Institute of Terrestrial Ecology RADIONUCLIDES IN TERRESTRIAL ECOSYSTEMS

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Natural Environment Research Council Institute of Terrestrial Ecology

# RADIONUCLIDES IN TERRESTRIAL ECOSYSTEMS

A review of their distribution and movement

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The Institute of Terrestrial Ecology (ITE) was established in 1973 from the former Nature Conservancy's research stations and staff, joined later by the Institute of Tree Biology and the Culture Centre of Algae and Protozoa. ITE contributes to and draws upon the collective knowledge of the fourteen sister institutes which make up the Natural Environment Research Council, spanning all the environmental sciences.

The Institute studies the factors determining the structure, composition and processes of land and freshwater systems, and of individual plant and animal species. It is developing a sounder scientific basis for predicting and modelling environmental trends arising from natural or man-made change. The results of this research are available to those responsible for the protection, management and wise use of our natural resources.

Nearly half of ITE's work is research commissioned by customers, such as the Nature Conservancy Council who require information for wildlife conservation, the Department of Energy, the Department of the Environment and the EEC. The remainder is fundamental research supported by NERC.

ITE's expertise is widely used by international organisations in overseas projects and programmes of research.

Captions for cover plates. From the top, clockwise: Saltmarsh, Photo. A. D. Horrill; Mixed woodlands, Photo S. E. Allen; Ravenglass estuary, looking north, Photo. C. Quarmby; Oystercatchers feeding on mudflats, Photo. J. D. Goss-Custard; Upland pasture, Photo. A. D. Horrill.

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This report summarizes information on the distribution and movement of radionuclides<sup>\*</sup> in semi-natural terrestrial ecosystems in north-west England with particular emphasis on inputs to, and outputs from ecosystems; on plant and soil aspects; and on radionuclides in fallout and in discharges by the nuclear industry.

Radionuclides may enter terrestrial ecosystems along several pathways:

- (a) Deposition in precipitation of particles and dissolved gases or vapours.
- (b) Dry deposition of particles of resuspended dust or fine droplets (aerosols) from soil, vegetation and water surfaces.
- (c) Deposition of mineral matter, seaweed and other flotsam on the seashore and around freshwater courses and lakes.
- (d) Transfer in birds, other vertebrates or invertebrates which feed in the sea or on the shore, but which occasionally move inland.
- (e) Transfer from the sea in fish or shellfish harvested by man.
- (f) Removal of material from the shore by man and deposition inland, e.g. seaweed as fertilizer, and sand.
- (g) Use of fertilizers, especially phosphate.
- (h) Burial of radioactive waste.

(a), (b), (c), (g) and (h) are likely to be the main input pathways.

Radionuclides deposited on plants and on soil surfaces occur in various chemical forms in particles, as vapours or in solution. Each nuclide may be deposited in more than one chemical or physical form. Retention by a plant depends on the surface characteristics and the growth form of the plant. Wind removes all particles  $>44 \ \mu$ m deposited on the plant. As much as 83-90% of deposited water-soluble radionuclides may be lost from plants in a month, with little difference in loss between plant species and radionuclides. Retention half-life for radioactive aerosols and fine particles in various agricultural and non-agricultural vegetation is from 13-87 days.

The degree of retention of stable radionuclides by soil varies with the physical and chemical characteristics of the element involved, the physical and chemical form of the nuclide, the concentrations of the nuclide and of ions which compete with nuclide ions for retention sites, soil pH, soil organic matter content, soil mineral type, concentrations of chelating agents or other compounds which form complexes with the nuclide, and the time allowed for equilibration of the nuclide between solid and liquid phases.

Nuclides are retained by soils in water films, in hydrated oxide or humus on particle surfaces, and in the crystal lattice of soil minerals. They occur in solution, as cations, anions or complexes with mineral or organic matter, and as particles.

Sr\*\* radionuclides are retained by soil largely in solution or on minerals or organic matter in an ion-exchangeable form. Cs, Ru, Zr, Nb, Ce and other rare earths, and the transuranides are held more strongly than Sr, particularly by soil minerals.

Sesquioxides appear to be especially important in retention of I. Both I and Ru can occur in anionic form and are held strongly by acid organic matter. Maximum retention by soil occurs at pHs 7-11 (Sr), 3.5-11 (Cs), 1-5 and >8 (Zr + Nb), 6-8 and > 11 (rare earths), 4-9 (transuranides) and 5.5-8.5 (I). The effect of pH on Ru retention by soil varies with the experimental conditions. Chelating agents affect retention and migration of Sr significantly only in soils low in clay. They have little effect on Cs and Ru retention but promote desorption of the rare earths and transuranides.

The distribution of radionuclides in soil profiles is affected by several factors including retention characteristics of the nuclides, precipitation, temperature, moisture, soil composition and hence porosity and aeration, degree of soil fissuring, soil biological activity, soil cultivation and fertilizer application.

Radionuclides may be absorbed by plants either through the leaves, the stems, the flowers and fruits or via the roots. Absorption by the above-ground parts appears to have been little studied although many data are available on gross retention of 'natural' or simulated fallout by these parts. These indicate that 4-75% of radioactivity in plants in the field is derived from direct deposition, the value varying with radionuclide and plant type.

Absorption of radionuclides by plant roots varies with the many factors which affect retention and distribution of the nuclides in soil. Absorption varies by one to several orders of magnitude between plant species, soil types and with radionuclide according to the following order  $Sr \ge 1 > Cs \simeq Ru > Ce \simeq Y \simeq Pm \simeq Zr +$ Nb>Pu. Some elements, e.g. Cs, are mobile in all

\*A RADIONUCLIDE is any species of atom which exists for a measurable time, which is distinguished from other species atoms by the constitution of its nucleus, and which emits radiation by spontaneous transformation of its nucleus.

\*\*For explanation of symbols see Table 1.

directions in the plant whereas others, e.g. Sr, are mobile largely from root to shoot, and yet others, e.g. Zr, tend to remain near the site of absorption. Cs and Sr, consequently, tend to become evenly distributed throughout plants, whereas Zr becomes concentrated in the roots.

Absorption by plant roots varies with distribution of the roots in the soil profile. Rye-grass, a shallowrooting species, for example, absorbs decreasing amounts of <sup>90</sup>Sr over several years as the nuclide migrates downwards from the point of application. Lucerne, a deep-rooting species, absorbs less <sup>90</sup>Sr than rye-grass when the contamination is near the soil surface. Ploughing or soil mixing by earthworms changes the distribution of radioactivity in the soil profile and hence would be expected to affect plant uptake.

The absorption of some radionuclides, e.g. Pu, by plants increases with time, possibly as a result of changes in availability of the radionuclide during the root growth/decay cycle. Micro-organisms may be involved as some are capable of solubilizing unavailable radionuclides, e.g. in Pu compounds.

Uptake and retention of radionuclides by animals vary greatly with radionuclide and animal type. Vertebrates and those invertebrates with a calcareous exoskeleton or shell, for example, have a higher demand for Ca and Sr than other invertebrates. Uptake occurs via respiratory surfaces, the skin and the gastrointestinal tract of animals. Elements such as Ca and Sr, which are readily taken up by plants from the soil, are also readily absorbed in the animal gut whereas others, e.g. the transuranides, are absorbed negligibly. The distribution of absorbed radioactivity in the animal body is usually very uneven and varies with the radionuclide and animal involved. Elimination of radionuclides by the animal is a complex process and occurs at different rates from different parts of the body.

Rates of release of radionuclides during decomposition of plant remains vary with the radionuclide involved. <sup>85</sup>Sr and <sup>106</sup>Ru, for example, are lost at the same rate as dry matter whereas <sup>134</sup>Cs is lost at more than twice that rate as a result of leaching by rain. Like stable nuclides, radionuclides are immobilized by microorganisms in plant remains. Retention half-life for radionuclides in decomposing animal remains appears to vary between a day or two in soft tissues to many thousands of years in bones and shells.

Radionuclides may be lost to the air as gases, vapours and in resuspended soil particles; to freshwater and the sea in solution or in eroded soil; and to other ecosystems in crops, timber and animal products. Information on losses of dry matter from UK ecosystems is sometimes available but losses of radionuclides appear to be unknown. 0.35-4.5% of the annual fallout input of <sup>90</sup>Sr is lost annually in eroded soil and solution from some agricultural soils in USA. 46-57% of the incoming <sup>90</sup>Sr is lost from other agricultural soils. Eroded soil is about ten times richer in <sup>90</sup>Sr than the uneroded soil. A strong association between soil erosion and radionuclide loss occurs also for <sup>137</sup>Cs and <sup>23 9, 240</sup>Pu in USA.



Figure 1. The main pathways along which radionuclides are transferred into, through and out of terrestrial ecosystems.

### INTRODUCTION

This report is the outcome of a study by the Institute of Terrestrial Ecology (ITE) of the Natural Environment Research Council funded by the Department of the Environment. The main objective was to summarize and collate the relevant information on the distribution and movement of radionuclides in semi-natural terrestrial ecosystems in the United Kingdom, with particular reference to north-west England.

In the late 1950s and 1960s, during and immediately after the period of most frequent above-ground tests of nuclear weapons, the distribution and movement of fallout radionuclides in air, precipitation, agricultural produce and soils in the UK were intensively and extensively studied by the Agricultural Research Council (ARC) Letcombe Laboratory, the Atomic Energy Research Establishment (AERE) Harwell and the Ministry of Agriculture, Fisheries and Food (MAFF) (Russell, 1966a). Semi-natural ecosystems, e.g. woodlands and upland grazings, received little attention.

Monitoring by AERE of radionuclides in air and precipitation over the whole of the UK has continued but, until recently, other terrestrial radioecological studies have been restricted largely to routine monitoring by ARC, British Nuclear Fuels Ltd. (BNFL) and the Central Electricity Generating Board (CEGB) of radionuclides in air, vegetation, agricultural produce and soils, mainly in agricultural areas in the vicinity of nuclear installations. The National Radiological Protection Board (NRPB) has now assumed responsibility for monitoring the radioactivity of milk and other agricultural products, previously the concern of ARC.

With the development of the UK nuclear power industry. during the 1960s and 1970s, the amount of radioactive waste has increased. The majority of this waste is now stored for ultimate disposal in stable sites such as certain deep geological strata. The bulk of the unstored waste is discharged to the sea or, to a much lesser extent, to freshwater (Taylor et al., 1976). The fuel reprocessing plant at Windscale discharges more waste than any other nuclear installation in the UK, hence current marine and terrestrial ecological studies are centred on the north-east Irish Sea and west Cumbria. These studies received added stimulus from the Sixth Report of the Royal Commission on Environmental Pollution (1976) and the Windscale Enquiry (Parker, 1978) and involve AERE Harwell, BNFL, Lancaster University, the MAFF Fisheries Radiobiological Laboratory (FRL), MAFF Food Sciences Division, Natural Environment Research Council (Institute of Geological Sciences, Institute of Marine Environmental Research and Institute of Oceanographic Sciences), and NRPB.

Possible transfer of radioactivity from sea to land in aerosols and resuspended estuarine sediment is being examined separately by AERE with BNFL, by NRPB and by Lancaster University. MAFF is measuring levels of radioactivity in a range of environmental materials from agricultural areas, primarily agricultural products. AERE is examining radionuclide concentrations in soils and vegetation in semi-natural ecosystems over the whole of the UK and, more intensively, in Cumbria, and is making some measurement of radionuclide uptake by plants. ITE is assisting AERE in some aspects of these studies (see below). The continuing monitoring of the terrestrial environment in the UK by BNFL, AERE and NRPB is intended to detect accidental releases of radioactivity and changes in the concentrations of radionuclides in the long term.

ITE's particular interest in terrestrial environmental radioactivity is in the transfer and distribution of radionuclides in ecosystems (Figure 1), in the processes involved in these transfers and in the factors which affect both transfer and distribution, e.g. the activities of organisms; physical factors such as temperature; chemical factors such as soil acidity; or complexes of factors associated with land management or land use. The ecosystems of interest range from littoral areas, such as salt marshes, to mountain tops and include, where necessary for comparative purposes, agricultural systems, commercial forests, grazed upland and related freshwater aspects. In the case of littoral and freshwater Biological Association and Lancaster University.

The radionuclides considered in this report are those occurring in fallout and particularly in discharged or stored waste and accidental emissions from nuclear installations (Table 1). Relevant published information is abundant, hence emphasis will be placed on those radionuclides which are associated with the nuclear fuel cycle in the UK (Royal Commission on Environmental Pollution, 1976; British Nuclear Fuels Ltd., 1978) and which are considered to present the greatest potential hazard to man and other organisms (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 1977; Parker, 1978). These include particularly the nuclides which are long-lived and hence which are most likely to become involved extensively in environmental pathways (Table 1). Tritium (<sup>3</sup>H) and radiocarbon (<sup>14</sup>C) fall into this category but for practical reasons their ecology cannot be examined by ITE in the near future and they will not be considered here.

The noble gases, <sup>41</sup>Ar, <sup>85</sup>Kr, <sup>133</sup>Xe and <sup>222</sup>Rn, become generally distributed in the environment and enhance background radiation but they are biologically inert and hence will not be considered here.

ITE's studies of the functioning of ecological systems may be greatly facilitated by regarding pollutant radionuclides as labels or tracers in natural processes. The Institute already uses bomb <sup>14</sup>C in dating organic materials (Harrison and Harkness, 1976). Studies of the distribution and movement of <sup>33</sup>S and <sup>131</sup>I or <sup>129</sup>I from nuclear installations may help our understanding of the environmental behaviour of the related stable nuclides. In America, knowledge of soil erosion processes has been increased by studying Pu movement (Muller *et al.*, 1978; Sprugel and Bartelt, 1978) and of movement and effect of stemflow water in forest soils by studying Cs movement (Gersper, 1970).

In expanding its research interest to include the environmental behaviour of radionuclides, the Institute will use its previous experience and relevant expertise in studies of the distribution and flow of plant nutrients and energy in ecosystems, mathematical modelling of nutrient pathways, chemical analysis of environmental materials, use of radiotracers in plants and animals, examination of plant-soil-nutrient relationships, and studies of the environmental behaviour of pollutants.

Recent data on the concentrations and distribution of pollutant radionuclides in the environment indicate

that further detailed studies of the behaviour of radionuclides in terrestrial ecosystems are justified not solely in relation to current hazards to man and other organisms but more generally in relation to assessment of future possible hazards to organisms. The recent unexpected finding of concentrations of radioactivity above maximum permitted levels in the bodies of islanders from Bikini after the islands had been decontaminated to apparently 'safe' levels (Robinson et al., 1977) emphasises the continual need for environmental pathway studies even in much studied areas. This general principle applies more broadly in the less extreme situation in the UK and other countries exposed to low levels of ambient radioactivity, but subject to possible higher levels of radionuclides released accidentally.

References and keywords for the literature examined in preparation of this report have been stored in a computer-based information bank described in the Appendix.

