8	-----	-----	-----	-----
16E	12.1 ± 4.3	-----	0.23 ± 0.04	0.27 ± 0.04	0.25 ± 0.06
Mean (E)	13.6 ± 1.0	-----	0.19 ± 0.06[1]	0.26 ± 0.01[1]	0.31 ± 0.08[1]
7F	7.2 ± 3.1	28F, 34F	0.05 ± 0.01	0.18 ± 0.02	0.16 ± 0.02
28F	13.2 ± 2.1	-----	-----	-----	-----
34F	17.5 ± 4.7	-----	-----	-----	-----
12F	12.9 ± 2.3	30F	0.07 ± 0.02	0.24 ± 0.03	0.14 ± 0.03
30F	13.5 ± 4.0	-----	-----	-----	-----
13F	4.8 ± 2.1	39F	0.13 ± 0.03	0.15 ± 0.03	0.08 ± 0.02
39F	13.4 ± 5.4	-----	-----	-----	-----
6F	7.1 ± 2.5	9F	0.14 ± 0.03	0.55 ± 0.06	0.07 ± 0.02
9F	11.8 ± 3.2	-----	-----	-----	-----
Mean (F)	11.3 ± 1.35	-----	0.10 ± 0.02	0.28 ± 0.09	0.11 ± 0.02
	Cheshire Reference Site				
43B[2]	<1.7	-----	0.20 ± 0.04	0.34 ± 0.04	0.55 ± 0.05
24B[2]	<1.4	-----	0.17 ± 0.03	0.07 ± 0.02	<0.05
45B	<4.6	-----	-----	-----	-----
Mean (B)	<2.6	-----	0.19 ± 0.02[1]	0.21 ± 0.19[1]	<0.30

E = March 1994, and F = July 1994. B = August 1993 trap in Cheshire. All values in Bq kg^{-1} (± 2σ counting error) except means which are ± standard error and except [1] which is reported as standard deviation.

[2] Several mammal samples pooled to produce these values.

high levels of actinides measured in the principal prey species such as adults and larvae of Coleoptera, Isopoda, Araneida, Gastropoda and Collembola.

In contrast to the actinide data, ^{137}Cs levels are greater for all three small mammal species when compared to the Cheshire site. This is significant ($p<0.01$) as Table B8 (Appendix B) shows. In addition to elevated levels of ^{137}Cs, the mammals also exhibit different inter-specific concentrations. For example, for *A. sylvaticus* ^{137}Cs levels range between 13 to 26 Bq kg^{-1}, for *M. agrestis* between 11 and 13 Bq kg^{-1} and for *S. araneus* between 42 and 72 Bq kg^{-1}. The ^{137}Cs concentrations are significantly higher in *S. araneus* compared to the other two species ($p<0.01$, Table B8, Appendix B).

Overall, the levels of radionuclides in the small mammals are low, as expected from the results in the previous two chapters. It is probable that the small mammals are in equilibrium with the levels of ^{137}Cs in their environment. However, this is unlikely to be true for the actinides which will very slowly accumulate within the body although the rate will be determined by their low transfer coefficient across the gastro-intestinal tract.

Table 5.31: Radionuclide activities in individual common shrews, *Sorex araneus*, from the Sellafield sand dunes and Cheshire reference site.

Sample Code	^{137}Cs	*Combined with*	^{238}Pu	$^{239+240}Pu$	^{241}Am
20A	38.7 ± 4.1	34A	0.18 ± 0.04	0.41 ± 0.06	0.80 ± 0.13
34A	47.0 ± 10.1	-----	-----	-----	-----
23A	39.0 ± 19.5	33A	0.14 ± 0.03	0.35 ± 0.05	0.41 ± 0.06
33A	44.7 ± 3.7	-----	-----	-----	-----
Mean (A)	42.4 ± 2.1	-----	0.16 ± 0.03[1]	0.38 ± 0.04[1]	0.61 ± 0.28[1]
43F	195.5 ± 24.1	51F	0.07 ± 0.03	0.74 ± 0.09	0.64 ± 0.10
51F	34.7 ± 15.2	-----	-----	-----	-----
48F	16.2 ± 3.8	53F	0.30 ± 0.05	0.18 ± 0.04	0.37 ± 0.07
53F	39.6 ± 7.0	-----	-----	-----	-----
Mean (F)	71.5 ± 41.6	-----	0.19 ± 0.16[1]	0.46 ± 0.40[1]	0.51 ± 0.19[1]
	Cheshire Reference Site				
21B	<5.2	5B	0.14 ± 0.04	0.13 ± 0.04	0.34 ± 0.07
5B	<1.5	-----	-----	-----	-----
6B	<0.5	-----	0.07 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
Mean (B)	<0.2	-----	0.11 ± 0.05[1]	0.10 ± 0.04[1]	0.20 ± 0.20[1]

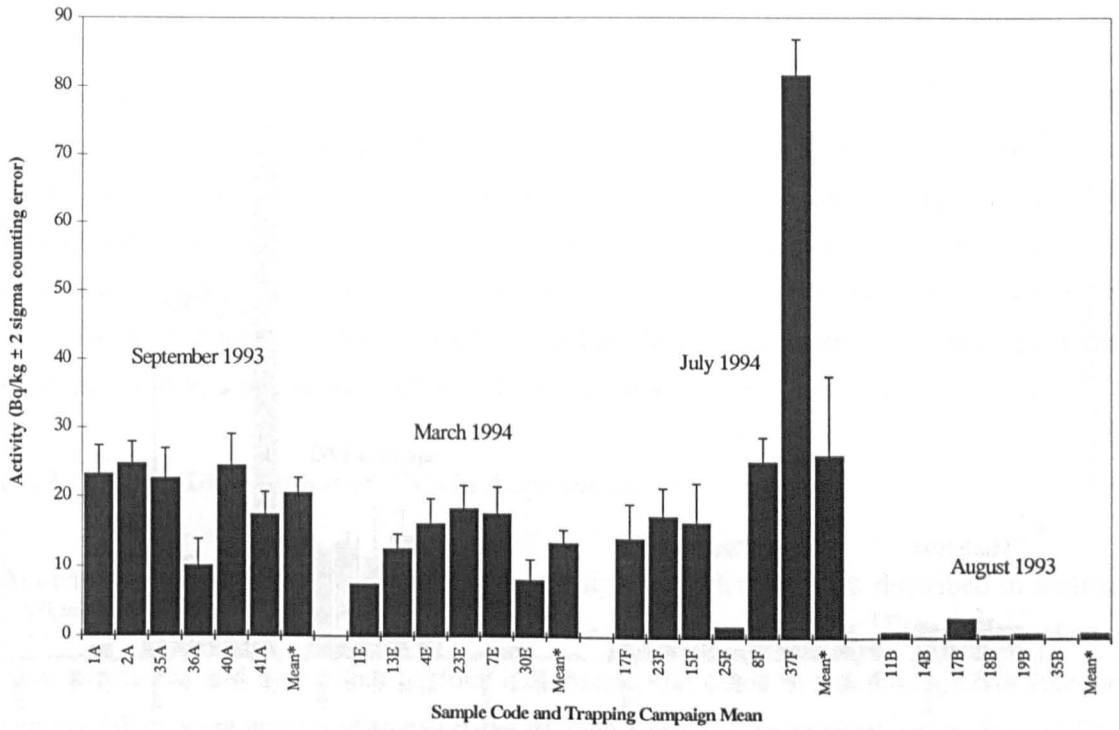
A = September 1993, and F = July 1994. B = August 1993 trap in Cheshire. All values in Bq kg^{-1} (± 2σ counting error) except means which are ± standard error and except [1] which is reported as standard deviation.

5.8.1 Temporal Variation in Radionuclide Body Burdens

Figures 5.18 to 5.21 present the data for the small mammals caught at the sand dunes and, for comparison, those from the Cheshire site. For *A. sylvaticus* there is limited variation between individuals for the first two trapping campaigns; only in the third campaign are there any significant differences for ^{137}Cs. Greater individual variation in radionuclide levels was expected based on the results from the previous two chapters. However, it is evident from the data presented that this is not the case. It is postulated that this is a function of the more uniform deposition and concentration of each radionuclide across the sand dunes compared to the more heterogeneous nature of the salt marsh and woodland. Also, from Figures 5.20 and 5.21, it is very clear that the concentration of each radionuclide is very different for each species, with *S. araneus* showing approximately twice the levels of ^{137}Cs in *A. sylvaticus*. Surprisingly, *M. agrestis* had the lowest overall concentration of ^{137}C. From previous studies, this species was anticipated to have a radionuclide content similar to *A. sylvaticus* (Rudge, 1989). It is probable that the higher ^{137}Cs concentration in *A. sylvaticus* is related to the increased consumption of invertebrates described in section 1.4.4.

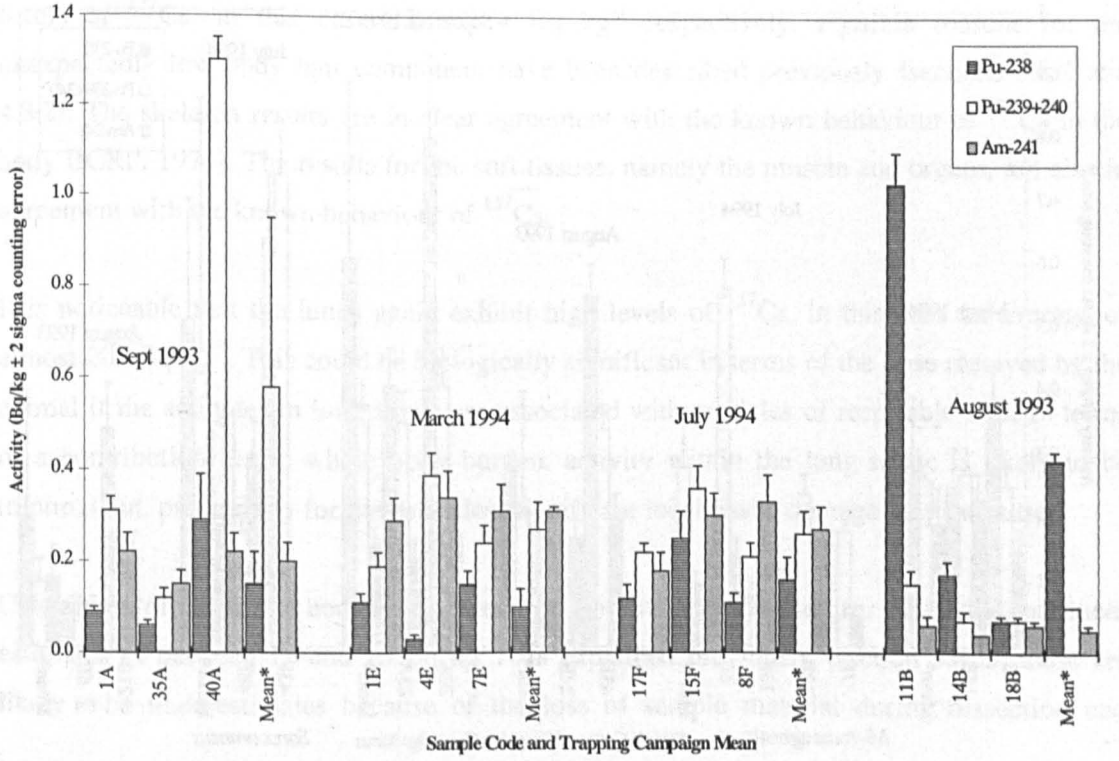
Unlike ^{137}Cs, the actinide data exhibit little by way of variation, temporal or otherwise. The data for animals caught in Cumbria tend to be generally higher than for Cheshire but the difference is insignificant ($p>0.05$). Given the much greater concentrations of the actinides in

Figure 5.18: ^{137}Cs in *Apodemus sylvaticus*, on the Sellafield sand dunes and in Cheshire.