Table 1. The main radion	uclides present in the	IK environment from	nuclear devices	and the nuclear	industry
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Element	Radio- nuclide	Half-life (a)	Type of emission	Energies of emission (MeV) (b)	Main sources and discharge routes (c)
HYDROGEN		12·3 y	β	U·019	F, PL, RL, PA, RA
CARBON	<sup>14</sup> C	5730 y	β	0.156	F, RL, PA, RA
PHOSPHORUS	<sup>32</sup> P	14·3 d	β	1.71	RL
SULPHUR	<sup>35</sup> S	88 d	β	0.167	RL, RA
ARGON	<sup>41</sup> Ar	1 · 83 h	β Υ	<i>1 · 20,</i> 2 · 49 1 · 29	PA, RA
CALCIUM	<sup>45</sup> Ca	165 d	β	0-258	RL
CHROMIUM	<sup>51</sup> Cr	27 · 8 đ	Υ EC	0·32 0·752	RL
MANGANESE	<sup>54</sup> Mn	303 d	Ŷ	0.835	F, RL
IRON	<sup>55</sup> Fe <sup>59</sup> Fe	2 · 6 y 45 · 1 d	EC EC β Υ	0·232 0·273, 0·475, 1·57 0·142-1·29 (1·10, 1·29)	F, RL RL
COBALT	<sup>58</sup> Co	71 · 3 d	β Υ	0·474 0·810, 0·864, 1·67 2·21	RL
	<sup>60</sup> Co	5·3 y	β Υ	2 · 31 <i>O · 315,</i> 1 · 49 1 · 17, 1 · 33	F, RL
NICKEL	<sup>63</sup> Ni	92 y	β	0.067	RL
ZINC	<sup>65</sup> Zn	244 d	β Υ Υ ΕC	0·325 1·11 1·11	RL
ARSENIC	<sup>76</sup> As	1 · 1 d	β Υ	0·35-2·96 ( <i>2·40. 2·96</i> ) 0·510-2·66 ( <i>0·559</i> )	RA
KRYPTON	<sup>85</sup> Kr	10·8 y	β Υ	0 · 67 0 · 14	F, PA, A

Element	Radio- nuclide	Half-life (a)	Type of emission	Energies of emission (MeV) (b)	Main sources and discharge routes (c)
STRONTIUM	<sup>89</sup> Sr <sup>90</sup> Sr	52 d 28 • 1 y	β β	1 · 46 0 · 546	F, PL, <b>RL</b> , A F, PL, <b>RL</b> , PA, A
YTTRIUM	90Y	2 · 67 d	β	2 · 27	RL, daughter of <sup>90</sup> Sr
	<sup>91</sup> Y	58·8 d	β Υ	0 · 33, <i>1 · 55</i> 1 · 21	PL, RL, A'
ZIRCONIUM	<sup>95</sup> Zr	65 d	β Υ	0 • 360 - 1 • 1 3 <i>(0 • 360, 0 • 396</i> ) 0 • 236, 0 • 723, 0 • 757	F, PL, RL, PA, A'
NIOBIUM	<sup>95</sup> Nb	35 · 1 d	β Υ	0·160 0·766	F, PL, RL, PA, A' daughter of <sup>95</sup> Zr
TECHNETIUM	<sup>99</sup> Tc	2 · 12 × 10⁵ y	β	0.292	F, UEL
RUTHENIUM	<sup>103</sup> Ru	'39·6 d	β	<i>0·203-</i> 0·90	F, PL, A
	<sup>106</sup> Ru	367 d	Υ β Υ	0·040-0·610 ( <i>0·497</i> ) 0·039 0·512-2·64 ( <i>0·512, 0·616</i> )	PLA, A (Y from daughter <sup>106</sup> Rh, 30s half-life)
SILVER	<sup>110</sup> mAg	253 d	β Υ ΙΤ	<i>0 · 087,</i> 0 · 529, 1 · 50 0 · 657, 0 · 818, 1 · 48 0 · 12	RL
ANTIMONY	<sup>124</sup> Sb	60 · 3 d	β	0.06-2.32 ( $0.621$ , $2.32$ )	RL
	<sup>125</sup> Sb	2 · 7 y	Υ β Υ	0·044-2·30 ( <i>0·603, 1·69</i> ) 0·10-0·619 ( <i>0·125, 0·299, 0·619</i> ) 0·036-0·671 ( <i>0·408</i> )	⊦, RL
TELLURIUM	<sup>125</sup> mTe	58 d	Ŷ	0.035, 0.110	RL
	<sup>132</sup> Te	3∙25 d	β Υ	0·22 0·049-0·228	A
IODINE	<sup>129</sup>	1 ⋅ 7 x 10 <sup>7</sup> y	β	0.189	F, PL, PA
	131	8∙07 d	β Υ	0·040 0·257-0·806 ( <i>0·606</i> ) 0·080-0·723 ( <i>0·364</i> )	F, PA, RA, A
XENON	<sup>131</sup> mXe	11 · 8 d	Ŷ	0.164	PA, A
	<sup>133</sup> Xe	5 • 27 d	β Υ	0 • 163 0 • 267, 0 • 347 0 • 080, 0 • 382 ( <i>0 • 810</i> )	ΡΑ, Α
CAESIUM	<sup>134</sup> Cs	2·05 y	β	0.089, 0.410, 0.662	F, PL, RL
	<sup>136</sup> Cs	13 d	β	0.341, 0.560, 0.657 0.073, 1.24, (0.201, 0.010, 1.05)	F
·	. <sup>137</sup> Cs	30 · 2 y	Υ β Υ	0·067, 1·24 (0·347, 0·879, 1·05) 0·511, 1·18 0·662	F, PL, RL, PA, A
BARIUM	<sup>140</sup> Ba	12·8 d	β Υ	0 · 47 - 1 · 02 (0 · 47, 1 · 01) 0 · 139 - 0 · 537	F
LANTHANUM	<sup>140</sup> La	1 · 67 d	β Υ	1 ·25-2 ·17 ( <i>1 ·36, 2 ·1</i> 7) 0 ·110-2 ·55 (0 · <i>329</i> ) (0 ·487, 0 ·816, 1 ·60)	F, RL
CERIUM	<sup>141</sup> Ce	33 d	β	0 · 444, 0 · 582	F, RL, A
	<sup>144</sup> Ce	285 d	Υ β Υ	0·145 <i>0·175-0·309</i> 0·034-0·134	F, PL, PA, A
NEODYMIUM	<sup>147</sup> Nd	11 ·1 d	- β Υ	0·38, <i>0·82</i> 0·091-0·69 <i>(0·091, 0·53</i> )	F, A'
PROMETHIUM	<sup>147</sup> Pm	2·5 y	β	0.225	RL, A'

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Element	Radio- nuclide	Half-life (a)	Type of emission	Energies of emission (MeV) (b)	Main sources and discharge routes (c)
EUROPIUM	<sup>154</sup> Eu	16 y	β	0.27-1.86	
			γ	0·060-1·60 (0·123)	
	<sup>155</sup> Eu	1·81 y	β	0.10-0.25	RL
			γ	0.043-0.105	
POLONIUM	<sup>210</sup> Po	138·4 d	α	5.30	А
			Υ	0.803	
THORIUM	<sup>234</sup> Th	24 · 1 d	β	0 · 100. <i>0 · 191</i>	UPI
			Ŷ	0·030-0·094 ( <i>0·063, 0·93</i> )	
URANIUM	<sup>234</sup> []	2 · 47 x 10⁵ v	α	4.60 4.72 4.77	LIFA LIPA daughter of 238 Pu
	•	,	Ŷ	0.053.0.580 (0.053 0.121)	
	<sup>235</sup> U	7 ⋅ 1 x 10 <sup>8</sup> v	à	$4 \cdot 16 - 4 \cdot 60$ ( $4 \cdot 37$ $4 \cdot 40$ )	
			Ŷ	0.074-0.367 ( $0.185$ )	daughter of <sup>239</sup> Pu
	<sup>236</sup> U	2 · 39 x 10 <sup>7</sup> y	à	$4 \cdot 33, 4 \cdot 44, 4 \cdot 49$	daughter of <sup>240</sup> Pu
			Ŷ	0.050	
	<sup>238</sup> U	4 ⋅ 51 x 10 <sup>9</sup> y	ά	4 • 14, 4 • 15, 4 • 20	UEL UEA UPA
			Υ	0.048	daughter of <sup>242</sup> Pu
NEPTUNIUM	<sup>237</sup> Np	2 ⋅14 x 10 <sup>6</sup> y	α	4 • 40 - 4 • 87 (4 • 77, 4 • 79)	PL A
	,		Υ	0.020-0.240 (0.030, 0.086)	,
PLUTONIUM	<sup>' 238</sup> Pu	86 v	α	5-36.5-46 5-50	
		•	Ŷ	0.044	·, · E, · A, A
	<sup>· 239</sup> Pu	2 ⋅ 44 x 10 <sup>4</sup> y	ά	5.01-5.16	Ε ΡΙ ΡΑ Δ
		,	Ŷ	0.039-0.769	.,,
	<sup>240</sup> Pu	6580 y	ά	5.02, 5.12, 5.17	F. PL. PA. A
			γ	0.045, 0.104, 0.160	
	<sup>241</sup> Pu	13·2 y	α	4·80-5·05 ( <i>4</i> ·90)	F. PL. PA. A
			β	0.021	
	<sup>242</sup> Pu	3 · 79 x 10⁵ y	ά	4.86,4.90	PL, PA, A
AMERICIUM	<sup>241</sup> Am	458 y	α	5·39-5·55 ( <i>5·48</i> )	F, PL, PA, A
		-	Υ	0.026-0.060	daughter of <sup>241</sup> Pu
CURIUM	<sup>242</sup> Cm	163 d	α	5 · 97 - <i>6 · 11</i>	PL, PA, A
			γ	0.044	

Footnotes to Table 1. a. d=days y=years. Only radionuclides with half-lives>1day are included. b. Dominant energies of emission are in italics.

С.

F=fallout from nuclear weapon tests.

F=fallout from nuclear weapon tests. PN=low radioactive liquid discharge from a reprocessing plant (Windscale). RL=low radioactive liquid discharge from a reactor. UEL=low radioactive liquid discharge from a uranium enrichment plant (Capenhurst). UPL=low radioactive liquid discharge from a uranium processing plant (Springfields). PA=emissions to the atmosphere from a reprocessing plant. RA=emissions to the atmosphere from a reactor. UPA=emissions to the atmosphere from a uranium processing plant (Springfields), radionuclides not specified. UFA=emissions to the atmosphere from a uranium enrichment plant (Capenhurst).

A=of particular importance if accidents occur, many of the other nuclides listed may also be released. A=non-volatile fission product, likely to be released only if the fuel is disrupted by explosion or fire.

EC=Electron capture.

IT=Isomeric transition.

Main sources of this information are as follows:

Beattie and Bryant (1970), Booker (1962), British Nuclear Fuels Ltd. (1978), Cambray et al. (1977). Dunster et al. (1958), Groom and Short (1977). Hetherington et al (1976), Hunt et al (1976), Parker (1978), Royal Commission on Environmental Pollution (1976), Russell (1966), Taylor et al (1976), Weast and Astle (1980).

## INPUTS OF RADIONUCLIDES TO TERRESTRIAL ECOSYSTEMS

#### General aspects

Radionuclides currently in terrestrial ecosystems have originated from natural sources with or without the aid of man, in fallout from explosion of nuclear devices, and in discharges to the atmosphere, liquid discharges and solid waste from the nuclear and the non-nuclear industries (Table 1). Natural sources and fallout from nuclear devices provide the main inputs to terrestrial ecosystems, except for certain radionuclides in some situations near nuclear installations. For example, <sup>239, 240</sup>Pu and <sup>241</sup>Am, originating in routine discharges from Windscale, occur at higher than usual levels in west Cumbrian estuarine sediments (James *et al.*, 1978), and <sup>131</sup>I occurred at high levels in south-west Cumbria after accidental release from Windscale in 1957 (Dunster *et al.*, 1958).

Fallout from nuclear devices and discharges from nuclear installations often contain similar radionuclides but the proportion of the latter vary greatly with source e.g. fallout, fuel enrichment plant, fuel fabrication factory, power station and reprocessing plant, (UNSCEAR, 1977). Different types of installations performing the same function, e.g. pressurized water, boiling water, Magnox and gas-cooled reactors, may produce wastes containing different proportions of the same radionuclides (UNSCEAR, 1977). Even at one installation, e.g. the Windscale reprocessing plant, the composition of the waste may change with time or with the process and equipment involved (UNSCEAR, 1977). The composition of fallout from nuclear devices changes with time, type of device and explosion conditions (UNSCEAR, 1977).

Identification of the main source of particular nuclides in individual situations is often possible by examining the geographical distribution and ratios of selected nuclides. These ratios are characteristic of individual sources at certain times.

#### Input pathways

Radionuclides enter terrestrial ecosystems in the UK along several pathways. Each pathway tends to be associated with several sources but one source usually dominates. Routes include the following:

- (a) Deposition in precipitation of particles and dissolved gases or vapours.
- (b) Dry deposition of particles of resuspended dust or fine droplets (aerosols) from soil, vegetation and water surfaces.
- (c) Deposition of mineral matter, seaweed and other flotsam on the sea-shore and around freshwater courses and lakes.
- (d) Transfer in birds, other vertebrates or invertebrates which feed in the sea or on the shore, but which occasionally move inland.
- (e) Transfer from the sea in fish or shell-fish harvested by man (Primary routes to man, including this one, are the concern of other organizations, and are not considered in this report).
- (f) Removal of material from the shore by man and deposition inland, e.g. seaweed as fertilizer, and sand.
- (g) Use of fertilizers, especially phosphate (UNSCEAR, 1977).
- (h) Burial of radioactive waste.

Precipitation scavenging of air has long been assumed to be responsible for about 80-90 per cent of the delayed deposition of fallout (Osburn, 1967) and presumably also for radionuclides emitted to the atmosphere from nuclear installations. About 85 per cent is an appropriate value for the UK (N. J. Pattenden, personal communication). Deposition of fallout in the UK tends to be linearly related to precipitation (Peirson and Salmon, 1959).