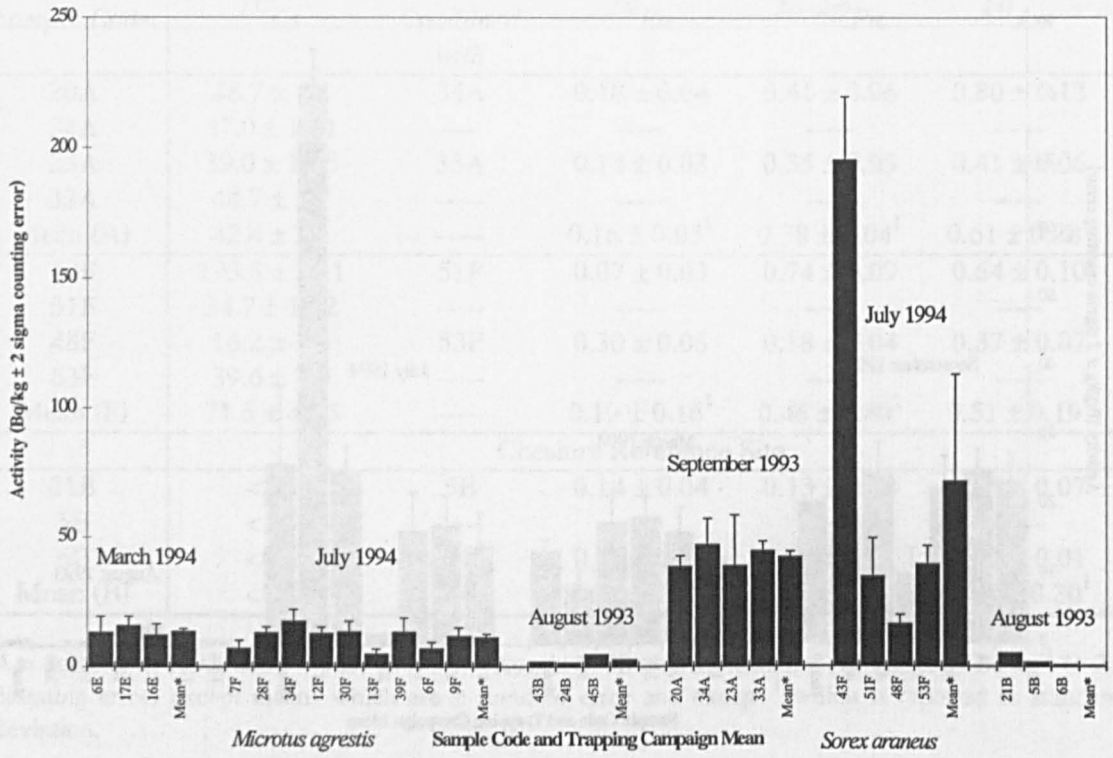
* Mean reported with standard error. Results from the Cheshire animals (August 1993) are < LOD.

Figure 5.19: ^{238}Pu, $^{239+240}Pu$ and ^{241}Am in *Apodemus sylvaticus*, on the Sellafield sand dunes and in Cheshire.



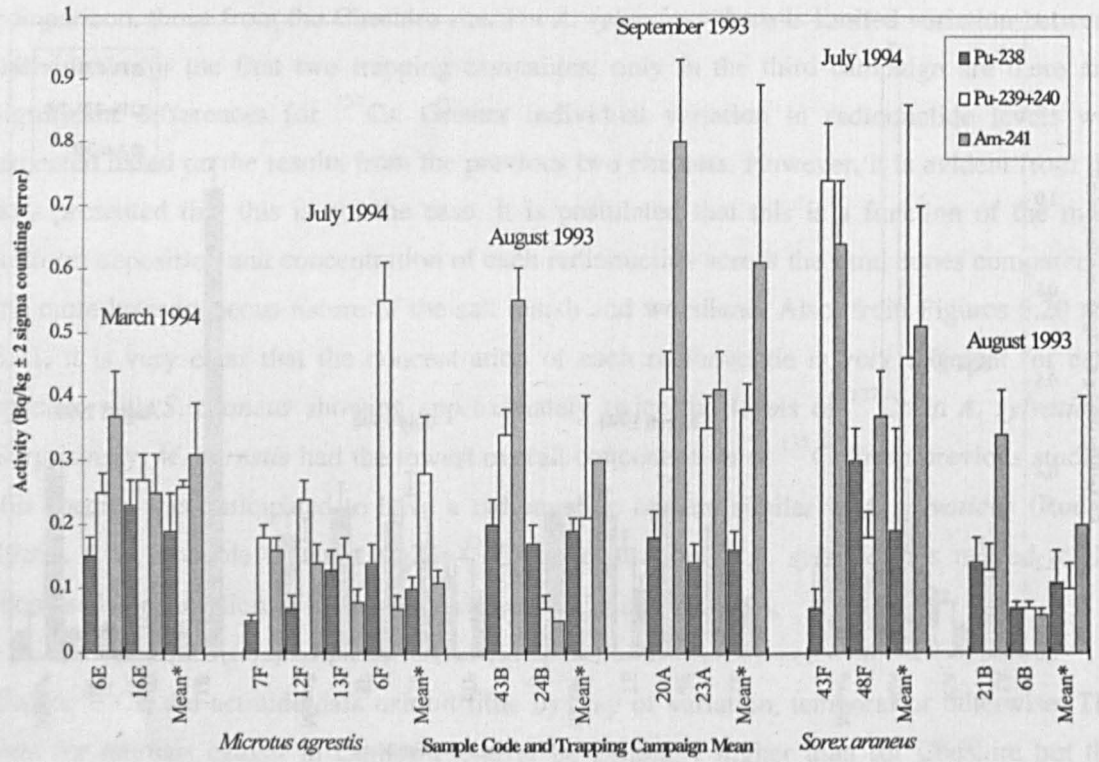
* Mean reported with standard error. Results from the Cheshire animals (August 1993) are < LOD.

Figure 5.20: **^{137}Cs in *Microtus agrestis*, and *Sorex araneus*, on the Sellafield sand dunes and in Cheshire.**



* Mean reported with standard error. Results from the Cheshire animals (August 1993) are < LOD.

Figure 5.21: **^{238}Pu, $^{239+240}$Pu and ^{241}Am in *Microtus agrestis*, and *Sorex araneus*, on the Sellafield sand dunes and in Cheshire.**



* Mean reported with standard error. Results from the Cheshire animals (August 1993) are < LOD.

the sand, vegetation and invertebrates in Cumbria, the low transfer coefficient of the actinides across the mammalian gut is clearly a major factor in determining the overall mammalian body burden of the actinides.

The overall lack of temporal variation indicates that the animals are not exposed to greatly different concentrations of the radionuclides in their diet throughout the year. This is not surprising since the vegetation and invertebrates also showed little temporal variation. Furthermore, given the isolated nature of the field site, it is unlikely that large-scale migration of the small mammals occurs; for the most part, animals caught will have been present on the dunes for most if not all of their lives. This, and the fact that uniform conditions exist across the dunes, will lead to a similar radionuclide exposure regime for the animals.

5.8.2 Tissue Distribution of ^{137}Cs in *A. sylvaticus*

Animals caught during the final trapping campaign were dissected (as described in sections 3.8.2 and 4.8.2) to provide information about the tissue distribution of ^{137}Cs. Two replicates were produced, one by combining four individuals, the other by combining five. Random number tables were employed to select the pooling combination and both sexes were utilised equally. Table 5.32 presents the data from the six body compartments analysed.

The results presented in Table 5.32, although more variable, are very much in line with the results in Tables 3.33 and 4.35 with both the body hair and skeleton containing very low levels of ^{137}Cs, in this case 32 and 4 Bq kg^{-1} respectively. Possible reasons for the unexpectedly low body hair component have been described previously (sections 3.8.2 and 4.8.2). The skeleton results are in clear agreement with the known behaviour of ^{137}Cs in the body (ICRP, 1976). The results for the soft tissues, namely the muscle and organs, are also in agreement with the known behaviour of ^{137}Cs.

It is noticeable that the lungs again exhibit high levels of ^{137}Cs, in this case an average of almost 205 Bq kg^{-1}. This could be biologically significant in terms of the dose received by the animal if the actinides in lung tissue are associated with particles of respirable size. In terms of a contribution to the whole body burden, activity within the lung tissue is likely to be minimal but, particularly for the actinides, significant local tissue damage may be caused.

Calculation of the whole body ^{137}Cs levels for the nine animals used for dissection, produces estimates of between 12 and 15 Bq kg^{-1}. As explained previously (section 3.8.2), these are likely to be underestimates because of the loss of sample material during dissection and

Table 5.32: **^{137}Cs in tissues and organs of *A. sylvaticus* from Sellafield sand dunes.**

Tissue	*Combined Wet Weight (g)*[a]	*^{137}Cs Bq kg^{-1} (replicate 1)*[b]	*^{137}Cs Bq kg^{-1} (replicate 2)*[b]	*^{137}Cs Bq kg^{-1} ± Standard Deviation*
Body Hair	2.52	45.1 ± 7.0	19.1 ± 4.1	32.1 ± 18.4
Gut (full)	50.40	20.9 ± 5.5	108.5 ± 6.7	64.7 ± 61.9
Lungs	3.54	58.4 ± 16.5	349.4 ± 62.8	203.9 ± 205.8
Muscle	11.51	54.0 ± 11.1	371.1 ± 27.1	212.6 ± 224.2
Organs	32.19	37.1 ± 6.1	112.7 ± 15.0	74.9 ± 53.5
Skeleton[c]	4.8	<2.5	4.4 ± 1.7	3.5 ± 1.3

[a] all nine animals. [b] Replicate values are reported ± 2σ counting error. [c] dry weight reported.

preparation. The estimates are however comparable to the whole body burdens reported for *A. sylvaticus* in Table 5.29.

5.8.3 Isotopic and Nuclide Ratios

Table 5.33 shows calculated isotopic (^{238}Pu:$^{239+240}Pu$)and nuclide (^{137}Cs:$^{239+240}Pu$ and ^{137}Cs:^{241}Am) ratios for small mammals caught on the Sellafield sand dunes. The mean ratio is presented for each trapping campaign. As previously (sections 3.8.3 and 4.8.3), analytical difficulties with ^{134}Cs prevented determination of the ^{134}Cs:^{137}Cs ratio.

The data for ^{238}Pu:$^{239+240}Pu$ are higher than expected given the isotopic ratios for soil, vegetation and invertebrates (Tables 5.6, 5.11 and 5.26). In contrast, the nuclide ratios for small mammals are considerably greater than for soil and vegetation. This was expected

Table 5.33: **Isotopic and nuclide ratios for small mammals from the Sellafield sand dunes.**

Trapping Session	^{238}Pu:$^{239+240}Pu$	^{137}Cs:$^{239+240}Pu$	^{137}Cs:^{241}Am
		A. sylvaticus	
September 1993	0.34 ± 0.08	94 ± 50	122 ± 14
March 1994	0.43 ± 0.30	51 ± 11	43 ± 9
July 1994	0.59 ± 0.05	77 ± 22	70 ± 8
		*M. agrestis**	
September 1993	0.73 ± 0.18	49 ± 6	42 ± 9
July 1994	0.42 ± 0.15	35 ± 9	75 ± 13
		*S. araneus**	
September 1993	0.42 ± 0.03	103 ± 12	72 ± 33
July 1994	0.88 ± 1.12	177 ± 123	175 ± 185

* Reported as mean ± standard deviation (n=2).

because of the low transfer of actinides across the gut. Therefore, the uptake of ^{137}Cs is considerably greater than that of the actinides even though in the sand dune ecosystem environmental levels of caesium and the actinides are reasonably similar as a consequence of sea to land transfer. This is in agreement with the known behaviour of caesium, plutonium and americium and clearly indicates a preferential uptake of caesium by mammals.

5.8.4 Radionuclide Concentration factors for *M. agrestis* and *S. araneus*

Table 5.34 presents concentration factors for four radionuclides in *M. agrestis* and *S. araneus*. Factors were not calculated for *A. sylvaticus* for the reasons described previously (section 4.8.4). The values for *M. agrestis* were determined against *F. rubra* because, as mentioned earlier, *A. arenaria* is not believed to form a significant part of the diet. The vegetation results were averaged over the two transects because the home range of these animals easily covers the width of the sand dunes.

It is evident from Table 5.34 that the concentration factors for the actinides are low. This was anticipated from earlier results and from the measured actinide body burdens for *M. agrestis* reported in Table 5.30. In contrast, the ^{137}Cs concentration values are higher and are consistent with the range of 0.10 to 0.49 reported by Rudge (1989) for the same species.

S. araneus exhibits a similar, and again low, set of concentration factors for the actinides. The range was determined by assessment of the likely prey species found on sand dunes. These concentration factors are much lower than those reported for the invertebrates at this site but strongly reflect the low transfer coefficients for the actinides compared to ^{137}Cs. The ^{137}Cs concentration factors exhibit a wider range and are the result of the greater bioavailability of ^{137}Cs in prey. Closer investigation of the more abundant prey species, suggests that the real concentration factor probably lies close to unity. For example, calculation of the concentration factor from Isopoda, Coleoptera and Oligochaeta produces such values.

It is probable that the very high concentration factor in Table 5.34 for *S. araneus* caught in July 1994 is due to the single individual which contained over 195 Bq kg^{-1} of ^{137}Cs. Recalculation of the concentration factors, without this outlier, produces a range of values between 0.57 and 7.71, much closer to the values for individuals caught in September 1993.

These concentration factors are comparable to those obtained by Rudge (1989) although there is a much greater range reported here. However, caution must be used during the interpretation of these factors. This is because *S. araneus* will equilibrate very rapidly with the environmental levels of radionuclides present but the pooling of invertebrates over time means that it is not possible to determine rapid changes in the invertebrates.

Table 5.34: Radionuclide concentration factors for *M. agrestis* and *S. araneus* from the Sellafield sand dunes.

Trapping Session	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
		M. agrestis		
September 1993	0.43	0.11	0.03	0.01
July 1994	0.26	0.11	0.05	0.01
		S. araneus		
September 1993	0.29 - 7.07	0.01 - 0.20	0.004 - 0.11	0.002 - 0.071
July 1994	0.57 - 19.32	0.005 - 0.41	0.002 - 0.23	0.001 - 0.26

5.8.5 Summary of small mammal results

Activity levels for ^{137}Cs in all three small mammal species were significantly higher along the sand dunes than at the reference site ($p<0.01$) and ranged from 13 to 26 Bq kg^{-1} (*Apodemus sylvaticus*), 11 to 13 Bq kg^{-1} (*M. agrestis*) and 42 to 72 Bq kg^{-1} (*S. araneus*). ^{137}Cs concentrations in shrews were significantly higher than those for the other two species ($p<0.01$). In contrast, the actinide data are very similar to data for the reference site being 0.15, 0.58 and 0.20 Bq kg^{-1} (*A. sylvaticus*), 0.19, 0.26 and 0.31 Bq kg^{-1} (*M. agrestis*) and 0.16, 0.38 and 0.61 Bq kg^{-1} (*S. araneus*) for ^{238}Pu, $^{239+240}Pu$ and ^{241}Am, respectively. Only the $^{239+240}Pu$ and ^{241}Am data for *S. araneus* are significantly higher than for the reference site ($p<0.05$). No significant differences were found between sexes ($p>0.05$) in respect of any radionuclide.

Temporal variation in ^{137}Cs levels was observed for *A. sylvaticus* and *S. araneus*. This is believed to reflect seasonal changes in the availability and abundance of prey items for *S. araneus* , and the consumption of seeds, vegetation and invertebrates and, particularly during the autumn, the prominence of fruits, berries and fungi in the diet of *A. sylvaticus*. Temporal variation was not observed for the actinides and their generally low uptake is a result of the very low transfer coefficient across the mammalian gut.

Measurements of the tissue distribution of ^{137}Cs showed elevated levels in muscle (210 Bq kg^{-1}) and lungs (200 Bq kg^{-1}), with lower levels recorded for pooled principal organs, gut contents and body hair. The lowest levels were observed in the skeleton with less than 5 Bq kg^{-1}. Calculations for the whole body burden gave conservative estimates up to 15 Bq kg^{-1}, the lower end of the scale for ^{137}Cs body burden measurements.

Chapter 6
SITE COMPARISONS AND CONCLUSIONS

The databases generated in this research project indicate, quite clearly, that the deposition mechanisms serving as the vector for the input to each of the three field sites is different. Moreover, the subsequent behaviour and transfer of radionuclides within the food chain at each site has its own distinct character even though species representing the higher trophic levels, particularly the small mammals, have similar features as regards their uptake and concentration of active material.

6.1 Soils

Soil or sediment samples from each site exhibit highly significant differences in terms of activity levels for all the target radionuclides; specifically, in order of increasing concentrations: Sand dunes < Lady Wood < River Esk, with the River Esk containing between 5 and 10 times the activity of Lady Wood soils and 10 times that of the sand dunes. Soils in Lady Wood show evidence of the accumulation of radionuclides due to atmospheric deposition over an extended period of time. Accumulated deposits lie within the range 250 to 320 Bq m^{-2} (^{134}Cs), 2,600 to 53,000 Bq m^{-2} (^{137}Cs), 160 to 470 Bq m^{-2} (^{238}Pu), 3,600 to 5,500 Bq m^{-2} ($^{239+240}Pu$) and 1,100 to 2,900 Bq m^{-2} (^{241}Am).

The deposits of plutonium in Lady Wood are characterised by low ^{238}Pu:$^{239+240}Pu$, ratios in the range 0.039 to 0.086. This is characteristic of plutonium produced from uranium of low irradiation and indicates that much of the activity has arisen from airborne deposition during the early years of operation of the Sellafield complex. Much of the ^{134}Cs in Lady Wood originates in active debris emitted to the atmosphere during from the Chernobyl nuclear power station on 26 April 1986, and fluxed into the woodland ecosystem by wet- and dry deposition which occurred as the radioactive plume passed over Cumbria between the 2nd and 3rd May 1986. Measurements imply that Chernobyl could account for up to 4,500 Bq m^{-2}, or about 10 to 20%, of the ^{137}Cs deposit.

The total inventory of all radionuclides in Lady Wood except ^{134}Cs shows a consistent spatial decline in accumulated deposit along a linear transect away from the dominant Sellafield source. In Lady Wood, activity declines from the 'front' to the 'back' of the wood because of the 'edge effect' whereby greater deposition occurs at the leading edge of the woodland facing Sellafield. Activity in surface soils from the sand dune ecosystem do not show such variation but reflect the predominant influence of sea to land transfer involving re-suspension of the sea surface microlayer. Accumulated deposits within the ranges: 160 to 280 Bq m^{-2} (^{134}Cs), 25,000 to 38,000 Bq m^{-2} (^{137}Cs), 1,400 to 3,600 Bq m^{-2} (^{238}Pu), 11,600 to 19,600 Bq m^{-2}

($^{239+240}$Pu) and 14,500 to 21,400 Bq m^{-2} (^{241}Am), are consistent with the relative deposition rates for actinides compared to ^{137}Cs in sea to land transfer.

The sand dune inventory of plutonium is characterised by ^{238}Pu:$^{239+240}$Pu ratios in the range 0.19 to 0.21. These are comparable to measurements reported previously for similar ecosystems and to the average ratio of these two plutonium isotopes present within marine sediments as a consequence of historic marine discharges.

There is evidence that the ^{137}Cs inventory has been influenced by deposition from the Chernobyl accident across all three field sites. Assuming that all the ^{134}Cs present derives from Chernobyl, it is estimated that between 8 and 15% of the ^{137}Cs is of this origin. For example, estimates of the Chernobyl contribution range from 2,300 and 3,500 Bq m^{-2} in the sand dune ecosystem, around 4,500 Bq m^{-2} in Lady Wood and between 13,000 and 30,000 Bq m^{-2} for the salt marsh at the River Esk. The more uniform pattern of ^{134}Cs compared to the other radionuclides measured at each site is consistent with much of the deposit having arisen from wet deposition when the Chernobyl 'cloud' reached Cumbria during May 1986.

Data from the cores taken in Lady Wood show greatest accumulation of activity at the soil surface horizons due to the efficient retention of nuclides by soil particles and the pine needle litter. In contrast, levels of radionuclides increase with depth in the sand dune as downward migration by leaching is emphasised by the coarse texture, low organic carbon and clay mineral content of the substrates with a low resulting cation exchange capacity. Sediment core profiles from the salt marsh site in the River Esk showed a contrasting pattern to Lady Wood and the sand dunes, with subsurface maxima for each radionuclide with a rapid decline in activity below 20 to 30 cm. This reflects accumulation of Sellafield-derived activity in the surface horizons over the last 40 years of salt marsh deposition history reflecting the stratification of the salt marsh. Much of the radioactivity within the marsh is thought to be sediment- and organic matter bound, which will limit its labile properties and bioaccumulation potential.

Sediment core samples from the salt marsh on the River Esk showed temporal and spatial variation in radionuclide activity, the latter being related mainly to the distance from the river. The spatial variation is exemplified by the range of 3,000 to 8,000 Bq kg^{-1} (^{137}Cs) at the riverward side of the marshland ecosystem. Lower activity levels were characteristic of the landward section of the marsh, consistent with the less frequent inundations of this area.

That the main influence upon the deposition of activity in the salt marsh is tidal patterns is apparent from the similar distribution of ^{137}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am. Measured activities for surface sediments from the central marsh ranged from 10,800 to 12,300 Bq kg^{-1} for ^{137}Cs,

to 12,500 to 15,000 Bq kg^{-1} for ^{241}Am, 1,100 to 1,450 Bq kg^{-1} for ^{238}Pu and 5,700 to 7,000 Bq kg^{-1} for $^{239+240}Pu$, with the ranges reflecting proximity to the river.

6.2 Vegetation

The vegetation cover varied dramatically between sites both in biomass production and species diversity. Only a sparse data set was obtained for activity in and on the vegetation of Lady Wood, with concentrations ranging from: 1 to 5 Bq kg^{-1} (^{134}Cs), 65 to 280 Bq kg^{-1} (^{137}Cs), 0.3 to 1.5 Bq kg^{-1} (^{238}Pu), 0.8 to 8 Bq kg^{-1} ($^{239+240}Pu$) and 0.6 to 16 Bq kg^{-1} (^{241}Am). The sand dunes, supporting mature stands of marram grass (*Ammophila arenaria*) and red fescue (*Festuca rubra*), showed radionuclide concentrations that were surprisingly similar for the two species. At 20 to 70 Bq kg^{-1} (^{137}Cs), values for ^{137}Cs were lower than for Lady Wood but actinide levels were similar at: 1 to 5 Bq kg^{-1} (^{238}Pu), 10 to 30 Bq kg^{-1} ($^{239+240}Pu$) and 10 to 65 Bq kg^{-1} (^{241}Am).

As with soils, little temporal variation was observed for either sand dune species, reflecting both the proximity of the two transects to each other, and the dominant influence of deposition by sea to land transfer. The little temporal variation which was observed is thought to result from variations in climatic conditions antecedent to sampling. In contrast to the soils, both species of vegetation exhibit spatial differences for ^{137}Cs, $^{239+240}Pu$ and ^{241}Am, with higher levels of activity in the areas closest to the sea. Fluxes in sea to land transfer of activity decline exponentially within a few hundred metres of the shoreline. This factor combined with the influence of accretion around *A. arenaria*, is thought to be the cause of local variation in activity, some of which is spatially predictable and some less so.

Activity levels in vegetation from the salt marsh showed significant temporal and spatial variation within, and between, the three sampling transects on the lower, middle and upper marsh. This mainly reflects the intensity and periodicity of tidal inundation. Thus, live plant material, mainly *Bromus mollis,* ranged from 10 to 80 Bq kg^{-1} for ^{137}Cs, 2 to 21 Bq kg^{-1} for $^{239+240}Pu$ and 3 to 54 Bq kg^{-1} for ^{241}Am. Senescent material showed higher and even more variable levels of activity. *Festuca rubra* showed considerable temporal variation but consistently low values at <100 Bq kg^{-1} for ^{137}Cs, $^{239+240}Pu$ and ^{241}Am. Values were higher in winter than in summer, probably due to tidal patterns and the impact of growth dilution. Predictable peaks in activity occurred along the frontal and strand line transects where, for example, levels of ^{137}Cs were, at 96 and 78 Bq kg^{-1} respectively, an order of magnitude higher than for the landward (rear) transect. Once again, ^{137}Cs levels in senescent samples collected in late autumn and winter were, at 40 to 120 Bq kg^{-1} and 40 to 75 Bq kg^{-1} for the seaward and riverward sectors of the marsh, much higher than the respective values for live tissue, 18 to 41 and 8 to 18 Bq kg^{-1}.

Live samples of *Juncus maritimus* and *Halimione portulacoides* from the salt marsh showed higher activities than *B. mollis* and *F. rubra*, a reflection of their habitat and the light coating of grey sediment associated with regular inundation. Further inter-specific differences between *J. maritimus* and *H. portulacoides* occurred in the same community, with the latter showing higher activity levels due to its physical structure being conducive to entrapment of sediment on leaf and branch surfaces.