Amounts of precipitation are recorded in the UK by the Meteorological Office, by many national or local organisations and by very many amateur meteorologists. Data from all sources for Cumbria, which are of particular interest in relation to Windscale discharges,

Table 2. Input of radionuclides in UK precipitation and air—some sample data

a) Mean deposi	tion in the U	K in 1975, m	Ci km <sup>−2</sup> (Hunt	<i>et al.,</i> 1976)					
	<sup>54</sup> Mn	<sup>90</sup> Sr	<sup>95</sup> Zr	<sup>103</sup> Ru	<sup>106</sup> Ru	<sup>125</sup> Sb	<sup>137</sup> Cs	<sup>141</sup> Co	144Co
	0.044	0.48	0.24	0.50	0.97	0.071	0.66	0.14	3.8
b) Monthly vari	iation of activ	vity concentra	tion in air nea	r ground level.	fCi kg <sup>-1</sup> (C	ambray <i>et al.,</i> 1	977).		
	⁵⁴Mn	<sup>90</sup> Sr	<sup>95</sup> Zr	<sup>103</sup> Ru	<sup>106</sup> Ru	<sup>125</sup> Sb	<sup>137</sup> Cs	<sup>141</sup> Co	<sup>144</sup> Co
Milford Haven	<0.1		<0.1—	' N. D.—	0 · 9—	< 0.1	0 · 3	N.D.—	1.0
1975	1 · 4		7.9	2.0	17.0	1.2	2.2	2.0	14.0
Eskdalemuir	<0.1—		<0.1—	N.D.—	0 · 2	<0.1—	0 · 1—	N.D	0.3
1975	0.3		3 · 9	1.5	3.6	0.6	1.1	1.1	5.6
Milford Haven	<0.1—	-	N.D.—	N.D	0.3—	<0.1	0.1	N.D.—	0.3
1976	0.5		4 · 6	10.0	3.1	0.7	0.7	9 · 4	4.0
Eskdalemuir	<0.1—	_	N.D.—	N.D.—	0 · 3	<0.1-	0.1—	N.D.—	0 · 2
1976	0.1		2.5	2.3	1.6	0.6	0.4	3 · 4	1.7

N.D. Not detected

Not measured.

are summarized as monthly and yearly totals by Mr. P. R. Cutforth.

Concentrations of radionuclides in UK precipitation and air are available in yearly summaries from AERE and NRPB. Data for some recent years (Table 2) indicate the levels of selected radionuclides present in these sources. Variation in levels between different periods and between different sites is high and depends on location of source, location of sampling site, global and local weather patterns and characteristics of individual radionuclides.

In certain areas of the world, not necessarily dry and hot areas, dry deposition of distant origin may account for as much as 28 per cent of radionuclide input to terrestrial ecosystems (Osburn, 1967). In west Cumbria, resuspension of dried estuarine silt has been suggested as a possible input pathway to more inland ecosystems (Royal Commission on Environmental Pollution, 1976). Mean concentrations of <sup>95</sup>Zr + <sup>95</sup>Nb, <sup>106</sup>Ru, <sup>137</sup>Cs and 144Ce in the silt in some Cumbrian estuaries near Windscale in 1972-73 were respectively 470-960, 660-940, 68-110 and 360-560 pCi g<sup>-1</sup> dry weight (Mitchell, 1975). <sup>239, 240</sup>Pu and <sup>241</sup>Am are each present at about 50-3000 pCi g<sup>-1</sup> (James et al., 1978), but for these, and probably for other radionuclides which are adsorbed by minerals, articles  $<1 \,\mu$ m diameter, which are most likely to be resuspended, have a specific activity about twice that of the whole silt.

Tests by the MAFF Fisheries Radiobiological Laboratory and others to date have failed to obtain evidence of this resuspension transfer (Parker, 1978). Evidence may be obtained from the current long-term monitoring by NRPB and others of concentrations of radionuclides in the air near Windscale and measurement by AERE and MAFF of accumulation of radionuclides in soil.

Precipitation in north-west England, especially that falling within 10 km of the coast, includes components derived from the sea (Gorham, 1958; Allen et al., 1968). The importance of this pathway in relation to the Windscale effluent discharged to the sea is unknown but is being examined currently by AERE.

I provides an example of the transfer of a radionuclide along the sea-to-land pathway. 75 per cent of the I throughput, mainly from irradiated fuel, is discharged to the sea from Windscale (Bryant and Jones, 1972). Atmospheric water is exchanged with water elsewhere in the environment in about ten days (UNSCEAR, 1977) 1-containing aerosols originate largely from the sea (Martens and Harriss, 1970; Robbins, 1970) and marine aerosols contain up to ten times the I/CI ratio of seawater (Duce et al., 1967). This enrichment is considered to be controlled by sea-surface temperature, ultraviolet light and bubbling rate (Martens and Harriss, 1970), gaseous I from the sea being adsorbed by freshlyproduced aerosols near the sea surface (Robbins, 1970). It is not thought to be associated with the sea-surface microlayer which is known to contain much higher concentrations of various pollutants than the deeper layers (Duce et al., 1972). However, it is possible that other less volatile radionuclides than I are concentrated in the microlayer and transferred to aerosols and hence to land.

Seaweeds accumulate many elements which occur as radionuclides in seawater. I, for example, is present at about 0.05 ppm in seawater but is concentrated by seaweed to about 50-9000 ppm (Shacklette and Cuthbert, 1967). Recent concentrations of radionuclides in seaweeds from the Windscale area are given in Table 3. The amount of seaweed deposited on the shore in the UK each year is currently unknown but, on one Californian beach, 105 kg km-1 containing  $4 imes10^4$  pCi <sup>239, 240</sup>Pu were deposited annually (Cheng et al., 1977). 0.2 per cent of the Pu was transferred to flies breeding in the weed and hence possibly to other ecosystems. Invertebrates are common amongst deposited seaweed in the UK but their role in transferring radionuclides has not been investigated. Seaweed from Cumbria contains about 10 times more Pu than that in the Californian study (Table 3).

Fable 3. Radionuclide content	of two	seaweeds from	the vicinity	of Windscale.
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			(porg dry weight )					
	<sup>90</sup> Sr	<sup>95</sup> Zr+ <sup>95</sup> Nb	<sup>106</sup> Ru	<sup>134</sup> Cs	<sup>137</sup> Cs	<sup>144</sup> Co	<sup>239, 240</sup> Pu	<sup>241</sup> Am
Porphyra								
1972/730.	4	100-800	1200-3800	NP	11-22	26-230	12-27	2-33
1974°	6	180-270	2810-3400	NP	140-150	150-340	18-44	52-140
1976₫	NP	130	1600-2600	20-25	120-130	74-84	29	26
Fucus								
1972/73	NP	240-720	180-510	NP	70-200	68-100	NP	NP
1974°	NP	57-190	140-370	33-109	140-500	38-100	NP	NP

Original data converted from a wet weight to a dry weight basis assuming a moisture content of 90% for wet seaweed. а.

From Mitchell (1975). b.

c. From Hetherington (1976).

From Mitchell (1977). d.

NP No published data. (nCi a<sup>-1</sup> dry woight<sup>a</sup>)

### RETENTION OF RADIONUCLIDES BY THE ABOVE-GROUND PARTS OF PLANTS

Radionuclides which are deposited on plants occur in particles, e.g. Pu, as vapours, e.g. elemental I, or as solutions in rain or fog, e.g. Cs (Russell, 1966b). Each nuclide may be deposited in more than one chemical or physical form. Retention may involve physical retention of particles, adsorption on the plant surface or absorption into plant cells.

The extent to which radionuclides deposited on plants are retained depends on the physical characteristics of the deposit, the growth form of the plants and the characteristics of the plant surface (Russell, 1966*b*). The mean size of particles deposited tends to decrease with distance downwind from source.

Particles > c.200  $\mu$ m diameter are seldom retained on leaves (Romney et al., 1963). Wind removes virtually all particles > 44  $\mu$ m, smaller particles being retained amongst hairs, in crevices and on sticky glandular secretions on plant surfaces. Retention varies with plant species. Plants which grow in tussocks or as tufts form particularly effective traps for particles (Russell, 1966b). In a comparison of retention of 1-44  $\mu$ m particles by sorghum, a grass-like plant with smooth angular stems and leaves, and soybean, a rougher bushy plant, both species retained about 39 per cent of the initial deposition (Witherspoon and Taylor, 1970, 1971); however, 81 per cent of this 39 per cent was retained by the soybean foliage but only 24 per cent by sorghum foliage, the remainder being retained by the stems. Half-lives for retention of the particles by the foliage over 34 days were 24.4 and 17.9 days for respectively soybean and sorghum.

Deposition of vapours and very small particles with a, very low terminal velocity is determined by diffusion processes (Russell, 1966*b*). The deposition is inversely related to the thickness of boundary layers of still air on the plant surface and greatest deposition occurs at places such as the edge of leaves, where the boundary layer is disrupted by turbulence. Loss of water-soluble radionuclides from plants appears to vary little with radionuclide under a given set of conditions. The fraction of the initial deposit of six soluble radionuclides which was lost from cabbage plants in 28 days ranged from 83-90 per cent (Russell, 1966*b*).

Chamberlain (1970) drew together the available data on interception and retention of radioactive aerosols and fine particles by vegetation in various agricultural and non-agricultural habitats. The proportion of the fallout retained initially by the herbage, p, was assumed to be related to an uptake coefficient,  $\mu$  (m<sup>2</sup> kg<sup>-1</sup>), by the relation  $1 - p = exp - \mu w$ , where w is the herbage density in kg m<sup>-2</sup> dry matter. Experimental values of  $\mu$  based on data from several sources fell in the range 2.3-3.3 m<sup>2</sup> kg<sup>-1</sup>. The physical form of the radioactive contamination had no marked influence on the value. The field loss coefficient, i.e. the retention mean life,  $1/T_b$  or  $\lambda b$ , was of the order of 0.05 days<sup>-1</sup> for field experimental data. This value is equivalent to a retention half-life, 0.693  $\lambda b^{-1}$ , of 20 days. The actual variability in retention half-life was 12.8-86.7 days. Some decreases in retention during dry periods were noted.

Representative values of p and  $\lambda_b$  were used to calculate the normalized specific activity (N.S.A.) where

N.S.A. = 
$$\frac{\text{Activity per kg dry matter foliage}}{\text{Activity deposited day}^{-1} \text{ m}^{-2} \text{ of ground}}$$
 (1)

N.S.A. values calculated from experimental results for  $\mu$  and  $\lambda_b$  ranged from 27-55 m<sup>2</sup> days kg<sup>-1</sup>, depending on the value assumed for  $\lambda_b$  and the time between successive croppings of herbage. N.S.A. values deduced from survey data for herbage in the growing season using (1) were similar to the calculated values. Values were higher, generally in the range 100-1000 m<sup>2</sup> days kg<sup>-1</sup>, for herbage in winter and for natural vegetation in upland and sub-arctic regions where growth is slow. Highest values were obtained for lichens in Finland and Alaska.

## RETENTION AND MIGRATION OF RADIONUCLIDES IN SOIL

#### General aspects

The soils in north-west England vary greatly in type, composition, chemical characteristics and biology (Pearsall and Pennington, 1973). In relatively undisturbed areas they often have a permanent superficial layer of slowly-decomposing plant remains. They are strongly leached by the moderate to high rainfall and hence are base-poor and acid, with a pH of 4-5 or less. The mineral composition of the  $< 2 \,\mu$ m soil particles, as indicated by the average composition of lake sediments in the area, is 42 per cent illite and 58 per cent chlorite (Dr. S. R. Aston, Lancaster University, personal communication).

Retention and migration of pollutant radionuclides in soil have been much studied because most of these radionuclides accumulate in the upper soil. This leads to direct exposure of man to radiation from the soil surface, uptake of radioactivity by food plants which root mainly in the top 10-30 cm of soil, possible changes in the level of radionuclides in water during its passage over or through the soil, and movement during soil erosion of fine particles which are much more radioactive than the whole soil.

Factors which affect the degree of retention of radionuclides by different soils include characteristics of the nuclide involved (Graham and Killion, 1962; Essington and Nishita, 1966), concentration of the nuclide (Tamura, 1964), concentration of ions which compete with radionuclide ions for retention sites (Tamura, 1964; Essington and Nishita, 1966), time available for sorption (Tamura, 1964), soil pH (Rhodes, 1957; Essington and Nishita, 1966), soil organic matter content (Graham and Killion, 1962; Essington and Nishita, 1966), soil mineral type (Graham and Killion, 1962; Tamura, 1964), concentration of chelating agents or other chemicals which could form complexes with the radionuclide (Essington and Nishita, 1966; Nishita and Essington, 1967).

Radionuclides are retained by soils in the films of water and hydrated oxide or humus on particle surfaces, and by incorporation in the surface layers of the crystal lattice of soil minerals. They may occur in solutions, as cations or anions retained on mineral or organic particles, in colloidal form, as complexes with minerals and particularly with organic matter, and as particulate material. Examples of these various types of retention are given below.

Information on retention and migration of radionuclides in soil has been obtained by measuring sorption of radionuclides under various conditions, by extracting radionuclides from soil in various ways, by adding radionuclides to soil columns and observing migration, and by measuring the depth distribution of components of fallout in the field soil, Great caution is required in using this information. Laboratory studies may allow insufficient time for radionuclides to equilibrate fully with soil, whereas changes in availability of nuclides to extractants or plants can occur in field soil over many months or years. As in stable nuclide chemistry, extraction procedures often rely on one leaching with a strong extractant, e.g. M ammonium acetate. Successive leachings often increase total extraction markedly (Kahn, 1956) and are normal in the field, but, in the latter, the extractant is extremely dilute and weak. Unless amounts of radionuclides at various depths in a field soil profile are related to cumulative fallout (Peirson and Salmon, 1959), a lack of knowledge of amounts which have moved through the profile before examination may lead the investigator to false conclusions.

#### Strontium (Sr)

A few per cent of the Sr in soil is held in a form largely inaccessible to plants but the majority is water-soluble or retained on minerals and organic matter in an ion-exchangeable form (Nishita *et al.*, 1956*a*; Russell and Newbould, 1966). In laboratory tests with various soil components, the highest retentions were by peat and montmorillonite (Graham and Killion, 1962). However, application of normal agricultural levels of manure had no significant effect on the concentration of fallout Sr accumulated by Danish soils (Aarkrog and Lippert, 1977). Presumably, as in the case of organic matter and plant uptake of <sup>90</sup>Sr, applications have to be abnormally high before a significant effect occurs (Nishita *et al.*, 1961).

Sr sorption is most favoured by a pH of 7-11 and may be reduced to less than 50 per cent of added nuclide at pH 3-5 (Rhodes, 1957; Essington and Nishita, 1966). Soil suspensions retain more Sr in solutions of low ionic strength than in solutions of high ionic strength.

The effect of competing ions may be summarized as  $AI > Fe > Ba \ge Ca > Mg \ge H^*$ ,  $Cs > K > Na > NH_4$ . Essington and Nishita (1966) indicated that the sorption of Sr in soil is reduced by addition of Ca, for example as lime and fertilizers. In widely different UK soils, the median depth of added Sr six years after application, 3-5 cm, was increased four-fold by annual additions of ammonium nitrate or mixed fertilizer in lime-poor soils, but was unaffected in calcareous soil (Squire, 1966).

Synthetic chelation agents, the effect of which may simulate naturally-occurring chelation agents, affect retention and migration of Sr significantly only in soils rich in organic matter or sand or low in clay (Essington and Nishita, 1966; Nishita and Essington, 1967).