The concentration factors for all radionuclides for Lady Wood vegetation were higher than could be explained by root uptake alone and therefore indicate either the importance of contemporary atmospheric deposition or high mobility/availability of radionuclides within forest soils. Isotopic and nuclide ratios for vegetation from the River Esk salt marsh also gave strong evidence in support of root absorption being a minor factor in vegetation activity levels, with sediment deposition being of over-riding importance. Activity transfer from sediment to vegetation by root uptake was generally low due to strong adsorption coefficients between radionuclides and both clay minerals and organic matter. Moreover, the concentration factors for the four nuclides of greatest interest suggest a lower bioavailability of ^{137}Cs borne in salt marsh sediment than in the case of agricultural soils. Nuclide concentration factors for the two main species comprising the sand dune community were similar: 0.05 to 0.14 for ^{137}Cs, 0.025 to 0.097 for ^{238}Pu , 0.022 to 0.057 for $^{239+240}Pu$ and 0.025 to 0.212 for ^{241}Am. Once again, these values are higher than would be expected if root uptake was the exclusive transfer mechanism. They certainly reflect the general low level of transfer of radionuclides from substrate to plant and the relatively high external contamination of radionuclides adhered to the plant foliage.

6.3 Litter

Pine needle litter in Lady Wood shows elevated levels of all radionuclides measured, ranging from 130 to 1,100 Bq kg^{-1} (^{137}Cs), 2 to 14 Bq kg^{-1} (^{238}Pu), 15 to 110 Bq kg^{-1} ($^{239+240}Pu$), and 18 to 105 Bq kg^{-1} (^{241}Am). Given the hypothesis that much of the deposition to soils is historic, concentrations are surprisingly high in relation to underlying soils, the ratio between leaf litter and soil concentrations typically lying between 0.3 and 1.0. The elevated concentrations in needle litter cannot be explained by adventitious soil contamination, and there are significant differences between isotopic ratios of ^{134}Cs:^{137}Cs and ^{238}Pu:$^{239+240}Pu$ which imply that contamination on the needle litter is of more recent origin than soils. It has been suggested that mechanisms involving fungal mycelia can lead to reconcentration of material into the litter layer and this may account for the relative levels of contamination on needle litter and soil.

Concentrations in needle litter from Lady Wood show the same spatial variation as in soils, with a fall in concentrations from the 'front' to 'back' edge of the wood, confirming that interception of aerosols by the woodland canopy is a dominant mechanism in deposition processes at the site. There is no valid comparison to be made between litter from Lady Wood and either the salt marsh or sand dune ecosystem. In the latter case, comparisons are negated by the absence of any meaningful litter layer. Comparisons between the salt marsh and the woodland are hindered by the dramatically different species composition of the litter layer.

In the salt marsh, the extent and biomass of the strand line varied seasonally and comprised mainly the leaf litter of *H. portulacoides*. Activity levels were similar for the seaward and riverward sectors of the marsh, with the ranges being: 180 to 600 Bq kg^{-1} for ^{137}Cs, 20 to 60 Bq kg^{-1} for ^{238}Pu, 80 to 250 Bq kg^{-1} for $^{239+240}Pu$ and 150 to 450 Bq kg^{-1} for ^{241}Am. Temporal differences between the riverward and seaward sectors of the marsh were more obvious than were spatial differences. The riverward sector showed higher activity levels between autumn and spring and peaked in mid-winter. For the seaward side the activity peak was in summer, a fact that is attributed to high rates of deposition in the summer and subsequent attrition and weathering later in the year.

6.4 Invertebrates

Whole body burdens of ground-living invertebrates from the sand dunes and Lady Wood varied between species and pooled taxonomic groups. In the sand dunes, levels in Araneida and Isopoda ranged from 4 to 145 Bq kg^{-1} (^{137}Cs), 1 to 34 Bq kg^{-1} (^{238}Pu), 4 to 150 Bq kg^{-1}, 4 to 150 Bq kg^{-1} ($^{239+240}Pu$) and 4 to 165 Bq kg^{-1} (^{241}Am). Low levels of ^{134}Cs within the samples precluded its quantification. At Lady Wood, activities varied over a broader range, from: 6 to 1,800 Bq kg^{-1} (^{137}Cs), 0.3 to 148 Bq kg^{-1} (^{238}Pu), 0.3 to 59 Bq kg^{-1} ($^{239+240}Pu$) and <0.3 to 400 Bq kg^{-1} (^{241}Am). The low levels again precluded quantification of ^{134}Cs. Levels of activity in invertebrates from the salt marsh also varied considerably between taxonomic groups, and were generally higher on the riverward sector of the salt marsh than on the seaward side. Extremes of the activity range for ^{137}Cs were 9 Bq kg^{-1} for adult Carabid beetles and 197 Bq kg^{-1} for the detritivorous Isopoda. Similar broad inter-taxonomic variation was recorded for the actinides: 3 to 134 Bq kg^{-1} for ^{238}Pu (Carabidae to Formicidae), 9 to 78 Bq kg^{-1} for $^{239+240}Pu$ (Carabidae to Isopoda) and 4 to 94 Bq kg^{-1} for ^{241}Am (Carabidae to Isopoda).

The detritivorous fauna, arguably the most important in Lady Wood, a coniferous woodland, showed higher values than for other taxa, namely: 43 to 1,800 Bq kg^{-1} (^{137}Cs), 1 to 15 Bq kg^{-1} (^{238}Pu), 7 to 74 Bq kg^{-1} ($^{239+240}Pu$) and <0.3 to 400 Bq kg^{-1} (^{241}Am). In the case of ^{137}Cs and ^{241}Am, the highest values were for Oligochaete earthworms wherein the gut contains residual

quantities of contaminated soil ingested involuntarily along with fragments of food. Isopods also showed strongly elevated levels of all measurable nuclides, reflecting the level of contamination of their litter food source.

Detritivores also exhibited the highest levels of activity, within the strand line community of the salt marsh system, ranging from 30 to 200 Bq kg^{-1} (^{137}Cs), 6 to 17 Bq kg^{-1} (^{238}Pu), 30 to 78 Bq kg^{-1} ($^{239+240}Pu$) and 40 to 100 Bq kg^{-1} (^{241}Am). Of the predatory species, the Araneida (spiders) showed the highest activities, especially for ^{137}Cs (10 to 110 Bq kg^{-1}). This is attributed to their exoenzyme feeding strategy in which the soft tissue of their prey, in which ^{137}Cs predominates, is pre-digested prior to consumption. Coleopteran larvae, voracious predators upon the detritivorous adult Isopoda, also showed very high levels of all four radionuclides: 175 to 300 (^{137}Cs), around 30 Bq kg^{-1} ($^{239+240}Pu$) and 80 to 420 Bq kg^{-1} (^{241}Am). Feeding strategy is clearly the dominant factor in determining activity burdens in ground-living invertebrates, regardless of habitat and community structure. Not unexpectedly, herbivorous weevils, beetles, slugs and snails caught on at the salt marsh and sand dunes all exhibited high levels of radionuclides, for example radionuclide levels between 117 and 210 Bq kg^{-1} (^{137}Cs), 25 and 50 (^{238}Pu), 92 and 230 ($^{239+240}Pu$) and 132 and 455 Bq kg^{-1} (^{241}Am) were recorded for Gastropoda. Using *F. rubra* to calculate concentration factors within the sand dune ecosystem, snails exhibit very high accumulation rates. This probably reflects their feeding mechanism, which may encourage the ingestion of particulates adhered to the external surfaces of vegetation. In many cases, again the sand dune system provides a good example, the ^{241}Am levels in invertebrates are equivalent to or greater than ^{137}Cs, possibly reflecting differences in the bioavailability of ^{241}Am attributable to its marine origin.

Radionuclide concentration factors (predator:prey) were mostly variable across species and seasons, with only the detritivores showing any stability, and factors consistently higher than unity. The latter observation was restricted to ^{137}Cs, with the actinides showing much lower concentration factors due to their lower absorption and biological transfer coefficients. Seasonal patterns of radionuclide activity were such that, with the exception of ^{238}Pu, levels in biota were lower in the spring and summer, compared to the autumn and winter. This is thought to be coincident with changes in feeding strategy, and the increased role of higher activity food items, such as fungi, which are favoured by detritivores in the over-wintering period.

6.5 Small mammals

No significant differences were found between sexes ($p>0.05$) in respect of any radionuclide at any site so, where it was appropriate, datasets were pooled for between-site comparisons. Activity levels in wood mice (*Apodemus sylvaticus*) from the woodland site were 7 to 150 Bq

kg^{-1} (^{137}Cs). This range encompasses mean values of 55, 11 and 29 Bq kg^{-1} for ^{137}Cs for the three trapping campaigns, values which compare with <0.86 Bq kg^{-1} for the reference site remote from airborne inputs of direct airborne or secondary marine-derived radioactivity. At the salt marsh site, mean activity levels for ^{137}Cs in common shrews (*Sorex araneus*) ranged from 7 to 81 Bq kg^{-1} compared to 4 to 48 Bq kg^{-1} for wood mice (*A. sylvaticus*) and field voles (*Microtus agrestis*). These values were significantly ($p<0.01$) greater than for the control site. Activity levels for ^{137}Cs in all three small mammal species were also significantly higher along the sand dunes than at the reference site ($p<0.01$) and ranged from 13 to 26 Bq kg^{-1} (*A. sylvaticus*), 11 to 13 Bq kg^{-1} (*M. agrestis*) and 42 to 72 Bq kg^{-1} (*S. araneus*). ^{137}Cs concentrations in shrews were significantly higher than those for the other two species ($p<0.01$).

Actinide levels in *A. sylvaticus* at Lady Wood (0.07 to 0.25 (^{238}Pu); 0.14 to 0.56 Bq kg^{-1} ($^{239+240}Pu$); 0.16 to 0.41 Bq kg^{-1} (^{241}Am) were little different from the reference site, where they reflect only residual inputs from historical atmospheric weapons testing. A similar situation was noted for the salt marsh ecosystem where actinide concentrations for *A. sylvaticus*: 0.21 Bq kg^{-1} (^{238}Pu), 0.27 Bq kg^{-1} ($^{239+240}Pu$) and 0.28 Bq kg^{-1} (^{241}Am) compare with 0.42, 0.10 and 0.05 Bq kg^{-1} for the reference site. The same consistent picture emerged for the sand dune system, with actinide data very similar to the reference site at: 0.15, 0.58 and 0.20 Bq kg^{-1} (*A. sylvaticus*), 0.19, 0.26 and 0.31 Bq kg^{-1} (*M. agrestis*) and 0.16, 0.38 and 0.61 Bq kg^{-1} (*S. araneus*) for ^{238}Pu, $^{239+240}Pu$ and ^{241}Am, respectively. Only the $^{239+240}Pu$ and ^{241}Am data for *S. araneus* were significantly higher at the sand dunes than for the reference site ($p<0.05$).

Differences between the accumulation of radiocaesium and the actinides are attributed to differential mobility of the isotopes and particularly the immobilisation of ^{137}Cs by cation exchange and crystal lattice binding within the clay fraction of sediments compared to the actinides which are more associated with the organic fraction (humics) and the oxide/hydroxide components. ^{137}Cs has a higher mobility and transfer coefficient in food chains compared with members of the actinide group.

The clear between-site differences were not matched in temporal patterns, where there were significant ($p>0.05$) but less discriminating seasonal differences in body burdens within Lady Wood. These differences were found in respect of ^{137}Cs, ^{238}Pu and ^{241}Am and are thought to reflect changes in the age structure of the population and also seasonal changes in the diet of wood mice from green and invertebrate material to a diet with a greater content of contaminated fruits, berries and especially fungi. The involvement of age-accumulation is evidenced by a positive relationship between body burden of ^{137}Cs and wet weight used as a crude index of age. No differences were found in the body concentrations of actinides or

radiocaesium in animals from the riverward and seaward sectors of the salt marsh. Moreover, there was again no indication of temporal variation, probably because the age structure of the mammal populations is in balance as regards loss and recruitment. Despite the absence of inter-seasonal differences, there was considerable variation in ^{137}Cs values within each population sample, with age, diet and physiological differences between individuals the dominant factors involved. Concentrations of radiocaesium were notably higher in *S. araneus* than in *A. sylvaticus* and *M. agrestis*, which is surely a reflection of the activity levels in the invertebrates (especially *Orchestia* ssp.) comprising the diet of the shrew at this site. In contrast to the other two sites, temporal variation in ^{137}Cs levels was observed for *A. sylvaticus* and *S. araneus* from the sand dune ecosystem. This is believed to reflect seasonal changes in the relative abundance of prey items in the case of *S. araneus* , and a parallel change in the composition of the diet of *A. sylvaticus*. Temporal variation was not observed for the actinides and their generally low uptake is a result of the very low transfer coefficient across the mammalian gut. Between-site comparisons at the mammalian species level are hindered by the diet-specific considerations and the dramatic differences in the predominant prey items over the range of sites encompassed within this study.

Pilot studies into tissue distribution of radionuclides in small mammals from Lady Wood showed that the muscle and pooled viscera (kidneys, liver, spleen, heart and sex organs) were centres of accumulation of ^{137}Cs but that lung tissue was outstanding in this regard.. Activity measurements in single tissues of animals from the salt marsh showed high levels of ^{137}Cs in muscle, up to 2,100 Bq kg^{-1}, with elevated levels also in lungs (630 Bq kg^{-1}), and the pooled principal organs (700 Bq kg^{-1}). Low activity was recorded in the skeleton and gut contents, the latter reflecting the presence of artificial bait. Back calculations gave conservatively estimated whole body concentrations for ^{137}Cs of 45 to 80 Bq kg^{-1}. Measurements of the tissue distribution of ^{137}Cs at the sand dune and salt marsh sites also showed elevated levels in muscle (210 and 42 Bq kg^{-1} respectively) and lungs (200 and 75 Bq kg^{-1} respectively), and lowest levels in the skeleton at <7 Bq kg^{-1}.

6.6 Conclusions

Overall, it is notable that, despite the high levels of radionuclides measured in the soil/sediment of the three study sites, the transfer of radionuclides, in particular the actinides $^{239+240}Pu$ and ^{241}Am, to higher trophic levels is minimal. It is apparent from the data that, even if the chemical form of the actinides is different between sites, consistent with the different origins and influences upon deposited material, there is no concomitant difference in the uptake and transfer of radionuclides to the higher trophic levels in the semi-natural food chain. It appears that the uptake and concentration of the actinides is determined not by external factors but by the transfer coefficient across the mammalian gastro-intestinal tract.

This study provides a much-needed parallel to the profusion of work conducted on agricultural ecosystems and the data provide an insight into the behaviour and transfer of anthropogenic environmental radionuclides in the food chains of three semi-natural ecosystems. The absolute concentrations measured at each site were found to be low and unlikely to be of any overall significance for the health and normal functioning of the ecosystems.

Given current anxiety and the debate over the effects of ionising radiation in the natural environment and public perceptions of inadequacy about the ICRP statement made in 1977, particularly in the light of the Chernobyl incident, this work forms a baseline of knowledge. It is hoped that this will provide a focus upon which to design further research into specific areas of radionuclide transfer and behaviour in natural and semi-natural ecosystems; in particular, the development of techniques to assess the effects of radiation dose to small mammals and the development of predictive models for determining the fate of anthropogenic radionuclides released to the environment as a consequence of either an accident or routine operations of a nuclear station.

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Appendix A
CHEMICAL SEPARATION OF PLUTONIUM AND AMERICIUM

A.1 SAMPLE PRE-TREATMENT

Soil and sediment samples were air- or oven-dried until a constant dry weight was achieved. Samples were then sieved through a 2 mm mesh to remove any vegetative material and stones. A sub-sample of 5 g was placed into weighed labelled crucibles and these were placed into a muffle furnace. The bulk of the organic matter was destroyed by dry ashing the samples at 700°C for two hours.

Vegetation, leaf litter and flotsam samples were sorted after collection and oven dried to constant dry weight. The samples were homogenised in a commercial blender and a subsample of 25 g collected. These were placed in weighed labelled crucibles and dry ashed in two stages in a muffle furnace. In Stage 1 the temperature was raised to 450°C and left for two hours at which time the samples were removed from the furnace and stirred to prevent the formation of a solid crust. Samples were returned to the furnace and the temperature raised to 600°C for a further 4 hours (Stage 2) to complete all but the residual destruction of organic matter.

Invertebrate and mammal samples required a more exacting procedure to ensure the organic matter was destroyed, so as to prevent problems occurring at later stages during the chemical separation. Approximately half the quantity of sample collected was transferred into a weighed labelled crucible and placed in a muffle furnace. The temperature was raised in 4 stages: initially to 150°C and then in 100°C intervals to a final operating temperature of 450°C. At each temperature, the samples were allowed 30 minutes before being removed, stirred and returned to the furnace. At 450°C, the samples were left for 4 hours before the furnace was switched off. The samples remained in the furnace overnight and were stirred a final time the following morning. The temperature in the furnace was then raised to 450°C and the samples left for a further six hours. The samples were checked throughout the final six hours and stirred if required. Initially, problems were still encountered during the chemical separation due to the presence of organic matter. Therefore the wet ashing stage at the start of the chemistry (A.2.1) was extended. This involved placement of the samples in the beakers, addition of nitric and hydrochloric acid as given in A.2.1 and cold wet ashing for 24h before the samples were subjected to the chemical separation process.

Samples underwent the chemical separation in batches of eight. Occasionally, blank samples of 100 ml of distilled water were allowed through the separation to assess the build up of any contamination on the glassware. To minimise any contamination problems, samples were

treated as containing 'high' or 'low' activity. This was an arbitrary decision based in part upon the known activities of ^{137}Cs and ^{241}Am present from the gamma analysis and partly upon the type of samples. In general, the 'low' activity samples were considered to be small mammals, invertebrates and some vegetation, particularly from the Cheshire sites. Separate glassware was used for the high and low activity samples to reduce any possible contamination. Each sample was spiked with a radiochemical tracer. Again, either a high or low activity spike was used as appropriate, to enable chemical yields for each sample to be calculated. In general, consistent chemical yields of 85%+ and 60 to 65% were considered reasonable for plutonium and americium respectively.

A.2 CHEMICAL SEPARATION OF PLUTONIUM

A.2.1 Samples were transferred into 1000 ml beakers. 1 ml aliquots of tracer solution were added as required. ^{236}Pu and ^{243}Am were used as the tracers. These were obtained from AEA Technology, Harwell as 5 ml solutions containing approximately 200 Bq g^{-1} and were diluted either 1:200 or 1:2000 w/w for high and low activities respectively. To each sample, 2 ml orthophosphoric acid (H_3PO_4), 100 ml hydrochloric acid (HCl) and 100 ml nitric acid (HNO_3) were added. In addition, 2 ml of ferric chloride ($FeCl_3$) were added to all sample types except soil or sediment. This was required to aid the subsequent precipitation stage. For soil or sediment samples the beakers were placed on a hotplate and boiled down to dryness. For all other sample types, the beakers were placed on a hotplate and covered with a watch glass before digestion for two hours. The watch glass was then removed and the sample boiled down to dryness and left to cool. This completed the destruction of the organic matter through oxidation processes and leached the plutonium, americium and other metals from the sample matrix as most metal ions become more soluble at low pH.

A.2.2 150 ml of concentrated HNO_3 and 150 ml of distilled water were added to each beaker which was then covered with a watch glass. The beakers were returned to the hotplate and left for a further two hours. This stage re-solubilises any metal ions and converts the hydrolysed condensed phosphate to orthophosphate to provide an excess of PO_4^{3-} ions for the co-precipitation with Fe(III) (section A.2.4). After two hours, the watch glass was removed and solution was reduced in volume to 200 ml. The beakers were allowed to cool before the addition of a further 100 ml of distilled water to dilute the acid.

A.2.3 Each solution was filtered through a 240 mm Whatman No. 42 ashless filter paper into a 600 ml beaker. The 1000 ml beakers were rinsed with four 10 ml aliquots of

distilled water which also passed through the filter paper. Filtration was allowed to occur over night under gravity.

A.2.4 Two drops of hydrogen peroxide (H_2O_2) and 3-4 drops of bromocresol green indicator were added to each sample, giving an initial yellow colour. The addition of H_2O_2 oxidises the Pu (III) ions to Pu(IV) and higher because the higher oxidation states are more soluble and therefore the plutonium remains in solution. In addition, H_2O_2 ensures the all the iron present is converted to Fe(III). Concentrated ammonia solution (NH_4OH) was added by pouring with continuous stirring until the solution became a cloudy yellow. The concentrated NH_4OH was added dropwise until the solution turned blue green. The pH was adjusted to 4.9 using 2M NH_4OH solution and checked using pH 3.8-5.5 indicator strips. The final solution was a deep blue green colour and a white precipitate could be seen forming. The latter was encouraged by the addition of heat for ten minutes before the solutions were left to stand for one hour to complete the precipitation. At low pH values, hydroxides of the alkaline earth metals form. The exact pH value determines which of these metal hydroxides is insoluble. Therefore, adjusting the pH to 4.9 forms a precipitate of iron (III) phosphate ($FePO_4$). Plutonium, americium and other transition metals form complexes with the $FePO_4$ and co-precipitate.

A.2.5 Each sample was passed through a 90 mm Whatman GF/B filter paper into a 1000 ml Buchner flask in 100 ml aliquots. Vacuum filtration was applied to dry the papers. The beakers and filter papers were then washed with 300 ml of pH 4.9 solution, and 300 ml of distilled water in 100 ml aliquots. This removes most of the alkaline earth metals from the samples. The filtrates were discarded and the Buchner flasks washed with two 200 ml aliquots of tap water, 2 x 100 ml aliquots of demineralised water, 1 x 20 ml wash of distilled water and a 10 ml aliquot of concentrated HNO_3.

A.2.6 The precipitates were dissolved from the filter papers into the Buchner flasks by the addition of 6 x 10 ml washes of 8M HNO_3. The first three washes were allowed to pass through the filter paper under gravity and suction was applied for the last three. It was important to ensure that all precipitate was re-dissolved and further washes were applied if required. The use of nitric acid to reduce the pH has the additional benefit of ensuring that the plutonium ions are complexed with nitrate ions for passing through the anion exchange column in A.2.8.

A.2.7 200 mg of sodium nitrite ($NaNO_2$) were added to each flask and left for 30 minutes to oxidise Pu(III) to Pu(IV).

A.2.8 Glass anion exchange columns (60-80 mm) were prepared using glass wool, the anion exchange resin in the nitrate form. The resin type was a chromatographic grade anion exchange resin (0.15-0.3 mm) in the chloride form, which was converted to the nitrate form by washing the resin with a solution of HNO_3. The resin was conditioned with 3 x 10 ml washes of 8M HNO_3. Each wash was allowed to pass completely through the column before the addition of the next wash. The sample solution was then loaded on to the columns and allowed to completely pass through into labelled 150 ml beakers. The beakers and resin were washed with 3 x 10 ml aliquots of 8M HNO_3. All neutral and positively charged metals (which includes Am) pass straight through the column. However, Pu, in its higher oxidation states, is negatively charged and therefore binds to the resin. The raffinate was stored for use later during the chemical separation of Am if required (A.3).

A.2.9 The columns were then washed with 3 x 10 ml aliquots of 9M HCl to elute thorium from the column. Both Pu and Th ions form complexes with the nitrate but not with chloride ions. The strength of the bonds between the Pu or Th and the nitrate is also different so washing the column with 9M HCl removes Th which is less strongly bound from the resin column. Finally, the Pu was eluted from the column by washing with 3 x 10 ml aliquots of 9M HCl containing 2% hydriodic acid (HI). HI is a strong reducing agent and thus the Pu(IV) is reduced to Pu(III) making it more soluble in the 9M HCl. The samples were then prepared for electrodeposition (section A.4).

A.3 CHEMICAL SEPARATION OF AMERICIUM

A.3.1 Two dropsof H_2O_2 were added to the raffinate from A.2.9 and the solution made up to 300 mls with distilled water. pH was raised above 10 by the addition of 50% NaOH solution, whilst stirring. Samples were heated for 30 minutes. Then cooled for a further 30 minutes.