#### Caesium (Cs)

At the low levels generally found in the environment, radiocaesium is retained very strongly by illite and chlorite (see above) but less strongly by other clay

\*Relative effects of H and Cs uncertain.

minerals such as kaolinite and montmorillonite (Graham and Killion, 1962; Tamura, 1964). Retention occurs either at exchange sites at the edges of the crystal lattice interlayers or in the interlayers (Fredriksson *et al.*, 1966). Soils rich in sand retain far less Cs than other soils (Fredriksson *et al.*, 1966).

Organic matter appears to be far less important than mineral particles in retention of Cs. Application of manure had no significant effect on the levels of fallout Cs accumulated in Danish soils (Aarkrog and Lippert, 1977). In contrast, peats and plant remains on the surface of forest soils trap fallout before it reaches the mineral soil layers and can retain large amounts of Cs (Graham and Killion, 1962; Waller and Olson, 1965). Cs held in organic materials is readily available to plants (Fredriksson *et al.*, 1966), presumably because microbial immobilization (Witkamp *et al.*, 1967) and ion exchange are the main methods of retention.

Retention on minerals varies little with pH over the pH range 3.5-10.0, but decreases or increases below this range depending on soil type (Nishita *et al.*, 1956*a*; Rhodes, 1957).

Retention is reduced by more than 90 per cent in the presence of 4M NaCl (Rhodes, 1957) and hence would be expected to be reduced slightly in seawashed coastal soils. Annual additions of fertilizer over several years had no effect on the distribution of Cs with depth in four UK soils, an acid clay, a sand, a lower Greensand soil and a calcareous loam (Squire and Middleton, 1966). Median depths of the distributions were 1-3 cm after six years.

Synthetic chelation agents had little effect on retention and migration of Cs in different soils (Nishita and Essington, 1967).

Migration of Cs down the soil profile is usually very slow. 51-75 per cent of added Cs was retained in the top 5 cm of various UK soils, including a peat, after 4.8 years (Gale *et al.*, 1963). Booker (1962) found that 79 per cent of accumulated <sup>137</sup>Cs from bomb fallout and Windscale occurred in the top 5 cm of various north Cumbrian soils in 1961.

#### Ruthenium (Ru)

The chemical behaviour of Ru in soils is complicated by the many valence states which occur and the ease with which the element changes valence state and forms complexes (Essington and Nishita, 1966). Cationic Ru is readily sorbed by most soils by exchange or precipitation except under very acid or mildly alkaline conditions (Auerbach and Olson, 1963). Rhodes (1957), for example, found maximum sorption at about pH 3-5.5 and 14 in a calcareous sandy loam sub-soil. In contrast, in other studies, Ru was sorbed readily from neutral or highly alkaline solutions or under acid or mildly alkaline conditions (Essington and Nishita, 1966). Neutral or anionic Ru-complexes are not retained significantly by mineral soil but are held by acid organic matter, for example the litter layer in woodlands (Auerbach and Olson, 1963; Essington and Nishita, 1966; Ritchie *et al.*, 1970). These complexes may comprise the main part of the 20-40 per cent soil Ru which is water-extractable (Nishita *et al.*, 1956*a*; Nishita and Essington, 1967).

Low cation concentration and reducing conditions favour retention of Ru by soil. Synthetic chelating agents have little effect on movement of the element (Essington and Nishita, 1966; Nishita and Essington, 1967).

As a result of the above characteristics, on undisturbed sites, more than 50 per cent of the accumulated fallout Ru occurs commonly below the top 5 cm of soil and the element may be readily detectable at 30 cm depth (Franklin *et al.*, 1967; Ritchie *et al.*, 1970).

### Zirconium (Zr) and niobium (Nb)

Little information is available on the retention and migration in soil of <sup>95</sup>Zr and its daughter radionuclide <sup>95</sup>Nb (Held, 1963). In estuarine sediments, <sup>95</sup>Zr + <sup>95</sup>Nb are held strongly on the surfaces of particles, particularly in the silt and clay fractions (Hetherington and Jefferies, 1974). Organic matter appears to play little part in retention of these nuclides.

More than 70 per cent of <sup>95</sup>Zr in solution added to three different soils, a chernozem, a podzol and a krasnozem, was absorbed (Klechkovsky and Gulyakin, 1958a). The <sup>95</sup>Zr was not displaced from the chernozem or krasnozem by neutral salt solutions, but 3-6 per cent was displaced from the podzol.

In equilibration sorption studies using a sandy loam, greatest amounts of  ${}^{95}$ Zr +  ${}^{95}$ Nb were retained at pHs 1-5 and above pH 8 (Rhodes, 1957).

### The rare earth elements, cerium (Ce), yttrium (Y) and promethium (Pm) -

Cerium (Ce) and other rare earth elements are held very firmly by soil minerals. Mineral type may affect adsorption considerably (Nishita *et al.*, 1956*a*). Maximum adsorption occurs between pHs 6-8 and above pH 11. Apparent decreases in sorption between pHs 8-11 are believed to be due to peptization\* of colloidal particles bearing rare earths. Below pH 6, sorption decreases rapidly (Essington and Nishita, 1966). In a laboratory study, water extracted less than 5 per cent of the Ce added to various soils except in a sandy loam of pH 4.7 where the value was 9 per cent (Nishita *et al.*, 1956*a*). Exchangeable Ce, extractable with M ammonium acetate, varied from about one per cent for a sandy loam of pH 6.6 to about 90 per cent for a muck soil of pH 3.9 and rich in organic matter.

\*Conversion of colloids from an insoluble (gel) form to a soluble (sol) form.

Rare earths are sorbed and complexed by humus (Essington and Nishita, 1966). Some synthetic chelating agents and mobile complexing agents, such as citrate and acetate, inhibit adsorption and markedly desorb Ce and other rare earths from soils (Essington and Nishita, 1966; Nishita and Essington, 1967).

More than 60 per cent of fallout Ce present in undisturbed acid, well-drained forest and non-forest soils in Ohio occurred in the top 7.5 cm (Franklin *et al.*, 1967). Fallout Ce tended to be fairly evenly spread throughout the L, F, H and 3-8 cm thick layers to 11.4 cm in undisturbed sandy loam soils in North Carolina-Tennessee (Ritchie *et al.*, 1970). About 70 per cent of the Ce added to a sandy loam soil in West Germany was present in the top 5 cm ten months after application (Kuhn *et al.*, 1973).

## The transuranides, neptunium (Np), plutonium (Pu), americium (Am) and curium (Cm)

The chemistry of the transuranic radionuclides in soil is extremely complex and only partially understood (Wildung and Drucker, 1976). Many fewer data are available for Am, Cm, and Np than for Pu. Four different oxidation states of Pu (III-VI) can coexist in the environment and the state is known to influence behaviour (Prout, 1959; Dahlman *et al.*, 1976). Pu compounds form minute "pseudocolloids" above pH 2.8 and genuine aggregates above pH 7.5 (Bulman, 1976). Pu can form complexes with soil components (Bulman, 1976; Wildung and Drucker, 1976).

PuO<sub>2</sub> in fallout and emissions from nuclear installations enters soil as stable and water-insoluble microspheres. Solubilization may occur by the action of organic acids of plant or microbial origin (Wildung and Drucker, 1976). High specific activity <sup>238</sup>PuO<sub>2</sub> dissolves about 100 times faster than the lower activity <sup>239</sup>,<sup>240</sup>PuO<sub>2</sub> (Bulman, 1976). This fact may be important ecologically as modern nuclear fuel technology involves discharge of waste with a higher <sup>238</sup>Pu/<sup>239,240</sup>Pu ratio than that in fallout.

Transuranides are strongly adsorbed by the finer soil particles and colloids, sorption changing with oxidation state (Prout, 1959; Dahlman *et al.*, 1976). Pu is adsorbed more strongly than Am, Cm or Np (Dahlman *et al.*, 1976).

Adsorption is strongly associated with the hydrous oxide film on soil particles, particularly with Mn oxides rather than with organic matter (Means *et al.*, 1978; Muller, 1978). In Ohio silt loams, 73-88 per cent of the Pu was associated with the oxide film, 2-13 per cent with organic matter extractable with 0.1N NaOH, 2-19 per cent with the crystal structure of soil particles, and one per cent<sup>239,240</sup>Pu or 3 per cent <sup>238</sup>Pu as adsorbed ions readily removable by extraction with MgCl<sup>2</sup> (Muller, 1978).

Adsorption is affected by pH and is particularly high around neutrality. It decreases markedly about pH 9-12 and below pHs 2-4, depending on the test conditions used (Rhodes, 1957; Prout, 1959). In laboratory studies, percentage extraction of Pu varies considerably with extractant pH, soil organic matter, equilibrium time, soil/extractant ratio and wet or dry pre-treatment of soil (Nishita *et al.*, 1977). Synthetic chelation agents promote desorption more than organic acids such as citric.

Pollutant transuranides are retained in the top few cm of soil in the field, even in high rainfall areas (Wrenn, 1974) (Table 4).

Table 4. Distribution of fallout radionuclides with soil depth at Brookhaven, Suffolk County, New York, autumn 1970 (Hardy & Krey, 1971).

· · · · · · · · · · · · · · · · · · ·		ب من المرب المرب الم	
	<sup>137</sup> Cs	alstribution <sup>90</sup> Sr	<sup>239,240</sup> Pu
0-7	59	42	57
7-11	26	30	27
11-15	11	15	11
15-21	4	8	3
21-25	0	2	1
25-30	0	3	1

Factors which affect the migration of transuranides down soil profiles include precipitation, which influences leaching and down-wash of transuranide particles or clay particles to which transuranides have become attached, oscillation of soil properties such as temperature and moisture content, soil composition and hence porosity and aeration, degree of fissuring of soils, soil animal and microbial activity, soil disturbance by man's activities, addition of lime and fertilizer, concentration of naturally-occurring substances which could chelate or solubilize transuranic compounds, and soil type, in particular the degree of podzolization (Held *et al.*, 1965; Romney *et al.*, 1970; Wrenn, 1974; Beckert and Au, 1976; Bulman, 1976; Essington *et al.*, 1976; Jakubick, 1976a, 1976b).

Physical transport of transuranide particles or soil particles carrying transuranides is considered to be the main method of migration downwards in undisturbed soil profiles (Romney *et al.*, 1970; Francis, 1973; Jakubick, 1976*a*, 1976*b*) although physicochemical processes of the type involved in podzolization appear important in some profiles (Essington *et al.*, 1976). The possible importance of processes other than simple leaching and downwash is emphasised by the more rapid migration of <sup>241</sup>Am than <sup>239,240</sup>Pu in some low-rainfall desert areas (Essington *et al.*, 1976).

#### Iodine (I)

lodide is considered to be the main form of I in soil, but iodate occurs in nitrate fertilizer (Goldschmidt, 1958).

Hill and Grimwood (1978), in a review, concluded that I migrates through the rocks and the deeper mineral-rich soil horizons as fast as ground-water, and hence adsorption, ion exchange, chemical precipitation and colloidal filtration have negligible effects on retention; however, in near-surface soil horizons retention is often appreciable. In various soils from Rumania, Russia and Japan, for example, only 3.0-7.3 per cent of stable I was water-soluble (Chilean Iodine Educational Bureau, 1956).

Soils rich in organic matter contain more stable I, <sup>127</sup>I, than soils poor in organic matter (Hanson, 1963). In a study of the behaviour of <sup>131</sup>I in bentonite, kaolinite and three Californian soils, including a peat, pre-treatment by autoclaving, oxidation with peroxide and digestion with alcohol, as well as extraction of <sup>131</sup>I by salt solutions, all indicated that most of the I was retained by dead organic matter (Raja and Babcock, 1961). The peat soil retained 85.7 per cent of added <sup>131</sup>I when extracted with water, the clay minerals retaining only 0.2-4.3 per cent. A clay loam retained 90.4 per cent <sup>131</sup>I, but only 29.2 per cent after peroxide treatment. Retention by from Cumbria (Table 5), I content was highly correlated with oxalate-soluble Al ( $r = 0.834^{***}$ ), but not with oxalate-soluble Fe (Whitehead, 1978). However, for the five most acidic soils (pH<4.8), I was more closely correlated with Fe than with Al. Although there was an apparent association between I and organic matter in peat generally, the Cumbrian peat had high values for oxalate-soluble Al and a higher ratio I/oxalate-soluble AI than for other profiles suggesting that sesquioxides may have been largely responsible for I retention. In other tests, undried UK soils continued to adsorb I added as iodide for >48 h. Maximum sorption occurred at pH<5 and was associated with Fe and AI oxides (Whitehead, 1973a, 1974). A secondary peak of sorption occurred at pHs 8.5-9.0 and was associated with organic matter. Sorption of iodide by oxides was negligible above pH 7.0.

Table 5 indicates the variation in distribution of stable I with depth in Cumbrian soils. The behaviour of stable I is expected to resemble that of <sup>129</sup>I or <sup>131</sup>I. Kuhn *et al.* (1973) found most of the <sup>131</sup>I added to an undisturbed sandy loam soil in West Germany occurred either in the top 10 cm or at 30-40 cm 26 days after application.

Table	5.	Distribution of stable I wi	h soil	depth	in some Cumbrian soils	s
		(White	head,	1978).		

Depth (cm)	Andesite/rhyolite (igneous)	% distribution Ordovician slate (sedimentary/drift)	Peat over alluvial clay
0-10	10.1	6.1	19.4
10-20	10.3	7.7	16.2
20-30	15.1	9.6	21.3
30-40	21.0	11.4	16.4
40-50	17.1	11.5	11.5
<sup>-</sup> 50-60	14.3	11.8	9 · 1
60-70	12.1	11.7	4 · 8
70-80		12.0	1.3
80-90	_	11.9	_
90-100		6 · 3	-

soil organisms was negligible. Retention by the clay loam and a loam increased gradually over at least seven days and was high over the pH range 5.5-8.5 but decreased by 10-25 per cent at pH 4.5.

Pennington and Lishman's (1971) data for Cumbria suggest that organic matter and clay colloids are important in the retention of I by lake sediments derived from surface soils.

In studies of <sup>127</sup>I in 19 UK soil profiles, including three

#### UPTAKE OF RADIONUCLIDES BY PLANTS

#### General aspects

Radionuclides may be absorbed by plants either through the leaves, stems, flowers and fruits or via the roots (Russell, 1966b). Distinction of these two pathways for individual nuclides is important for several reasons. Many nuclides are relatively immobile in plants and the levels of nuclides in different tissues will depend considerably on the site of absorption. The extent of foliar or floral absorption will depend upon contamination during a short period, whereas absorption via the roots and plant-base may depend on accumulation of radionuclides on plants or in the soil over a long period. Once radionuclides enter the soil, they become generally less available to plants because of dilution with ions already in the soil solution and sometimes because of fixation in complexes and soil minerals.

Absorption of radionuclides via the above-ground parts of plants appears to have been little studied in comparison with absorption via the roots, although many authors give plant retention data without discriminating between absorbed and superficially retained radionuclides. This omission reflects perhaps the technical difficulties of separating the two fractions.