A.3.2 Each sample was passed through a 90 mm Whatman GF/B filter paper into a 1000 ml Buchner flask in 100 ml aliquots. Vacuum filtration was applied to dry the papers. The beakers and filter papers were then washed with 300 ml of pH 10 solution, and 450 ml of distilled water in 150 ml aliquots. The filtrates were discarded and the Buchner flasks washed as described in section A.2.5.

A.3.3 The precipitates were dissolved from the filter papers into the Buchner flasks by the addition of 6 x 10 ml washes of 9M HCl, H_2O_2. The first three washes were allowed to pass through the filter paper under gravity and suction was applied for the last

three. It was important to ensure that all precipitate was re-dissolved and further washes were applied if required.

A.3.4 Glass anion exchange columns (0-80 mm) were prepared using glass wool, the anion exchange was the same type as that described in A.2.8. Columns were conditioned with 4 x 10 ml washes of 9M HCl to convert it to the chloride form. The samples were passed through and the Am, being positively charged, passes through the column and is collected. The columns were then washed with 3 x 10 ml 9M HCl/H_2O_2 to complete elution of Am. 20 mg of Bi^{3+} (added as a 4 ml solution of bismuth nitrate) was added to each sample. Solutions were evaporated to dryness. 2 ml of conc^n. HNO_3 was added and the evaporation repeated. This encourages the solublisation of any metal ions present. The residue was dissolved in 10 ml of 8M HNO_3.

A.3.5 The samples were transferred into centrifuge tubes. 1 ml of orthophosphoric acid and approximately 9 ml of conc^n. NH_4OH was added very slowly whilst mixing. thoroughly. pH was adjusted to 1.6. (Produces a coprecipitate of $BiPO_4$). After cooling, the tubes were centrifuged for five minutes at 3,000 rpm. Supernatant was discarded and 20 ml of distilled water added to wash the precipitate. Centrifugation repeated and supernatant rejected again. 20 ml of 4M ammonium thiocyanate and 400 mg of hydrazinium chloride was added and the tubes shaken to ensure thorough mixing. Samples left for one hour to re-dissolve.

A.3.6 Glass anion exchange columns were prepared as in A 2.8. The resin was washed with distilled water to rinse acid free and a Whatman grade 2 filter paper placed in the column. Columns were conditioned with 3 x 10 ml washes of 4M ammonium thiocyanate (pre-treated to remove impurities). Samples were added to the columns and the raffinate discarded. Am was eluted with 6 x 10 ml washes of 1M HCl, all residue was redissolved.

A.3.7 Cation exchange columns were prepared as in A.2.8 using chromatogaphic grade cation resin (0.15-0.3 mm) in the sodium form. Resin was conditioned with 4 x 10 ml 6M HCl and 3 x 10 ml of 1M HCl washes and the sample loaded. Am was eluted with with 4 x 10 ml 6M HCl and the samples prepared for electrodeposition (A.4). This stage removes any silica impurities, which can affect electrodeposition.

A.4 ELECTRODEPOSITION

A.4.1 2 ml of concentrated HNO_3 were added to each sample. The solutions were then placed on GF/B's on a hotplate and evaporated to dryness. The samples were allowed to cool and a further 2 ml of concentrated HNO_3 was added and the samples evaporated to dryness. The addition of nitric acid helps to drive off the acids HCl and HI. It is particularly important to remove the HI otherwise problems are encountered during the electrodeposition.

A.4.2 6 ml 6M HCl and 6 ml 5% potassium hydrogen sulphate ($KHSO_4$) solution were added to each sample. Again this was evaporated to dryness. The addition of HCl re-solubilises the Pu and adds an excess of Cl^- ions. The $KHSO_4$ was added to prevent the Pu from adhering to the sides of the glass beaker.

A.4.3 The samples were allowed to cool before 20 ml of electrodeposition solution was added. The electrodeposition solution was made from 6 g ammonium oxalate ($CCONH_4.H_2O$) and 4 ml concentrated HCl diluted up to 250 ml with distilled water. The samples were then stored overnight.

A.4.4 To avoid the build up of contamination, disposable electrodeposition cells were used. These cells were prepared the day before being used and leak tested for several hours before being placed in the electrodeposition cell units. The electrodeposition cells could hold a volume of about 50 ml and held a 25 mm diameter stainless steel disc which was scribed with the sample code. The anodes were made from 1 mm diameter platinum wire and were maintained and centred at a constant height of 15 mm above the stainless steel discs.

A.4.5 Samples were loaded into the electrodeposition cells and the beakers were rinsed with four 2.5 ml washes of the electrodeposition solution. The washings were transferred to the electrodeposition cells making a total volume of 30 ml. Three drops of concentrated NH_4OH were added to adjust the pH of the solution to 2. A 400 mA current was applied across the solution and the cells were left for 7.5 hours.

A.4.6 After 7.5 hours 5 ml of concentrated NH_4OH was added to each cell. After a further 60 seconds the current was switched off and the solution emptied. The addition of concentrated NH_4OH raises the pH to around neutral and thus avoids the Pu being re-solubilised into solution when the current is turned off.

A.4.7 Each cell was then rinsed twice with distilled water and dismantled. The disc was rinsed with 20-30 ml of distilled water and dried on a hotplate. Once cool the sample was ready for counting on the alpha spectroscopy equipment.

Appendix B

OUTPUT FROM STATISTICAL TESTS

Table B1: Sexual differences in *Apodemus sylvaticus* from Lady Wood (section 3.8) using ^{137}Cs data. Mann-Whitney U Test

Month	*Sex*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	Male	4	48.2	15.4	15	0.8170
	Female	2	32.8			
July 1994	Male	4	20.8	-11.97	13	0.8170
	Female	2	32.8			
All animals	Male	13	17.8	-8.05	114	0.375
	Female	5	25.5			

Cannot reject at alpha = 0.05

Table B2: Spatial differences in Apodemus sylvaticus body burdens from Lady Wood (section 3.8). Mann-Whitney U Test.

^{137}Cs data

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	E	3	22.1	-49.2	7	0.1904
	H	3	71.3			
March 1994	E	4	11.9	1.65	8	0.8170
	He	2	10.3			
July 1994	E	4	20.80	-20.15	12	0.4875
	H	2	42.90			
All Animals	E	9	17.8	-7.4	78	0.5365
	H	9	25.2			

Cannot reject at alpha = 0.05

Actinide data from September 1993

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
^{238}Pu	E	3	0.18	0.07	13.5	0.2752
	H	3	0.11			
$^{239+240}Pu$	E	3	0.48	0.19	13	0.3827
	He	3	0.29			
^{241}Am	E	3	0.35	-0.03	10	1.0000
	H	3	0.38			

Cannot reject at alpha = 0.05

Table B3: Comparison of Reference and Lady Wood *Apodemus sylvaticus* - ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am data. Mann-Whitney U Test.

^{137}Cs

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	Lady Wood	6	34.4	32.85	57	0.0025**
	Reference	6	0.6			
March 1994	Lady Wood	6	11.0	9.54	57	0.0025**
	Reference	6	0.6			
July 1994	Lady Wood	6	28.1	26.6	57	0.0025**
	Reference	6	0.6			

** Difference is significant ($p<0.01$)

^{238}Pu

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	Lady Wood	6	0.12	-0.055	28.5	0.3973
	Reference	3	0.17			
March 1994	Lady Wood	3	0.09	-0.080	10	0.5000
	Reference	3	0.17			
July 1994	Lady Wood	3	0.07	-0.100	7.5	0.1341
	Reference	3	0.17			

Cannot reject at alpha = 0.05

$^{239+240}Pu$

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	Lady Wood	6	0.37	0.2600	38	0.0264*
	Reference	3	0.07			
March 1994	Lady Wood	3	0.14	0.06	13	0.1879
	Reference	3	0.07			
July 1994	Lady Wood	3	0.19	0.09	14	0.0920*
	Reference	3	0.07			

Cannot reject at alpha = 0.05, * difference is significant ($p<0.05$)

^{241}Am

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	Lady Wood	6	0.37	0.315	39	0.0138*
	Reference	3	0.06			
March 1994	Lady Wood	3	0.17	0.110	15	0.0383*
	Reference	3	0.06			
July 1994	Lady Wood	3	0.14	0.080	12	0.3290
	Reference	3	0.06			

Cannot reject at alpha = 0.05, * difference is significant ($p<0.05$)

Table B4: Temporal variation in *Apodemus sylvaticus* from Lady Wood. Mann-Whitney U Test.

^{137}Cs

Month	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	6	34.4	22.68	56	0.0082**
March 1993	6	11.0			
September 1993	6	34.4	12.65	44	0.4712
July 1994	6	28.1			
March 1994	6	11.0	-17.04	29	0.1282
July 1994	6	28.1			

Cannot reject at alpha = 0.05, ** difference is significant ($p<0.01$)

^{238}Pu

Month	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	6	0.12	0.020	31	0.8969
March 1993	3	0.09			
September 1993	6	0.12	0.055	37.5	0.0684
July 1994	3	0.07			
March 1994	3	0.09	0.030	13	0.3827
July 1994	3	0.07			

Cannot reject at alpha = 0.05

$^{239+240}Pu$

Month	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	6	0.37	0.225	36	0.1556
March 1993	3	0.14			
September 1993	6	0.37	0.210	36	0.1556
July 1994	3	0.19			
March 1994	3	0.14	-0.050	9	0.6625
July 1994	3	0.19			

Cannot reject at alpha = 0.05

^{241}Am

Month	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	6	0.37	0.195	37	0.0933
March 1993	3	0.17			
September 1993	6	0.37	0.245	37.5	0.0695
July 1994	3	0.14			
March 1994	3	0.17	0.030	12	0.6625
July 1994	3	0.14			

Cannot reject at alpha = 0.05

Table B5: **Assessment of spatial variation of *Festuca rubra* between the two sides of the River Esk marsh using Student *t* tests.**

	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
Mean Activity Riverward side[a]	132.9 ± 17,900.8	14.0 ± 165.1	62.3 ± 3,547.5	211.9 ± 47,688.0
Mean Activity Seaward side[a]	95.3 ± 4,446.2	9.2 ± 61.3	38.9 ± 1,064.6	130.8 ± 10,706.8
n	17	11	11	17
df	23	17	16	23
t stat	1.036	1.060	1.156	1.383
P(T<=t) one-tail	0.155	0.152	0.132	0.090
t Critical one-tail	1.714	1.740	1.746	1.714
P(T<=t) two-tail	0.311	0.304	0.264	0.180
t Critical two-tail	2.069	2.110	2.120	2.069

[a] Mean value ± variance

Table B6: **Assessment of spatial variation of *Bromus mollis* between the two sides of the River Esk marsh using Student *t* tests.**

Front transects

	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
Mean Activity Riverward side[a]	29.3 ± 116.9	2.1 ± 2.0	8.6 ± 34.2	34.9 ± 434.2
Mean Activity Seaward side[a]	18.9 ± 796.3	0.7 ± 1.1	2.8 ± 17.5	20.5 ± 1452.4
n	9, 9	7, 5	7, 5	9, 8
df	10	10	10	11
t stat	1.036	1.913	2.003	0.951
P(T<=t) one-tail	0.162	0.042	0.036	0.181
t Critical one-tail	1.812	1.812	1.812	1.796
P(T<=t) two-tail	0.325	0.05	0.073	0.362
t Critical two-tail	2.23	2.223	2.228	2.201

[a] Mean value ± variance, n reported for the two sides

Middle transects

	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
Mean Activity Riverward side[a]	13.5 ± 39.9	1.3 ± 3.6	3.9 ± 19.1	11.7 ± 95.6
Mean Activity Seaward side[a]	14.9 ± 211.4	0.8 ± 0.5	3.0 ± 14.0	11.7 ± 470.6
n	8, 6	8, 3	8, 3	8, 6
df	6	9	4	7
t stat	-0.215	0.741	0.322	-0.000
P(T<=t) one-tail	0.418	0.239	0.382	0.499
t Critical one-tail	1.943	1.833	2.132	1.895
P(T<=t) two-tail	0.836	0.477	0.764	1.000
t Critical two-tail	2.447	2.262	2.776	2.365

[a] Mean value ± variance, n reported for the two sides

Table B6: (Continued).

Back Transects

	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
Mean Activity Riverward side[a]	9.8 ± 19.0	0.2 ± 0.03	0.6 ± 0.3	1.9 ± 2.3
Mean Activity Seaward side[a]	9.6 ± 13.7	1.1 ± 2.61	1.7 ± 8.3	3.0 ± 7.7
n	9, 10	7, 6	7, 6	10, 9
df	16	5	5	12
t stat	0.114	-1.319	-0.876	-1.027
P(T<=t) one-tail	0.455	0.122	0.211	0.162
t Critical one-tail	1.746	2.015	2.015	1.782
P(T<=t) two-tail	0.911	0.244	0.421	0.325
t Critical two-tail	2.120	2.571	2.571	2.179

[a] Mean value ± variance, n reported for the two sides

Table B7: Comparison of Reference and River Esk site small mammals - ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am data. Mann-Whitney U Test. Data for animals caught on the riverward side of the marsh

^{137}Cs (Apodemus sylvaticus)

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	River Esk	6	7.4	6.1	57	0.0025**
	Reference	6	0.6			
March 1994	River Esk	6	6.45	5.8	57	0.0025**
	Reference	6	0.6			
July 1994	River Esk	4	13	11.51	34	0.0071**
	Reference	6	0.6			

** Difference is significant ($p<0.01$)

^{238}Pu (Apodemus sylvaticus)

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	River Esk	3	0.14	-0.05	9	0.3313
	Reference	3	0.17			
March 1994	River Esk	3	0.52	0.04	11	0.5000
	Reference	3	0.17			
July 1994	River Esk	2	0.11	-0.065	5	0.3864
	Reference	3	0.17			

Cannot reject at alpha = 0.05

Table B7: (Continued).

$^{239+240}Pu$ (Apodemus sylvaticus)

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	River Esk	3	0.30	0.20	15	0.0383*
	Reference	3	0.07			
March 1994	River Esk	3	0.28	0.19	15	0.0383*
	Reference	3	0.07			
July 1994	River Esk	2	0.32	0.21	9	0.0745
	Reference	3	0.07			

Cannot reject at alpha = 0.05, * difference is significant ($p<0.05$)

^{241}Am (Apodemus sylvaticus)

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	River Esk	3	0.28	0.22	15	0.0404*
	Reference	3	0.06			
March 1994	River Esk	3	0.17	0.11	15	0.0404*
	Reference	3	0.06			
July 1994	River Esk	2	0.45	0.40	9	0.0745
	Reference	3	0.06			

Cannot reject at alpha = 0.05, * difference is significant ($p<0.05$)

^{137}Cs (Microtus agrestis)

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	River Esk	4	12.45	9.45	26	0.0152*
	Reference	4	2.15			
July 1994	River Esk	5	8.5	6.75	35	0.01**
	Reference	4	2.15			

* and ** difference is significant ($p<0.05$ and $p<0.01$ respectively)

^{238}Pu (Microtus agrestis)

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	River Esk	2	1.13	0.945	7	0.1226
	Reference	2	0.19			
July 1994	River Esk	2	0.255	0.07	5	0.5000
	Reference	2	0.185			

Cannot reject at alpha = 0.05

Table B7: (Continued).

$^{239+240}Pu$ *(Microtus agrestis)*

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	River Esk	2	1.94	1.735	7	0.1226
	Reference	2	0.21			
July 1994	River Esk	2	0.77	0.56	6	0.3493
	Reference	2	0.21			

Cannot reject at alpha = 0.05

^{241}Am *(Microtus agrestis)*

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	River Esk	2	3.47	3.165	7	0.1226
	Reference	2	0.30			
July 1994	River Esk	2	1.32	1.015	6	0.3493
	Reference	2	0.30			

Cannot reject at alpha = 0.05

^{137}Cs *(Sorex araneus)*

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	River Esk	6	27.6	24.7	39	0.0141*
	Reference	3	1.5			
March 1994	River Esk	2	8.8	5.9	9	0.0745
	Reference	3	1.5			

Cannot reject at alpha = 0.05, * difference is significant ($p<0.05$)

^{238}Pu *(Sorex araneus)*

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	River Esk	3	0.39	0.28	12	0.0745
	Reference	2	0.11			
March 1994	River Esk	2	0.48	0.375	7	0.1226
	Reference	2	0.11			

Cannot reject at alpha = 0.05

$^{239+240}Pu$ *(Sorex araneus)*

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	River Esk	3	0.89	0.79	12	0.0745
	Reference	2	0.10			
March 1994	River Esk	2	0.71	0.605	7	0.1226
	Reference	2	0.10			

Cannot reject at alpha = 0.05

Table B7: **(Continued).**

^{241}Am (Sorex araneus)

Month	*Transect*	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
September 1993	River Esk	3	1.05	0.85	12	0.0745
	Reference	2	0.20			
March 1994	River Esk	2	0.62	0.42	7	0.1226
	Reference	2	0.20			

Cannot reject at alpha = 0.05

Table B8: **Differences between the three small mammal species caught at on the Sand Dunes field site. Data from September 1993.**

Transect	*n*	*Median ^{137}Cs*	*Point estimate*	*W*	*p*
Apodemus sylvaticus	6	22.9	8.95	36	0.1556
Microtus agrestis	3	13.2			
Apodemus sylvaticus	6	22.9	-21.55	21	0.0142*
Sorex araneus	4	41.85			
Microtus agrestis	9	12.9	-26.3	46	0.0054**
Sorex araneus	4	37.2			

Cannot reject at alpha = 0.05, * and ** difference is significant ($p<0.05$ and $p<0.01$ respectively).

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Radionuclide behaviour in a coniferous woodland ecosystem in Cumbria, UK.

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Abstract

The behaviour of ^{134}Cs, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am, in food chains in a semi-natural woodland has been investigated and doses to the ecosystem due to the presence of these radionuclides of anthropogenic origin have been assessed. The woodland is located within 1 km of the coastal British Nuclear Fuels plc (BNFL) reprocessing plant at Sellafield, Cumbria (O. S. Grid Reference: NY 037045) and has received an input of radionuclides primarily through atmospheric discharges from the Sellafield site throughout its operational history of more than 40 years.

Deposition has been enhanced by interception by the canopy, such that deposits in the woodland are significantly higher than adjacent pastureland. Within the wood, deposition is greatest along the front (or leading) edge in relation to aerosols transported to the woodland from Sellafield, due to the 'edge effect'. Despite the high radionuclide deposits, relatively low uptake and mobility within the ecosystem was observed. Estimated doses to the ecosystem at around 2 mGy a^{-1}, were dominated by external irradiation and were well below the levels thought to be necessary to harm terrestrial ecosystems.

A provisional conclusion at this stage is that the measures taken to control emissions from Sellafield in line with radiological protection standards for humans have also been adequate to protect this potentially vulnerable ecosystem.

Site Description

The woodland is dominated by sitka spruce, *Picea sitchensis*, of quite uniform age (planted over 40 years ago) and covers 4.7 ha with a dense and uniform canopy structure. The woodland is surrounded by agricultural land, principally pasture, and has European larch, *Larix decidua*, along the woodland fringes. Few grass and herbaceous species are present under the canopy. Those which do exist, occur primarily along the woodland edges and in clearings. Common species include: bracken, *Pteridium aquilinum*; bluebell, *Endymion nonscriptus*; white clover, *Trifolium repens*; red fescue, *Festuca rubra*; and plaintain, *Plantago lanceolata*. The remaining forest floor is covered by a 3 cm thick layer of contemporary pine needle litter overlying a profile of partially decomposed litter of approximately 5 cm depth. The soil is a modified, juvenile iron pan podzol. A slow rate of decomposition and low pH are characteristic of this soil type, in which only a limited range of bacteria, fungal and invertebrate species flourish. The woodland faces BNFL Sellafield and the sea with no major intervening natural or artificial features to intercept any airborne particulates or aerosols. Along the woodland edge facing the sources of input, there is a steep slope (33 to 150%) which presents a large surface area of canopy for the interception of radionuclides from the ambient atmospheric aerosol.

Materials and Methods

A study area comprising four line transects running parallel to the front edge of the woodland was established over an area of 50 m by 100 m. At regular time intervals between 1993 and 1995 soil cores of depth 9 cm and diameter 10 cm were removed along each transect. Samples of leaf litter, approximately 0.5 kg by wet weight, were likewise collected along each transect. All samples were sorted before oven drying at 80°C. The samples were then homogenised before presentation to a high purity germanium detector for gamma spectroscopy to determine ^{134}Cs, ^{137}Cs and ^{241}Am activities. ^{238}Pu and $^{239+240}Pu$ levels were determined after radiochemical separation by alpha spectroscopy.

Invertebrates were collected using pitfall traps containing 100 ml of 2% formalin. Six traps were set along each transect and the samples were pooled over quarterly time intervals to obtain sufficient biomass for each taxonomic group. Small mammals were trapped using Longworth live traps placed throughout the woodland along the transects. Adult specimens were culled and immediately frozen to prevent tissue degeneration. Invertebrates and small mammals were washed prior to being dried, pooled and then homogenised

prior to radionuclide determination. Small mammals from the final trapping session were dissected to excise specific tissues in order to assess the distribution of ^{137}Cs within the body.

Results and Discussion

Soils

Soils in the woodland show evidence of the accumulation of radionuclides due to atmospheric deposition over an extended period of time. Accumulated deposits vary geographically and lie within the range 250 to 320 Bq m^{-2} (^{134}Cs), 2,600 to 53,000 Bq m^{-2} (^{137}Cs), 160 to 470 Bq m^{-2} (^{238}Pu), 3,600 to 5,500 Bq m^{-2} ($^{239+240}Pu$) and 1,100 to 2,900 Bq m^{-2} (^{241}Am) (Table 1). These inventories are consistent with other measurements in the immediate vicinity, although the ^{137}Cs levels in the woodland are much higher than would be expected given the general pattern of decline with distance from Sellafield [1,2,3].

Table 1: Comparison of deposition values (Bq m^{-2}) from samples collected in April 1993 with previous studies.

Transect	*Sample Date*	*^{134}Cs*	*^{137}Cs*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*	*Reference*
Base of front slope	1993	250	53,000	470	5,500	2,900	This study
Top of front slope	1993	230	36,000	280	5,100	1,600	"
Middle	1993	260	26,000	160	3,600	1,100	"
Back	1993	320	32,000	230	4,000	1,400	"
This site (top 15 cm)	1978		6,100	93	3,500	---	[1]
This site (30 cm cores)	1987	---	---	180	7,000	1,300	[3]
This site (30 cm cores)	1987	---	39,000	---	---	---	Cawse, unpublished

All radionuclides, with the exception of ^{134}Cs, show a consistent, and significant ($p<0.001$), decline in accumulated deposit from the 'front' to the 'back' of the wood (in relation to Sellafield complex: Table 2); this is attributed to the 'edge effect' whereby greater deposition occurs at the leading edge of the woodland. Several studies have investigated the interception of radionuclides by leaf canopies which are effective accumulators of atmospheric radioactivity [4]. Interception is maximised at the woodland edge where disruption of the air flow is at its greatest [5,6]. In the present study, between 65 and 70% of the total deposited activity for ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am was found along the front aspect of the woodland.

Table 2: Mean whole core activity (Bq kg^{-1}) for woodland soils (± s.e.), n=3.

Date	*Transect*	*^{134}Cs*	*^{137}Cs*	*^{238}Pu*	*$^{239+240}Pu$*	*^{241}Am*
April 1993	Base of front slope	6.8 ± 1.8	1,500 ± 430	13.0 ± 2.8	160 ± 33.0	86 ± 25
	Top of front slope	4.4 ± 0.8	720 ± 170	5.5 ± 1.1	102 ± 18.0	31 ± 6.4
	Middle	3.9 ± 0.4	400 ± 25	2.5 ± 0.2	56 ± 1.2	17 ± 2.2
	Back	4.4 ± 1.4	460 ± 150	3.4 ± 1.0	61 ± 15.0	21 ± 7
September 1994	Base of front slope	2.5 ± 0.1*	650 ± 66	6.4 ± 0.1	98 ± 0.5	45 ± 4.2
	Top of front slope	4.3 ± 1.0	750 ± 350	7.9 ± 1.3	96 ± 18.0	42 ± 17.0
	Middle	3.0 ± 0.4	520 ± 150	3.1 ± 0.4	53 ± 3.8	25 ± 5.3
	Back	3.3 ± 1.3	430 ± 92	3.8 ± 1.0	59 ± 7.4	30 ± 6.2

* Limit of Detection values used during calculations

Much of the ^{134}Cs deposited in the woodland appears to originate from the Chernobyl incident in 1986, since back-calculating the ^{134}Cs results to May 1986 and using a ^{137}Cs:^{134}Cs ratio of 1.7 [7] implies that Chernobyl fallout accounts for about 4,500 Bq m^{-2}, or about 10-15% of the ^{137}Cs deposit in woodland. This agrees with the range of deposition values due to Chernobyl measured during 1986 [8].