In rice plants growing in the field in northern Italy, about 72 per cent of the <sup>144</sup>Ce, 13-15 per cent of the <sup>90</sup>Sr, <sup>106</sup>Ru and <sup>137</sup>Cs and 4 per cent of the <sup>54</sup>Mn in or on the shoots appeared to be derived from direct deposition (Verfaillie et al., 1967). Protection of the plants from fallout had little or no effect (<14 percent) on radionuclide levels in the roots. 55-76 per cent of the five nuclides in meadow plants in fields adjacent soil on a contaminated site, by growing plants in

influenced by many factors including concentration and chemical characteristics of the nuclide, soil pH, competition between ions in the absorption process, rate of movement of the nuclide out of the rooting zone, distribution of roots in the soil profile, soil colloid concentration, organic matter content, the complex of factors collectively referred to as soil fertility and management of an area including ploughing, fertilizing and lime application (Russell, 1966c). Examples of the effects of these factors are given below.

Absorption rate may vary considerably between plant species and with radionuclide (Nishita et al., 1961). The order of uptake is  ${}^{89}Sr \simeq {}^{90}Sr \gg {}^{131}|>{}^{140}Ba>$  $^{137}Cs \simeq {}^{106}Ru > {}^{144}Ce \simeq {}^{91}Y \simeq {}^{147}Pm \simeq {}^{95}Zr + {}^{95}Nb$ >239Pu (Nishita et al., 1961). Russell (1966c) agrees with this order, but suggests <sup>137</sup>Cs><sup>106</sup>Ru and <sup>144</sup>Ce> <sup>95</sup>Zr. Radionuclides which are absorbed most readily are those which are soluble in the soil water or are isotopes of elements which have some metabolic function in the plant (Menzel, 1965). Those that are least absorbed are virtually insoluble in soil water.

Absorption of elements by plants is often indicated in terms of a concentration factor or ratio. This ratio is expressed usually in element concentrations, plant/soil, on a dry weight basis, but occasionally on a fresh weight of plant and dry weight of soil basis. Concentrations are expressed in  $\mu$ g or Ci per unit weight (Table 6).

Radionuclide uptake by plants has been studied by measuring concentrations of radioactivity in plants and

Table 6.	Concentration factors or ratios* for elements of particular interest in this publication
	(Menzel, 1965 ; Bulman, 1976)

10-1000 Strongly concentrated	1-100 Slighly concentrated	0·1-10 Not concentrated	0 · 01 - 1 Slightly excluded	<0·01 Strongly excluded
. S	Sr H** C**	I	Cs Ru∙	Y Zr Ce Pm Pu (10 <sup>-5</sup> —10 <sup>-3</sup> ) Am (10 <sup>-1</sup> —10 <sup>-2</sup> ) Cm (10 <sup>-4</sup> —10 <sup>-2</sup> )

Amount of element in plant/amount in soil. Amounts in  $\mu$ g g<sup>-1</sup> or Ci unit weight<sup>-1</sup> plant and soil on a dry weight basis.

\*\* Published data not found, values calculated from published data for stable isotopes. "total H or total C", in plants and soil.

fallout.

Studies of absorption via the roots have concentrated has been added. Uptake is greater in culture solutions on crop plants in relatively short-term laboratory than in soil. The relative uptake of the different nuclides, studies (Russell, 1966b). This absorption may be or the relative distributions of a nuclide within a plant,

to the rice fields was derived from direct deposition of contaminated soil in pots under semi-controlled conditions, and by growing plants in soil or culture solutions to which a known amount of radionuclide are similar but not identical using the two methods (Klechkovsky and Gulyakin, 1958a).

Different radionuclides may enter plants or may be translocated in plants at different rates and timing of sampling or length of experiment may be critical. Russell *et al.* (1955) found that the fraction of <sup>144</sup>Ce which reached the shoots of rye-grass after six weeks equilibration was 0.0015 of that for <sup>90</sup>Sr, whereas the corresponding value 22 weeks later was 0.014.

Information on uptake of Sr, Cs and I by plants is abundant whereas information on the other nuclides is much scarcer. Accordingly, below, information for Sr, Cs and I has had to be condensed considerably more and presented more selectively than information for the other nuclides.

#### Strontium (Sr)

Sr is more readily absorbed from soil by plants than any other main component of fallout (Nishita *et al.*, 1961; Russell, 1966*c*). In the UK, field crops may absorb 0.03-4 per cent of the Sr in the soil per year (Russell and Newbould, 1966). Absorption may vary one-two orders with soil type or with plant species (Nishita *et al.*, 1961).

Soil characteristics, other than Ca content and factors which affect rooting depth, are considered to have only minor effects on uptake (Russell and Newbould, 1966). Nevertheless, laboratory experiments indicate that uptake may be greater from sandy soils because of their low content of clay which retains Sr in an ion-exchangeable form (Nishita *et al.*, 1961).

Sr uptake appears to be affected only slightly by pH in the range 5.5-8.5, as indicated by absorption from culture solution by pea seedlings (Klechkovsky, 1957). At pH 4.0, however, uptake was only about 15 per cent of that at pH 5.5-8.5. This contrasts with the uptake pattern observed for plants growing in soil where the presence of other ions, especially Ca, have a dominant influence.

The <sup>90</sup>Sr/Ca ratios for a plant and for the soil in which it is growing are usually about the same, suggesting that, for any one soil, Ca uptake is indicative of Sr uptake (Russell and Newbould, 1966). In a study of uptake by a wide variety of crop plants on four different Swedish soils, the <sup>90</sup>Sr/Ca ratio for the vegetative tissues varied only slightly between species in the same soil, but varied up to about seven times between soils (Fredriksson and Eriksson, 1965). <sup>90</sup>Sr uptake was greatest relative to Ca uptake in soil low in exchangeable Ca. Other workers have obtained similar results (Russell and Newbould, 1966). In field experiments with various UK soils, the <sup>90</sup>Sr/Ca ratio in pasture grass, wheat, sugar-beet and kale changed only slightly as the exchangeable Ca changed from about 8-23 meq 100 g<sup>-1</sup> soil whereas below 8 meg 100 g<sup>-1</sup> it rose sharply from about 1 to 7.5 (Milbourn et al., 1959). Changes in total uptake of Sr parallel these changes; hence, liming acid soils reduces the Sr/Ca ratio in the soil and plant by a factor which seldom exceeds three and decreases Sr uptake (Russell and Newbould, 1966). These effects are not significant in lime-rich soils. Stable strontium usually has little effect on uptake of radiostrontium by plants because the ratio of stable Sr/Ca is so low; however, addition of moderate amounts of stable Sr to soil can increase uptake of radiostrontium by plants by displacing <sup>89</sup>Sr or <sup>90</sup>Sr from ion exchange sites into soil solution (Nishita *et al.*, 1961).

It is well established that Sr and Ca move upwards from plant roots and that downward translocation is negligible; hence the observed decrease in Sr/Ca ratio from root to the tissues most remote from the roots indicates that Sr is less mobile than Ca (Russell and Newbould, .1966). This effect is not large enough to produce an appreciable difference between the mean ratios for the shoot and for the soil solution around the roots.

Sr accumulates in the shoots of plants, particularly in the leaves, rather than in the root. In barley, for example, 96 per cent of added <sup>89</sup>Sr accumulated in the shoots after 56 days of growth (Russell and Newbould, 1966).

The effect of soil organic matter content on Sr uptake is considered to be slight in agricultural soils except where very heavy applications are made (Russell, 1966c). Rotted manure reduced <sup>90</sup>Sr uptake by wheat in pot experiments, particularly on light sandy soils (Klechkovsky and Gulyakin, 1958a), presumably because it increased the retention of Sr by ion exchange significantly in the presence of very low clay concentrations. In pot experiments with barley, Sr uptake from a loam soil was depressed with increasing additions of dried lettuce of low Ca content (Nishita et al., 1956b) Reduction was due partly to immobilization of the <sup>90</sup>Sr in microbial cells. Addition of oak leaf mould with three times the Ca content of that in the lettuce approximately doubled the uptake of Sr into barley shoots which occurred when lettuce was added. Application to a loam soil of 1-2 per cent by dry weight of various types of organic matter, alfalfa leaves, barley straw and lettuce, increased Sr uptake by about 45-80 per cent (Nishita et al., 1956c).

Synthetic chelating agents, which may simulate the effects of naturally-occurring agents from organic matter, do not affect the uptake of Sr significantly (Essington *et al.*, 1962).

Organic matter distribution in soil may affect uptake. Milbourn *et al.* (1959) found 25 per cent greater uptake of <sup>89</sup> Sr by sugar-beet, rye and barley when a field was under stubble at the time of contamination than when it was bare. The Sr adsorbed on the plant remains which were subsequently ploughed into the soil was more readily available to plants than the contamination applied to the bare soil and presumably adsorbed largely on mineral particles. A similar effect may occur when earthworms draw plant remains into an undisturbed soil.

Variation in the distribution of Sr in a soil profile in relation to root distribution can markedly affect the uptake of Sr. In pot experiments involving various UK soils, variations up to three-fold in the <sup>90</sup>Sr/Ca ratio in samples of rye-grass collected over one year were attributed to seasonal variation in the depth of absorption of the Sr. Deeper penetration of the soil by 90Sr over several years caused the <sup>90</sup>Sr/Cr ratio in the grass to decrease by about 10-15 per cent per year (Squire, 1966). Rye-grass, a shallow-rooting species, absorbs most Sr when contamination is restricted to the surface soil layers whereas lucerne, which has particularly deep roots, absorbs considerably less Sr than grass when contamination is mainly in the superficial soil (Russell and Newbould, 1966). Cultivation reduces the uptake of Sr by rye-grass to about one third and would be expected to increase uptake by the deeper-rooting species.

#### Caesium (Cs)

In culture solution, Cs may be taken up by plant roots more readily than Sr, but, in mineral soil, absorption is low because of the high retention of Cs by mineral particles (Klechkovsky and Gulyakin, 1958b; Nishita *et al.*, 1961; Fredriksson *et al.*, 1966; and see above). Uptake may vary six-seven-fold with plant species and four-fold with mineral soil type (Nishita *et al.*, 1961).

Cs is readily mobile in all directions within a plant in contrast to the other radionuclides considered here except perhaps I (Selders and Rediske, 1954; Russell, 1966*b*). The distribution within the plant varies with plant species, length of experiment, and conditions for growth.

In pot experiments using a sandy loam soil and lasting one growing season, levels of <sup>137</sup>Cs in the leaves of beans and lettuces were almost twice that in the stems, and in the leaves of radishes and carrots were three-four times those in the roots (Romney *et al.*, 1957). In wheat grown in soil and water cultures over one season, levels of <sup>137</sup>Cs in the roots were respectively 28 and four times those in the stems (Klechkovsky, 1957). However, as percentages, the distribution appeared more even. In water culture, for example, 17.0, 38.4, 27.4 and 17.2 per cent of the <sup>137</sup>Cs absorbed occurred in the roots, stems, leaves, chaff and grain respectively.

Potatoes, sugar-beet and swedes showed appreciable transfer of <sup>137</sup>Cs to the roots 2-4 months after application as a foliar spray (Middleton, 1958). For potatoes, this transfer amounted to 25-30 per cent of the applied nuclide.

In white oak trees, 33 per cent of <sup>134</sup>Cs injected into the trunk was translocated to the leaves after 140 days, the remainder becoming distributed in stems, branches or roots (Witherspoon, 1963).

Many soil characteristics affect the uptake of Cs by plant roots, but pH has only a small effect. For example, pea seedlings growing in culture solutions showed maximum uptake of <sup>137</sup>Cs at about pH 7.0 with uptake reduced to 29, 76 and 55 per cent respectively at pHs 4.0, 5.5 and 8.5 (Klechkovsky, 1957).

Absorption of Cs from organic soils is considerably higher than from mineral soils (Fredriksson *et al.*, 1966). Barber (1964) found high correlations ( $r = 0.94^{***}$ and 0.95<sup>\*\*\*</sup>) between Cs uptake by rye-grass growing on UK permanent grassland soils and soil organic matter content and cation exchange capacity of the organic matter.

In pot experiments with Swedish soils, the level of Cs in rape growing in peat was about 40 times that of rape in loam, sand or clay (Fredriksson, 1963*b*). Correspondingly, Cs levels in milk were about 1.5-5 times higher for farms on organic soils than for those on clay soils (Fredriksson and Eriksson, 1965).

Synthetic chelating agents do not alter the uptake of Cs by plants significantly as indicated by uptake by bean plants growing in a loam (Essington *et al.*, 1962). The agents tested may have been unable to compete successfully with the soil clay for the Cs or the Cschelate may have been insufficiently stable. Whether naturally-occurring chelating agents behave differently is unknown.

A reduction in uptake of Cs by plants as the exchangeable K content of the soil increases has been noted by many workers (Fredriksson *et al.*, 1966). A similar reduction occurs when the 'residual' K\* increases (Fredriksson, 1963*a*). The addition of K to soils reduces <sup>137</sup>Cs uptake by plants appreciably only when soils contain low levels of K; hence, in pot experiments, <sup>137</sup>Cs uptake by clover was reduced by addition of K after exchangeable K had been reduced by prolonged cropping (Nishita *et al.*, 1960). Progressive fixation of Cs on soil minerals (Squire and Middleton, 1966) may explain reduction in uptake by plants in long-term studies. Exchangeable Ca, like exchangeable K, can reduce uptake of Cs but its effect is usually smaller than that for K (Fredriksson *et al.*, 1966).

Because of its very slow penetration into the soil, Cs is accumulated more by plants which root near the soil surface than by deeper-rooting species (Fredriksson *et al.*, 1966). Christenson and Fowler (1961) found that differences in the depth of the root system in clover, lettuces and grass can cause uptake of <sup>137</sup>Cs from near the soil surface to vary by a factor of twothree.

Reduction of soil oxygen levels, which occurs in some undisturbed soils because of impeded drainage, has a

\*K extractable with hot 2N HCI but additional to the exchangeable K. variable effect on uptake of Cs by plants (Birkle *et al.*, 1965). For example, reduction of oxygen level in soil air from 21 per cent to <1 per cent in laboratory tests increased Cs concentrations in tomato leaves sevenfold, but decreased the concentrations to 28 and 42 per cent of the controls in beans and barley respectively.

In contrast to higher plants (Table 6), certain lichens and fungi can accumulate fallout radionuclides especially <sup>137</sup>Cs (Hanson, 1967; Grueter, 1971). Lichens accumulate <sup>137</sup>Cs by direct sorption from aerial deposits (Hanson, 1967) but accumulation in fungi is probably related to absorption of Cs from the soil (Byrne *et al.*, 1979). Grueter quotes an activity concentration ratio of 16.5 for the mushroom *Paxillus involutus* and soil. Ratios of the same order apply for Cs on a  $\mu$ g g<sup>-1</sup> fungus/soil basis (Byrne *et al.*, 1979).

#### Ruthenium (Ru)

Ru is usually absorbed from soils by plants less readily than Sr or Cs but more readily than the transuranides or Zr (Klechkovsky and Gulyakin, 1958*a*, 1958*b*; Nishita *et al.*, 1961; Auerbach and Olson, 1963); however, Romney *et al.* (1957) found greater uptake than for Sr or Cs with barley on some soils. Uptake may vary by one-two orders of magnitude for the same plant species on different soils or between different plants on the same soil (Nishita *et al.*, 1961; Schulz and Babcock, 1974).

The chemistry of Ru is complex. The element may occur in several different oxidation states or forms in soil (Essington *et al.*, 1963). The form or state may influence uptake by plants.

Beans grown in an Oak Ridge shale soil of pH 4.5-4.9 and watered with effluent containing <sup>106</sup>Ru and high nitrate, and hence probably containing nitrosyl Ru, showed concentration ratios on a dry weight basis of 0.43-20.1 for roots, 0.16-9.8 for stems and 0.44-15.1 for leaves (Auerbach *et al.*, 1957*a*). Uptakes obtained with the commonly-used chloride of <sup>106</sup>Ru are invariably much lower (Auerbach and Olson, 1963). Schulz and Babcock (1974), for example, found mean concentration ratios of 0.0018-0.425 for clover growing in soils varying in organic matter content, cation exchange capacity and pH.

Little generalization seems possible on the relation between Ru uptake by roots and soil pH. In pot experiments with five soils, Schulz and Babcock (1974) found greatest uptake of <sup>106</sup>Ru by clover on the most alkaline soil (pH 8.2) and least on a peat (pH 7.0). Three other soils with pHs 6.3, 6.8 and 7.0 had intermediate uptakes. Romney *et al.* (1957) found greatest uptake by barley on a soil of pH 8.0 and least at pH 6.5. Uptake at pH 4.6 was intermediate. The order of soil pH in relation to uptake varied for different plants. Pea plants grown in culture solutions of pHs 4.0-8.5 showed greatest uptake of Ru at pH 5.5 (Klechkovsky, 1957). Plant uptake of soil Ru appears to be correlated with solubility of the element in water (Nishita *et al.*, 1961; Schulz and Babcock, 1974). The strong retention of Ru by soil organic matter (see above) suggests that uptake by plants on highly organic soils will be high if the Ru is available to the roots; however, Schulz and Babcock (1974) recorded least uptake by clover on a peat soil in a study involving several soil types. This result may be associated with formation of stable complexes of anionic Ru and organic matter.

Synthetic chelating agents had little effect on uptake of Ru by beans in a calcareous soil, possibly because the Ru was unavailable for chelation, the Ru-chelator complex was unavailable for uptake or the Ru was already in a strong complex (Essington *et al.*, 1963).

Ru is usually distributed unevenly in a plant after absorption. Individual bean plants grown on contaminated shale soil showed least Ru in the stems (Auerbach *et al.*, 1957*a*). Deciduous trees growing around radioactive waste pits at Oak Ridge accumulated levels of <sup>106</sup>Ru almost two orders higher than those in nearby trees unaffected by seepage of liquid effluent (Auerbach *et al.*, 1957*b*; 1958). Representative levels in tree leaves, twigs, bark and leaf litter were respectively 8-9 × 10<sup>-2</sup>, 4 × 10<sup>-2</sup> to 2 × 10<sup>-1</sup>, 6 × 10<sup>-3</sup> to 2 × 10<sup>-2</sup>, and 4 × 10<sup>-1</sup>  $\mu$ Ci 100 g<sup>-1</sup> dry weight. The level in the tree leaves increased as the leaves died (cf. I in clover leaves, described below). The level in whole trees decreased as the trees died.

Absorbed Ru often accumulates mainly in the roots of plants. Wheat roots in culture solutions retained 95.7 per cent of absorbed Ru (Klechkovsky, 1957). A similar but slightly lower percentage was retained when wheat plants were grown in soil. Almost all the Ru absorbed by beans, tomato, maize and two xerophytic shrubs, Ceanothus and Adenostoma, in culture solution was retained in the roots (Handley and Babcock, 1972). The level of Ru in the leaves of beans grown in a calcareous soil was 42 per cent of that in the roots (Essington *et al.*, 1963). However, in radish and carrot grown in four different soils, the level was 1-73 times higher in the leaves than the roots (Romney *et al.*, 1957).

Uptake of Ru and accumulation in the roots tend to be far greater for plants in water culture than for plants in soil (Klechkovsky, 1957), probably because of the presence of stable Ru or elements which compete with Ru in the soil but not in culture solution (Myttenaere *et al.*, 1967).

#### Zirconium (Zr) and niobium (Nb)

Of the commoner pollutant radionuclides, except the transuranides,  ${}^{95}Zr + {}^{95}Nb$  are least readily absorbed by plants (Nishita *et al.*, 1961; Russell, 1966*b*).

<sup>95</sup>Zr + <sup>95</sup>Nb accumulate in the roots of plants. <sup>95</sup>Zr + <sup>95</sup>Nb in roots, leaves, stems, pods and seeds of pea plants grown in soil cultures produced respectively 418, 7.1, 2.3, 2.3 and 0.3 disintegrations sec<sup>-1</sup> (Klechkovsky and Gulyakin, 1958*b*). Wheat plants in soil and culture solution showed a similar pattern, with uptakes in soil one to more than two orders less than those in culture solution (Klechkovsky, 1957). 82.7 per cent of the absorbed <sup>95</sup>Zr accumulated in the roots. About 99.9 per cent of the <sup>95</sup>Zr absorbed by barley growing in a clay suspension accumulated in the roots (Jacobson and Overstreet, 1948). If foliar application is made, <sup>95</sup>Zr remains near the site of application (Held, 1963).

Studies of retention by soil (see above) suggest that soil organic matter has little direct effect on uptake of  ${}^{95}Zr + {}^{95}Nb$  by plants except that it will dilute the mineral particles which retain the nuclides.

Studies of retention of  ${}^{95}Zr + {}^{95}Nb$  by soil suggest that uptake is likely to be greatest between pHs 5-8 (see above).

## The rare earth elements, cerium (Ce), yttrium (Y) and promethium (Pm).

Uptake of Ce and other rare earths may vary one-two orders for the same plant species on different soils or different species on the same soil (Romney *et al.*, 1957).

Absorption by plants is negligible except in acid soils (Palumbo, 1963) where the elements are less precipitated and more water-soluble than in neutral or alkaline soils (Russell, 1966b). Pea seedlings in culture solution absorbed ten times more Ce than Pu at pH 4.0, and 200 times more Ce at pH 4.0 than at pH 7.0 (Klechkovsky, 1957). Bean plants in a sandy loam, absorbed seven times more Ce at pH 4.6 than at pH 6.0 (Romney *et al.*, 1957).

As for Sr, <sup>95</sup>Zr + <sup>95</sup>Nb, Ru and Cs (Klechkovsky and Gulyakin, 1957), uptake of Ce is greater in culture solution than in soil. Only one per cent of added <sup>144</sup>Ce was absorbed by two varieties of rice in a sandy loam, low in organic matter (pH unstated), whereas 40 and 70 per cent were absorbed by the two varieties in culture solution (Myttenaere *et al.*, 1967).

Absorbed rare earths tend to accumulate in the roots of plants. In rice growing in flooded soil, in soil at field capacity and in culture solutions, 86.3, 96.4 and 99.5 per cent respectively of the absorbed Ce was found in the roots (Myttenaere *et al.*, 1967). In beans, tomatoes and barley growing in a silt loam or in culture solution, only a few per cent of the absorbed Y and Ce were found in the shoot (Birkle *et al.*, 1965).

Oxygen concentration in the rooting zone of plants influences the mineral content of plant shoots and in culture solutions. Uptake of Ce and Y by shoots of tomatoes, beans and barley increased with increasing oxygen concentrations around the roots (Birkle *et al.*, 1965). However, when the plants were grown in soil, results were too variable to allow generalizations about the effect of oxygen to be made.

Synthetic chelators increased the uptake of Ce, Y and Pm by bean plants growing in a calcareous soil (Essington *et al.*, 1963). Diethylene triamine pentaacetic acid (DTPA), in particular, increased the accumulation of the rare earths in the leaves. Evidence of translocation of a <sup>91</sup>Y DTPA complex from roots to leaves was obtained. This leads to speculation of the importance of naturally-occurring chelators in promoting uptake and translocation of the rare earths.

Little information appears to be available on the effect of organic matter on uptake of rare earths, but Ce is held largely in an ion-exchangeable form in some organic soils (Nishita *et al.*, 1956*a*).

## The transuranides, neptunium (Np), plutonium (Pu), americium (Am) and curium (Cm)

Uptake of transuranides by plant roots, as indicated by the results of laboratory studies and field survey of contaminated sites, is very low (Bennett, 1974; Durbin, 1974; Bulman, 1976; Dahlman *et al.*, 1976; Schulz *et al.*, 1976). Concentration ratios vary greatly with plant species, soil and experimental conditions (Table 6).

The order of uptake by plants, Np>Am=Cm>Pu, is approximately the inverse of their order of hydrolysis and order of adsorption of their oxidation states to colloids, Pu (IV)>Cm (III) = Am (III)>Np (V) (Dahlman *et al.*, 1976), although this order is provisional because of lack of data for Np, Am and Cm. In one test, wheat (grain) absorbed five times more Pu (III, IV, V, VI) than Am (III) from a calcareous fine sandy loam of pH 7.9 (Schulz *et al.*, 1976). <sup>238</sup>Pu appears to be more readily taken up or less readily lost by organisms than <sup>239</sup>Pu since <sup>238</sup>Pu/<sup>239</sup>Pu ratios in soil, plants and animals are 0.05, 0.1 and 1.0 respectively (Hakonson and Johnson, 1974).

Uptake may vary with the chemical state of the radionuclide. In laboratory studies, barley seedlings absorbed 0.0010, 0.0024 and 0.00045 per cent of Pu (II), Pu (IV) and Pu (III) respectively from calcium-bentonite clay suspensions (Jacobson and Overstreet, 1948). This order was the inverse of the extent of binding of the cations by the bentonite. In other studies, wheat absorbed the same amounts of Pu from a pH 6.3 clay loam and from a pH 7.0 loam regardless of whether the Pu was Pu (III) Cl<sub>3</sub> or Pu (III, IV, V, VI) NO<sub>3</sub> (Schulz *et al.*, 1976).

Little generalization seems possible on the uptake of transuranides in relation to soil characteristics. Barley grown for eighteen days on three different soils absorbed most added <sup>23</sup> Pu (0.00022 per cent) from a moderately acid silt loam forest soil and least (0.00006 per cent) from a calcareous silt loam (Wilson and Cline, 1966).

Wheat grown for 120 days on three other soils absorbed about 33 times more added Pu from a pH 7.9 fine sandy loam than from a pH 6.3 clay loam forest soil (Schulz et al., 1976). Am uptake was two-five times greater from the third soil, a pH 7.0 agricultural loam, than from the other two soils. Complexes between Pu (IV) and soil carbonate (Schulz et al., 1976) or organic acids, such as citric, which accumulate in leaves of iron-deficient plants on lime-rich soils (Bulman, 1976) may have facilitated uptake of Pu by wheat at pH 7.9. Some synthetic chelating agents stimulate the uptake of <sup>23</sup> Pu ten-fold (Romney et al., 1970) and <sup>241</sup>Am several hundred-fold (Hale and Wallace, 1970). A similar role has been suggested for natural chelators originating from plant remains in the soil (Romney et al., 1970).

Transuranides may become increasingly available to plants with time. Rye-grass growing on three UK soils, Greensand (pH 5.8, cation exchange capacity (CEC) 10.4 meq 100 g<sup>-1</sup>), Lias loam (pH 7.2, CEC 26.0 meq 100 g<sup>-1</sup>) and Gault clay (pH 7.3, CEC 39.8 meq 100 g<sup>-1</sup>), absorbed more Pu in the second year than in the first year of an experiment (Newbould, 1963). Clover grown on highly contaminated soils from the Nevada test site absorbed seven times as much <sup>239</sup>Pu in 1962 as in 1958 (Romney *et al.*, 1970). However, alfalfa growing on soils at Palomares, Spain, showed no increase in uptake of Pu over the four years after accidental contamination (Bucholz *et al.*, unpublished data, quoted in Francis, 1973).

Increased availability of Pu to perennial plants with time may be explained by the recycling of the element in the rooting zone with the natural chelation of Pu by substances produced in the decaying roots (Romney et al., 1970). Micro-organisms may be involved in laboratory studies since some have been shown to be capable of solubilizing Pu compounds (Wildung and Garland, 1977). In one study, the water-soluble Pu in soil increased from about 0.5 per cent to 1.5 per cent of total Pu in the  $<0.45 \ \mu m$  fraction with increased microbial activity. Correspondingly, the concentration ratio for barley increased from 0.019 to 0.071 for shoots and 0.060 to 2.2 for roots. Am showed similar increases in uptake by barley. In another study, the fungus Aspergillus niger absorbed <sup>238</sup>Pu supplied as nitrate, citrate or microsphere dioxide form and transported it to its spores (Beckert and Au, 1976). In general, specific activities of the spores were two-three times higher for fungus exposed to nitrate and citrate than for fungus utilizing dioxide. For all three forms, uptake by the fungus was greater at pH 2.5 than at 5.5.

Plant species vary in their ability to absorb transuranides (Bulman, 1976) but generalizations and detailed explanation of the differences appear to be currently impossible because of the complexity of the environmental chemistry involved. Lower plants accumulate many fallout radionuclides including transuranides far more than higher plants, but this difference may be due largely to trapping of particulate material and ion exchange at the plant surface (Miettinen, 1976). Levels of Pu in lichens in north temperate latitudes, for example, are one-three orders higher than those in higher plants (Holm and Persson, 1977).

#### Iodine (I)

Studies of the uptake and retention of radioiodine by plants have concentrated on levels of <sup>131</sup> in forage plants and leafy vegetables resulting from aerial deposition. Because <sup>131</sup> has a short half-life (8 days), uptake by plant roots has been considered unimportant. Examination of the possible hazard to man of the very long-lived <sup>129</sup> requires information on uptake by roots, but relevant information is scanty (Bryant, 1970) and is confined to results for <sup>131</sup> or <sup>127</sup> (stable I).

Soldat (1976) assumed that all the 129 deposited remains in the rooting zone and used a concentration factor of 0.02\* based on 127 behaviour to calculate that 1.3 and 0.97 per cent of the 129 accumulated in forage plants and leafy green vegetables respectively enter the plants annually via the soil-root pathway. These calculations appear to exclude the return of uneaten plant material to the soil surface, which is particularly important in uncultivated areas, and the variability in the concentration ratio. Soldat's 0.02 is equivalent to about 0.1-0.2 on a dry plant-dry soil basis. Selders and Rediske (1954) found a ratio of about one for barley on this basis. Menzel (1965) concludes that the ratio for <sup>127</sup>l is between 0.1 and 10. This agrees with the range of 0.85-5.5 found for rye-grass in a sandy loam and receiving various treatments (Whitehead, 1975). Clearly, detailed examination of the factors influencing variability in the soil-root pathway is more justified than Soldat's (1976) estimates suggest.