The woodland deposits of plutonium in soil are characterised locally by low ^{238}Pu:$^{239+240}Pu$ ratios, in the range 0.039 to 0.086. This typifies plutonium produced from uranium of low irradiation, which indicates that much of the deposition results from the early years of operation of the Sellafield site.

Leaf litter

Pine needle litter in the woodland shows elevated levels of all radionuclides, ranging from 2 to 10 Bq kg^{-1} (^{134}Cs), 130 to 1,100 Bq kg^{-1} (^{137}Cs), 2 to 14 Bq kg^{-1} (^{238}Pu), 15 to 110 Bq kg^{-1} ($^{239+240}Pu$), and 18 to 105 Bq kg^{-1} (^{241}Am). Given the hypothesis that much of the deposition to soils is historic, concentrations are surprisingly high in relation to underlying soils, the ratio between leaf litter and soil concentrations typically lying between 0.3 and 1.0. Furthermore, direct deposition of airborne particulates to the forest floor is unlikely because of the density of the canopy. Recent work on spruce canopies in wind tunnel experiments has shown that over 99% of the airborne particulate material can be intercepted by the tree canopy where radionuclides may be absorbed into the leaves or remain bound to the external leaf surface [6]. Subsequent redistribution of radionuclides to the forest floor may occur through rainfall dissolution of adsorbed particulates on leaf material or as a result of leaf fall.

The elevated concentrations in needle litter cannot be explained by adventitious soil contamination, and there are significant differences between isotopic ratios of ^{134}Cs:^{137}Cs and ^{238}Pu:$^{239+240}Pu$. These imply that contamination on the needle litter is of more recent origin than that present in the soil. Table 3 compares the soil and leaf litter activity. The ^{238}Pu:$^{239+240}Pu$ ratio clearly indicates that the origins of the radionuclides deposited in the soil and on the leaf litter are clearly different.

Table 3: Comparison of the soil and leaf litter data (Bq kg^{-1}) from transects 1 and 2.

Radionuclide	*Transect 1*		*Transect 2*	
	Soil	*Leaf Litter*	*Soil*	*Leaf Litter*
^{137}Cs	1500 - 650	1100 - 300	460 - 390	560 - 130
$^{239+240}Pu$	160 - 98	110 - 25	61 - 50	20 - 15
^{241}Am	86 - 45	105 - 50	30 - 15	38 - 18
Isotopic Ratios	*Soil*	*Leaf Litter*	*Recent Deposition*	
^{137}Cs:^{134}Cs	169:1 (± 20)	100:1 (± 14)	*	
^{238}Pu:$^{239+240}Pu$	0.07:1 (± 0.01)	0.16:1 (± 0.01)	0.16:1 (± 0.02)	

* ^{134}Cs measurements of aerial discharges are not routinely monitored by Sellafield (Evans, pers. comm.).

Although the relative levels of contamination on needle litter and soil cannot fully be explained, some researchers have suggested that mechanisms involving fungal mycelia can lead to reconcentration of material into the litter layer. The role of fungi in cycling ^{137}Cs in ecosystems is known to be important. For example, it has been estimated that up to 30% of the ^{137}Cs present in soil may be located within fungal mycelia [9], although this is likely to be species specific.

Concentrations in needle litter show the same spatial variation as concentration in soils, with a fall in concentrations from the 'front' to 'back' edge of the wood, confirming that interception of aerosols by the woodland canopy is a dominant mechanism in deposition processes at the site.

Invertebrates

Whole body burdens of invertebrates from the woodland varied between species and pooled taxonomic groups, in the range 6-1800 Bq kg^{-1} (^{137}Cs), 0.3-148 Bq kg^{-1} (^{238}Pu), 0.3-59.4 Bq kg^{-1} ($^{239+240}Pu$) and <0.3-400 Bq kg^{-1} (^{241}Am) (Table 4). The low levels precluded quantification of ^{134}Cs.

The detritivorous fauna, arguably the most important in a coniferous woodland ecosystem [10], showed higher values than for other taxa, namely 43-1800 Bq kg^{-1} (^{137}Cs), 1-14.5 Bq kg^{-1} (^{238}Pu), 6.5-74 Bq kg^{-1} ($^{239+240}Pu$) and <0.3-400 Bq kg^{-1} (^{241}Am). In the case of ^{137}Cs and ^{241}Am, the highest values were for Oligochaete (earthworms) wherein the gut contains residual quantities of contaminated soil ingested involuntarily along with fragments of food [10]. Isopods also showed strongly elevated levels of all measurable nuclides, reflecting the level of contamination of their litter food source.

Table 4: Radionuclide activities (Bq kg^{-1} ± 2σ counting error) in invertebrate samples caught between April and August 1993.

	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
Predators				
Araneida	40 ± 15	< 1	10 ± 1	< 3
Opiliones	115 ± 10	16 ± 2	11 ± 2	6 ± 2
Carabidae	25 ± 5	0.6 ± 0.1	5 ± 0.4	1.6 ± 0.4
Herbivores				
Coleoptera	12 ± 5	n/a	n/a	< 1
Detritivores				
Oligochaeta	870 ± 135	n/a	n/a	15 ± 6
Oniscus asellus	120 ± 20	8 ± 1	15 ± 1	30 ± 8

n/a Sample not analysed. < Indicates value below the calculated limit of detection.

Seasonal patterns of radionuclide activity were such that, with the exception of ^{238}Pu, levels in biota were lower in the spring and summer, compared to the autumn and winter. This is thought to be coincident with changes in feeding strategy, and the increased importance of higher activity food items, such as fungi, which are favoured by detritivores in the over-wintering period [12]. Of the predatory species, the Araneida and Opiliones showed the highest levels of ^{137}Cs. The high levels in spiders, compared to beetles at a similar trophic level, may reflect their exo-enzyme digestion system whereby the soft tissues of their prey are digested externally and then ingested as a partly processed foodstuff. ^{137}Cs is preferentially associated with soft, as opposed to skeletal, tissues [13].

Radionuclide concentration factors (predator:prey) were mostly variable across species and seasons, with only the detritivores showing any stability, and factors consistently higher than unity. The latter observation was restricted to ^{137}Cs, with the actinides showing much lower concentration factors due to their lower absorption and biological transfer coefficients.

Small mammals

Activity levels in wood mice (*A. sylvaticus*) from the woodland site were 7.1-150 Bq kg^{-1} (^{137}Cs), 0.07-0.25 Bq kg^{-1} (^{238}Pu), 0.14-0.56 Bq kg^{-1} ($^{239+240}Pu$) and 0.16-0.41 Bq kg^{-1} (^{241}Am). No significant differences ($p>0.05$) were found between sexes in respect of any nuclide, so datasets were pooled for between site comparisons (Table 5). The range for ^{137}Cs encompasses mean values of 55, 11 and 29 Bq kg^{-1} for the three trapping campaigns, values which compare with <0.86 Bq kg^{-1} for the reference site remote from airborne inputs of industrial or secondary, marine-derived radioactivity. Actinide levels at the woodland location were little different from the reference site, where they reflect only residual inputs from historical atmospheric weapons testing and the Chernobyl event.

Table 5: Mean radionuclide activities for *Apodemus sylvaticus*. Values in Bq kg^{-1} dry weight (± standard error).

Sample Code	^{137}Cs	^{238}Pu	$^{239+240}Pu$	^{241}Am
September 1993	55 ± 20	0.2 ± 0.003	0.56 ± 0.01	0.41 ± 0.01
March 1994	11 ± 2	0.3 ± 0.01	0.14 ± 0.001	0.21 ± 0.01
July 1994	29 ± 8	0.07 ± 0.001	0.17 ± 0.01	0.16 ± 0.01
Reference Site	<0.86	0.42 ± 0.02	0.10 ± 0.003	0.05 ± 0.01

The clear differences in levels of ^{137}Cs in wood mice from the woodland and reference site reflect not just the differences in contemporary exposure conditions for this nuclide at the Sellafield and reference locations, but also the higher mobility and transfer coefficient of caesium in comparison with members of the actinide group in food chains. The clear between-site differences were not mirrored in temporal patterns, where

there were significant ($p>0.05$) but less discriminating seasonal differences in body burdens within the woodland site.

These seasonal differences were found in respect of ^{238}Pu, ^{241}Am and particularly ^{137}Cs and are thought to reflect changes in the age structure of the population, and also seasonal changes in the diet of wood mice from green and invertebrate material to a diet with a greater element of contaminated fruits, berries and especially fungi [14]. The involvement of age-accumulation as a factor in apparent seasonal differences is evidenced by a positive relationship between the body burden of ^{137}Cs and wet weight used as a crude index of age ($r=0.78$; $p<0.01$). Body burdens of actinides showed much less variation with season, probably due to the lower transfer potential of these nuclides regardless of dietary changes during the year.

Activity measurements in single tissues showed very high levels of ^{137}Cs in muscle, up to 2100 Bq kg^{-1}. This is not unusual as the muscle is a recognised site of accumulation for ^{137}Cs [15]. Elevated levels of ^{137}Cs were also found in the lungs (630 Bq kg^{-1}), and the pooled principal organs (spleen, liver, kidneys and heart) (700 Bq kg^{-1}). Low activity was recorded in the skeleton and gut contents, the latter reflecting the presence of artificial bait. Back calculations gave conservatively estimated whole body concentrations for ^{137}Cs of 45-80 Bq kg^{-1}.

Doses to the ecosystem

Doses to biota were estimated from measured concentrations of radionuclides in biota and soil, using values representative of the upper quartile of the measured ranges, as indicated in Table 6.

Table 6: Radionuclide concentrations for dose estimation (Bq kg^{-1} wet weight)

Nuclide	*Invertebrates*		*Mammals*	*Soil*
	Predators	*Detrivores*	*Woodmice*	
^{137}Cs	23	300	20	1000
$^{239+240}Pu$	4	5	0.2	100
^{241}Am	1	8	0.5	50

Internal doses were calculated by evaluating the absorbed energy from decays within the body, using absorbed fractions calculated for representative idealised geometries [16]. External doses were calculated from concentrations in soil using the standard formulae for a semi-infinite absorbing medium [17]; this will overstate the dose from ^{137}Cs gammas by a small factor since most of the ^{137}Cs is contained within the upper 15 cm of the soil horizon. The resulting doses are summarised in Table 7.

Table 7: Doses to biota (mGy a^{-1})

Nuclide	*Invertebrates:*		*Mammals:*
	Predators	*Detrivores*	*Woodmice*
Internal doses:			
$^{137}Cs(g)$	2.E-4	2.E-3	9.E-4
$^{137}Cs(b)$	2.E-2	3.E-1	2.E-2
$^{239+240}Pu(a)$	1.E-1	1.E-1	5.E-3
$^{241}Am(a)$	3.E-2	2.E-1	4.E-3
External dose	2.E+0	2.E+0	2.E+0

The calculated doses are much lower than the level of 1 mGy d^{-1} below which damage to terrestrial ecosystems in considered unlikely [18]. It is noteworthy that external doses (beta plus gamma) from ^{137}Cs are much higher than the corresponding internal doses. Internal doses for invertebrates from the actinides are much higher than the corresponding doses for woodmice; depending on the relative biological effectiveness for alpha radiation in these biota the internal doses in this case may be comparable in significance to the external doses.

Conclusions

The site has been subject to relatively high levels of deposition due to past discharges to atmosphere from Sellafield. This deposition has been enhanced by interception by the canopy, such that deposits in the woodland are significantly higher than adjacent pastureland. Within the wood, deposition is greatest along the front (or leading) edge in relation to aerosols transported to the woodland from Sellafield, due to the 'edge effect'. Despite the high radionuclide deposits, relatively low uptake and mobility within the ecosystem was observed. Estimated doses to the ecosystem at around 2 mGy a^{-1}, were dominated by external irradiation and were well below the levels thought to be necessary to harm terrestrial ecosystems.

A provisional conclusion at this stage is that the measures taken to control emissions from Sellafield in line with radiological protection standards for humans have also been adequate to protect this potentially vulnerable ecosystem.

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