The form in which the I occurs in the soil is important. In pot experiments with rye-grass in a sandy loam, elemental I and iodide were absorbed to similar extents (Whitehead, 1975). lodate was taken up much more readily than iodide or elemental I, except where organic matter was added as a treatment. The latter addition markedly reduced uptake of all three forms of I. Addition of chalk, which raised the pH to 7.3, led to a marked reduction in uptake of iodide, but a marked increase in uptake of iodate. Katalymov and Churbanov (1960) found that liming decreased the uptake of 127 and 131 by oats, grass and lettuce in pot and field experiments. Uptake of 127 and 131 by bush beans in culture solution or by barley in soil was reduced about four times when the pH was raised from 4-7, apparently because of an effect on the root surface (Selders and Rediske, 1954). Changes in uptake of I with pH may be associated partly with changes in solubility particularly when the I occurs as iodate, and partly with effects of I compound on the plants, as in the case of iodide

\* pCi l kg -1 wet plant tissue per pCi kg +1 dry soil.

(Whitehead, 1975). Except in one study, a pot experiment involving fertilizer application (Katalymov and Churbanov, 1960), chloride, which may be added to soil in fertilizers and marine sources, appears not to decrease I uptake by plants (Whitehead, 1963b).

Uptake of I may vary with plant species. In culture solution, *Salicornia* removed 8.8 per cent of the <sup>131</sup> supplied as iodide whereas *Aster* absorbed only 1.7 per cent (Fowden, 1959). Both are salt marsh plants. In Whitehead's (1973b) studies, two clovers and two grasses absorbed similar amounts of I from a culture solution containing  $0.2 \times 10^{-7}$  M iodide. Insofar as differences did occur, they largely reflected differences in dry matter yield between species. Increasing the level of stable I in soil culture increased the uptake of radioiodine by barley (Selders and Rediske, 1954; cf. Ru).

I absorbed by roots tends to be rapidly but unevenly distributed throughout a plant (Selders and Rediske, 1954). Cereal grain may contain one tenth of the I in straw (Chilean Iodine Educational Bureau, 1956). In plants grown in culture solution, I tends to accumulate most in the roots at lower concentrations of I which are closest to field concentrations. About 50-70 per cent

of the absorbed I occurred in the roots of bean, tomato, barley and Russian thistle after growth for four days (Selders and Rediske, 1954). Whitehead (1973*b*) obtained similar results for clovers and grasses in the culture solution, but found higher accumulation of I in shoots than in roots at the higher of several concentrations of I used. In beans, I accumulated continually over the 28 days in one test (Selders and Rediske, 1954). The observed variation in distribution of I in plants may therefore be due partly to the time required for equilibration. Highest levels of I in plant shoots usually occur in the older leaves (Selders and Rediske, 1954; Langston, 1956) or in senescent and dead leaves (Whitehead, 1973*b*).

I content of plants, as an indication of relative I uptake, varied by one-two orders of magnitude, even for plants growing on the same soil (Chilean Iodine Educational Bureau, 1956; Johnson and Butler, 1957; Shacklette and Cuthbert, 1967). I content may vary over one order of magnitude for the same plant type growing on different soils, but I contents of plant and soil (Hanson, 1963) or I content and soil type (Shacklette and Cuthbert, 1967) are uncorrelated. This lack of correlation is not surprising in the light of the large number of factors which influence the I content of plants.

### UPTAKE AND RETENTION OF RADIONUCLIDES BY ANIMALS

Much information is available on the uptake and retention of radionuclides by terrestrial vertebrates and invertebrates. Only a brief outline of the main aspects of these topics can be given here. The main sources of information include publications from the Oak Ridge National Laboratory, Tennessee, and the Savannah River Ecology Laboratory, South Carolina, on various different animals, from several countries, on I in vertebrates including man, and from North America and Scandinavia on fallout radionuclides, particularly Cs, in the lichen-reindeer-man pathway.

The extent of uptake varies greatly with the radionuclide (Table 7) and with the animal type. Vertebrates and those invertebrates which possess a calcareous exoskeleton or shell, e.g. millipedes, crustaceans and many molluscs, will have a higher uptake of Ca and Sr than other invertebrates.

Table 7. Degree of absorption of elements by man (International Commission on Radiological Protection, 1959)

Absorption (%)	Element
70-100	C, S, I, Cs
20-70	Sr
1-20	Ru
< 1	Y, Zr, Nb, Ce, Pm,
	Np, Pu, Am, Cm

Uptake may occur through internal respiratory surfaces, e.g. in the lungs of vertebrates, via the gastrointestinal tract and probably also through the skin. Some radionuclides will enter the body largely by one route, others by another route and yet others appreciably by several pathways. In general, elements such as Cs and Sr, which are absorbed strongly from the soil by plants (Table 6) and are also absorbed readily by animals, will enter the body mainly through the gastrointestinal tract. Others, such as Pu and Am, which are not readily absorbed by plants and animals but which occur as particles  $< c.10 \ \mu m$  diameter (Comar, 1966), enter the body via the respiratory organs and gastrointestinal tract (Linsley *et al.*, 1978).

The time taken for the transfer of radionuclides to various parts of the body after ingestion or inhalation varies with radionuclide (Table 8), with body part and with animal type. Attainment of peak radioactivity in a particular part of an animal may take a considerable time even in metabolically active animals and for readily mobile radionuclides. For example, a peak in the <sup>131</sup>I in the thyroids of kangaroo rats feeding continuously on contaminated food was not reached until about the tenth day (French and Larson, 1963).

Distribution of a radionuclide within a body varies with radionuclide, animal and contamination method. Table 9

Table 8. Time of appearance of maximum concentration ofradioactivity in blood of dairy cows after ingestion(Lengemann & Comar, 1956)

Radionuclide	Time of peak activity ín blood (h)		
131	4-6		
<sup>137</sup> Cs	20		
<sup>90</sup> Sr	20-40		

illustrates variation with radionuclide in a vertebrate. In invertebrates, similar variation occurs. Bocock and Pickering (unpublished data) found that the bulk of <sup>85</sup>Sr ingested by a millipede occurred in the faeces within 24 hours. Over 90 per cent of the remainder occurred in the calcareous exoskeleton (cf. Table 9).

Table 9. Distribution of some radionuclides in various tissues of the rat after intramuscular or intravenous injection (Durbin, 1960)

	Percentage of absorbed dose									
Radionuclide	Time after administration (days)	Bone	Kidney	Liver	Muscle	GI tract & contents	Thyroid	Blood	Excreta	
<sup>90</sup> Sr	4	44.0							56.0	
<sup>95</sup> Zr	4	34 • 9	4 · 3	6.6	_		_	_	17.8	
<sup>95</sup> Nb	4	16.2	2.9	8 · 4	8.0		<del></del>	7.7	39.4	
<sup>97</sup> Ru	1	6.5	2.7	5.8	21.4		_	7.4	19.4	
<sup>131</sup>	0.375	1.2		1.0	_	8 · 4	23.6	_	34 • 4	
<sup>134</sup> Cs	4	1.5	_	2 · 1	38.5	_	—	_	44.9	
<sup>144</sup> Co	4	27 · 7	—	51·0	-	_	-	_	6 · 0 (Urine) 8 · 0 (Faeces)	
<sup>237</sup> Np	1	44 • 4	2.6	8.5	_	—	—	_	36.9	
<sup>239</sup> Pu	4	70·9	1.8	8.4	_	_	_	_	8.2	
<sup>241</sup> Am	4	19.1	2.3	35.7	_	_	_		34.7	
<sup>242</sup> Cm	4	29.0	1.6	40 . 2	_	_	_		20.0	

Retention of radionuclides varies greatly with the radionuclide, the animal species, the part of the animal under consideration and the elimination conditions. <sup>85</sup>Sr in millipedes, for example, is lost from soft tissues and the exoskeleton with effective half-lives of respectively a few days and 50-55 days (Bocock and Pickering, unpublished data). Elimination from some animals, for example <sup>32</sup>P from cotton rats, is temperature-dependent (Wagner, 1970). In some animals it follows a course which is best expressed by a multicomponent exponential curve, as in the millipede example.

Elimination from other animals, e.g. <sup>134</sup>Cs in a beetle (Reichle and Dodson, 1965), is essentially linear with time.

Animals such as millipedes and crustaceans conserve Ca and Sr when they moult by transferring a high percentage of the elements in the old exoskeleton to the new larger exoskeleton. The rate of radiostrontium loss from the body is therefore not changed appreciably by the moult (Bocock and Pickering, unpublished data).

## RELEASE OF RADIONUCLIDES FROM PLANT AND ANIMAL REMAINS BY DECOMPOSITION

A vast amount of information is available on the decomposition of plant remains in temperate soils (Dickinson and Pugh, 1974). The effects of plant species, soil physical and chemical characteristics, micro-organisms and fauna have all been examined in detail. Each of these factors can markedly affect the rate and course of decomposition. Information on the decomposition of animal remains is less abundant and often refers primarily to use of animal waste as a soil amendment.

Some data are available on the release of radionuclides from plant remains and the various factors which affect the release, particularly in publications from the Oak Ridge National Laboratory, Tennessee. Few, if any, data are available on release from animal remains but approximate values could be obtained from known decomposition rates and radionuclide content of live animals.

In field experiments, in which pine, oak and dogwood leaf litters labelled with <sup>60</sup>Co, <sup>85</sup>Sr, <sup>106</sup>Ru and <sup>134</sup>Cs were enclosed in nylon net bags and allowed to decompose for one year, decomposition, as indicated by weight loss, was only slightly higher in an oak stand than in a pine stand (Olson and Crossley, 1963). The rate of loss of <sup>60</sup>Co, <sup>85</sup>Sr and <sup>106</sup>Ru from the litter was about the same as that for weight loss due to microbial and faunal activity, whereas <sup>134</sup>Cs was lost considerably more rapidly as a result of leaching. In oak litter in the oak stand, for example, the retention half-life for dry weight was 44 weeks compared with only 17 weeks for <sup>134</sup>Cs.

In a similar study, labelled beech leaf litter lost 57-67 per cent of its weight in one year but 66-79 per cent, 84-90 per cent and 91-95 per cent of the <sup>134</sup>Cs after seven weeks, one year and two years respectively (Olson *et al.*, 1965). A 2.5 cm deep slab of soil placed under the litter retained 66, 66 and 42 per cent of the original added <sup>134</sup>Cs in pine, oak and maple stands respectively.

Immobilization of Cs by micro-organisms in deciduous tree litter has been demonstrated in both laboratory and field studies (Witkamp *et al.*, 1967; Witkamp, 1969). Immobilization of Sr was slightly less than that of Cs. Cs immobilization increased with increasing temperature and was nil when the litter was dry. Freezing or drying mobilized over 95 per cent of the Cs in microbial tissue, but only a third of the Cs in decaying litter because of resorption on to the litter.

Our own observations on invertebrates in southern Cumbria suggest that the soft tissue of animals decomposes in a few days to a few weeks, depending on weather conditions and the activities of carrion feeders. Decomposition of invertebrate exoskeletons may take several months at field soil temperature. In laboratory studies at 25°C, 50 per cent of the C in cuttlefish chitin and insect wings was released in 1.5 and 6 weeks respectively (Okator, 1966*a*, 1966*b*). Data on the decomposition of whole vertebrate skeletons are apparently unavailable, but from archaeological evidence, release of radionuclides would be expected to occur over tens to many thousands of years, depending on the size and type of bone and decomposition conditions. Radionuclides may move out of terrestrial ecosystems in several different ways. Those which are volatile (129]), involved in the water cycle (3H), associated with fine soil particles (90Sr, 137Cs, 239Pu) or involved in the metabolism of organisms (14C), may become airborne. The more soluble nuclides (1291) are lost mainly in surface water or water which has leached the soil profile. Nuclides associated with fine mineral particles (90Sr, 129], 137Cs, 239Pu) or organic matter (129], 137Cs) are lost in material eroded from soil surfaces especially from surfaces exposed by man's activities. Radionuclides are removed from ecosystems in crops, including timber and animal products.

Losses of radionuclides do not appear to have been calculated for UK ecosystems although many relevant data on levels of radioactivity in air, soil, plants and animals are available (e.g. Bryant et al., 1957; Cambray et al., 1977). Some data on rates of transfer out of ecosystems of dry matter and nutrients in forest products, cattle and sheep were assembled by Bocock and Adamson (1976). Any calculations of radionuclide losses using available data would give only approximate values because of lack of information on variability in transfer rates and on fractionation effects, e.g. selective

OUTPUTS OF RADIONUCLIDES FROM TERRES- loam, eroded soil contained 1300 pCi <sup>90</sup>Sr kg<sup>-1</sup> whereas soil from the plough-layer contained only 45 pCi kg<sup>-1</sup>. This result emphasizes the degree of fractionation of soil particles which can occur.

> Permanent pasture soil on experimental catchments in Ohio contained 34.7 mCi km<sup>-2</sup> <sup>90</sup>Sr whereas accumulated fallout was estimated to be 29 mCi km<sup>-2</sup> (Frere and Roberts, 1963). The fallout value may be within the calculation error of the content value but it suggests that <sup>90</sup>Sr loss from the catchments was small. No soil loss from these catchments was recorded. Six other catchments, carrying grass, maize or wheat crops, lost 46-57 per cent of incoming fallout in the study period. Loss was greatest towards the bottom of the slope and was attributed to soil erosion, soil leaching and removal in crops. The relative importance of these processes was not examined.

> Application of <sup>137</sup>Cs to experimental plots on a silt loam in Tennessee and subsequent monitoring of movement of the radionuclide indicated that most of the <sup>137</sup>Cs loss was associated with soil erosion rather than with soil leaching, the loss decreasing with increasing vegetation cover (Table 10). Condition of the vegetation, amount and intensity of precipitation appeared to be major factors affecting the rate of <sup>137</sup>Cs loss.

Table 10.	Results	of	a two-year	study of	mov	ement	of	<sup>137</sup> Cs	applied	to
experiment	al plots	in	Tennessee	(Rogows	ki &	Tamui	a,	1970)		

Plot treatment	Run off* (cm)	Infiltrate* (cm)	Soil loss (g m-²)	<sup>137</sup> Cs loss (%) Soil Solution		
Bare					,	
soil	149	96	5258	42·3	0.6	
Poor meadow	78	167	250	18.8	0 · 6	
Good meadow	40	205	85	6·4	0 · 4	

\* Precipitation 245 cm

removal of particles of a particular size and type and hence radionuclide content from a catchment.

Losses of Sr, Cs and Pu radionuclides by soil leaching and erosion have been studied for a few catchments in the USA. 0.35, 4.1 and 4.5 per cent of the annual <sup>90</sup>Sr input in fallout were removed annually in eroded soil and solution in run-off from a silt loam in Wisconsin carrying respectively clover, oats and maize (Menzel, 1960). Comparable values for a sandy loam in Georgia carrying oats, maize and peanuts were 0.45, 1.39 and 1.44 per cent. Percentages varied only slightly between months except in one month on a silt loam where 27.7 and 28.6 per cent of fallout <sup>90</sup>Sr were removed. Losses of <sup>90</sup>Sr were greater on plots with greater soil loss. For most plots, concentrations of <sup>90</sup>Sr in eroded soil were about ten times those in the soil remaining on the plots. For the clover plot on the silt

Although levels of Pu in the top few cm of soil are about three times greater for pastures and forests compared with those in agricultural soils, total erosional removal of Pu is probably greater from the latter since soil erosion rates for agricultural soils are typically ten or more times greater than those for pastures or forests (Muller et al., 1978). Although the mechanisms of attachment of Cs and Pu to soil particles differ, the erosional behaviour patterns of the two nuclides on an agricultural and on a pasture watershed in Ohio were similar (Muller et al., 1978). Cs may be leached considerably more readily from soils which are poor in illitic minerals which retain Cs strongly than from other soils; hence the erosional behaviour of Cs and Pu may differ on illite-poor soils (Muller et al., 1978). Cumbrian soils tend to be rich in illite and chlorite (see above).

Only about 0.05 per cent of the total <sup>239, 240</sup> Pu accumulated in drift soils on a 1401 km<sup>2</sup> predominantly agricultural watershed in Ohio was removed annually by erosion and leaching (Sprugel and Bartelt, 1978). Filtered water and suspended sediment contained 0.15fCi litre<sup>-1</sup> and 14fCig<sup>-1 239, 240</sup>Pu respectively, emphasizing the strong association between Pu and soil particles.

### DISCUSSION

Ideally, studies of the distribution and movement of pollutant radionuclides in ecosystems should be undertaken by direct measurement in the field under conditions which are as natural as possible, using radionuclides in the chemical and physical forms normally encountered in the field. This ideal approach is possible for distribution studies but not for many transfer studies because of the complexity and variability of the processes involved and the impracticability of measuring certain transfers. Transfer of radionuclides to plants via their leaves, for example, will be difficult to estimate because (a) input and loss via the leaf surface and translocation from the roots may occur simultaneously, (b) these transfers may vary considerably with season, plant form and weather conditions, and (c) for some plants and soils concentrations of some radionuclides may be too low and transfers too Given these problems, therefore, data have slow to be collected using several different approaches and ultimately synthesised to give the required information on distribution and transfer.

In some areas near nuclear installations, pollutant radionuclides occur in concentrations high enough to allow direct measurement of transfers between parts of ecosystems. Sedimentary soils in the salt marshes around the Ravenglass estuary in Cumbria, for example, contain 2-220 pCi g<sup>-1 239, 240</sup>Pu and 5-150 pCi g<sup>-1</sup><sup>134, 137</sup>Cs from Windscale, the lower values occurring around the high tide mark, the higher ones in the exposed sediment at the seaward end of the marsh and amongst silt-trapping plants (James et al., 1978; Dr. E. I. Hamilton, Institute for Marine Environmental Research, personal communication; A. D. Horrill and K. L. Bocock, ITE, unpublished data). Some other radionuclides, <sup>95</sup>Zr + <sup>95</sup>Nb, <sup>106</sup>Ru and <sup>144</sup>Ce, also occur in these sediments at easily measurable concentrations (Mitchell, 1975).

In terrestrial ecosystems which are not inundated periodically by seawater, concentrations of pollutant radionuclides approach the fallout background levels except near the coast where some transfer from the sea in aerosols and bird droppings may be expected. James *et al.* (1978), for example, found 0.4 pCi g<sup>-1</sup><sup>239, 240</sup>Pu and 0.3 pCi g<sup>-1 241</sup>Am (dry weight) in the top 5 cm of a sand dune soil about 3 km north of Ravenglass. This Pu value is fifty times the mean value for the top 30 cm soil in grasslands and woodlands over England and Wales (Cawse *et al.*, p. 81 in United Kingdom Atomic Energy Authority, 1978). The difference between the Pu concentrations in the dune soil and elsewhere was partly due to the difference in sampling depth and the presence of coarse material in

some of the UK samples but not in the dune soil. This is supported by data on <sup>137</sup>Cs. Cawse's mean concentration for <sup>137</sup>Cs in UK soils, 0-30 cm, was 0.5 pCi g<sup>-1</sup> whereas the <0.2 mm fraction of the top 10 cm of a deciduous woodland soil near Grange-over-Sands, southern Cumbria, contains about 2.4  $\pm$  0.2  $\,$  pCi g^{-1} (Dr. S. R. Aston, Lancaster University personal communication). All the radionuclide concentrations mentioned in this paragraph are likely to be too low for direct measurements of radionuclide transfers to be practicable. Inputs of some radionuclides, for example in rain, are often measurable, but uptake by plants from soil may be difficult to measure, especially for elements such as Pu and Am which have concentration ratios of 10-5-10-3 and 10-4-10-2 respectively (Bulman, 1976).

Where direct measurement of radionuclide transfers under natural field conditions is impracticable, estimates have to be obtained indirectly using any available information on residence times in particular parts of ecosystems and transfer rates for analogous stable nuclides between parts, data on patterns and amounts of discharge by nuclear installations, observed concentrations and total amounts of radionuclides in particular ecosystems, and transfer data obtained by experimental addition of appropriate radionuclides to ecosystems or organisms. Mathematical modelling is likely to be an important feature of this approach.

Experimental addition of radionuclides has to be used with great caution because the environmental behaviour of radionuclides may vary with concentration and chemical and physical form of nuclide and with experimental conditions used. Moreover, use of tracer concentrations of radionuclides facilitates measurement of concentrations but may be incompatible with measurement of fallout background levels without stringent precautions to avoid cross-contamination of samples or duplication of counting equipment.

Although the main emphasis in future studies by the Institute is likely to be placed on environmental pathways, radionuclide concentrations will also be surveyed and monitored for specific purposes: (a) in combination with measurement of various environmental variables and use of multivariate analysis to provide clues to the importance of individual environmental variables, pathways or processes in determining radionuclide concentrations, (b) to provide background data for studies of specific organisms or processes, and (c) where insufficient data on spatial and temporal variability are available for the UK, e.g. for the transuranides.

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### APPENDIX

## ITE data bank of radioecological literature developed by A. Nelson

This data bank was developed for information collected during the literature survey for the above paper. The aim was to organize the information into a rapidly retrievable form for use by research workers and interested organizations, and in particular, to provide lists of literature references and abstracts for specific radionuclides and aspects of their ecology (Table 11). Detailed numerical data are not included in the bank because of doubts about their value in isolation from full details of methods of data collection. However, literature containing a particular type of data can be identified using the keyword DATA with qualification as described below. The bank is essentially a small specialized bank of information highly relevant to a limited set of research objectives and is intended to complement rather than compete with the large international data bases such as those in the Department of Trade and Industry's Dialtech system or the Lockheed system.

The bank is stored on one of the disks of ITE's Merlewood PDP 11/34 computer. It contains references in a standard format consisting of author, date of publication, title and literature source with space for an abstract. Currently, the latter consists largely of a list of standard keywords and qualifying terms applicable to the reference, but any other relevant information may also be included. Each reference plus abstract may vary in length, size being limited only by the size of the computer disk. With the current average size of an entry, about 3,000 references and abstracts may be stored on one disk. A bank larger than this would be unwieldy to operate using the chosen system of information retrieval.

The bank consists of three main parts, a reference/ abstract store, a store index, and a keyword index. The reference/abstract store is in a virtual file, a type of computer file which allows rapid direct access to an individual item and storage of items of variable length in a minimum of space. The order of the references is determined by the input sequence and any subsequent editing, each reference being allocated an index number on input. The store index indicates the location and length of each item in the store. The keyword index consists of a series of files, one for each standard keyword. Each file contains the index numbers of all the references to which the keyword in question refers ; therefore, in a search of the bank using standard keywords, search of the reference/abstract store is unnecessary to determine the presence of references of interest and search time is correspondingly reduced.

Currently, each reference is given keywords according to a standard list of about 160 words. The latter may be modified or extended easily as new terms become necessary, or the interests of the users change. For convenience during the allocation of keywords and to facilitate rapid selection of keywords for searching, the words are grouped in 10 main subdivisions covering the following topics: radionuclide, type of ecosystem type of organism, soil characteristic, climatic variable, location of study in the world, management practice, aspect of methodology, aspect of the nuclear industry, and a miscellaneous section. These subdivisions are not used as distinct entities by the computer.

The references are compiled conveniently on index cards prior to input of information to the bank. Additional information, e.g. qualifying terms for keywords, is also entered on the cards for subsequent punching as part of the abstract. Code letters A or FT appear at the end of each abstract indicating whether the reference was given keywords from a published abstract or from the full text.

References are inserted in the bank in two ways: on-line, that is by punching them in directly on a computer terminal keyboard, or indirectly, as a paper tape input. The former method is convenient when adding only a few references; the latter enables large numbers of references to be punched off-line, edited, and inserted rapidly in the bank using a computer program which automatically updates the appropriate keyword files as each reference is inserted.

A search of the data bank may be performed using either any combination of specified keywords or any string of characters. The former search uses only the keyword files and is valuable where a particular topic, or combination of topics, is of interest. For example, the topic PLUTONIUM in FOREST SOIL INVERTE-BRATES would involve a search using the four keywords in capital letters. Such searches are made in one operation after input of all the relevant keywords, unlike some commercially available information systems where a keyword profile has to be built up manually in several operations. Search on a character string uses the main reference/abstract store and allows, for example, all the works of a particular author to be listed, or extraction of the references containing data of a particular type. In the latter case, the keyword DATA is gualified in the abstract and the computer searches for the string DATA (QUALIFICATION).

At the end of a search, the number of relevant references is indicated. If this number appears to be large, that is if the search field specified was rather wide, the option of narrowing the field and searching again is available. When the number of relevant references is acceptably low, the references may be printed in alphabetical order of names and authors, in date of publication order, or in order of access. The listing may be made in pages of A4 size if required and the references produced in any desired format after minor changes in the output

program. The lists are therefore suitable for direct inclusion in reports or scientific papers for publication.

The main disadvantages of the data banking system are the few minutes per reference required both for the allocation of keywords and for punching of references and abstracts for direct input to the computer. In contrast, the advantages are many. The system can be operated easily by users with a few minutes' training. Several fail-safe facilities in the computer programs prevent inexperienced users corrupting the bank and wasting time during searches. The bank may be updated simply and automatically at any time. It is available locally at short notice. The computer's time-sharing facility allows several workers to use the bank simultaneously. Printing occurs almost immediately and often more rapidly than for some commercial systems. The format of the print-out may be varied as required (see above). As REVIEW is one of the standard keywords, details of review papers, which are currently difficult to identify and list from commercial systems, are extracted easily. By juxtaposition of words in the abstract during the allocation of keywords and searching on an appropriate character string, literature with a particular type of content, e.g. data, may be identified. Currently, the bank contains about 140 references relevant to the current desk study. Further development and use of the bank depend on the future needs of the Institute.

Table 11. Sample computer print-outs of information from ITE's data bank of radioecological literature.

a) extraction of references and abstracts relevant to five keywords, PLUTONIUM, LEVEL, FOREST, SOIL, DATA.

b) extraction and listing in alphabetical order of author and (on A4 size pages) of references relevant to two keywords PLUTONIUM and SOIL.

Table 11 (a)

Enter the keywords you wish to search on as asked Terminate entries by typing END.

RUNNH

Key 1 ? PU Key 2 ? LEVEL Key 3 ? FOREST Key 4 ? SOIL Key 5 ? DATA Key 6 ? END Search on keywords complete A total of 3 relevant references Do you want to list these references? YES Do you want the references in ALPHA order? YES Do you want the ABSTRACT included ? YES

ADRIANO, D. C., PINDER, J. E. (1977). AERIAL DEPOSITION OF PLUTONIUM IN MIXED FOREST STANDS FROM NUCLEAR FUEL REPROCESSING. J. ENVIRON. QUAL.6(3),303-307. MENT, DEPOSITION, UPTAKE, RETENTION, FIELD, MODEL, A

PLATT, R.B., PALMS, J.M., RAGSDALE, H.L., SHURE, D.J., MAYER, F.G. & MOHRBACHER, J.A. (1973). EMPIRICAL BENEFITS DERIVED FROM AN ECOSYSTEM APPROACH TO ENVIRONMENTAL MONITORING OF A NUCLEAR FUEL REPROCESSING PLANT: IN: IAEA, ENVIRONMENTAL BE-HAVIOUR OF RADIONUCLIDES RELEASED IN THE NUCLEAR INDUSTRY: PP673-700.

RADIONUCLIDE (BE7, NA22, EU155, PU245), H3, KR85, SR89, SR90, Y90, Y91, ZR95, RU103; RU106, I129, I131, CS134, CS137, CE144, CE141, K40, MN54, CO57, CO60, SB125, BA140, RA226, TH232, PU238, PU241, PU242, ECOSYSTEM, FOREST, ARABLE, FRESHWATER, MAN, TREE, MAMMAL, VERTEBRATE, INVERTEBRATE, SOIL, TYPE, WATER, AIR, USA, SAMPLING (FIELD), CHEMICAL ANALYSIS, DATA ANALYSIS, REPROCESSING, LIQUID EFFLUENT, GASEOUS EFFLUENT, DATA, LEVEL, RADIATION, FIELD, SURVEY.

(USEFUL SUMMARY OF VALUES OF ECOSYSTEM APP-ROACH) FT

ROBINSON, W.L., PHILLIPS, W.A. & COLSHER, C.S. (1977). DOSE ASSESSMENT AT BIKINI ATOLL. 51PP. UCRL-51879 (PT.5).

CS137, SR90, PU239, PU240, AM241, PU241, CO60, PU238, MARINE, LITTORAL, FOREST, ARABLE, VERTEBRATE, MAN, PLANT, TREE, BIRD, INVERTEBRATE, SOIL, PACIFIC, FALL-OUT, DATA, SPATIAL VARIATION, TEMPORAL VARIATION, FOOD, UPTAKE, MODEL, FIELD, INHALATION, INGESTION. FT

#### TABLE 11 (b)

ADRIANO, D.C., PINDER, J. E. (1977). AERIAL DEPOSITION OF PLUTONIUM IN MIXED FOREST STANDS FROM NUCLEAR FUEL REPROCESSING. J. ENVIRON. QUAL.6(3), 303-307.

COHEN, B. (1977). HAZARDS OF PLUTONIUM TOXICITY. HLTH. PHYS.32, 359-380.

COMAR, C.L. (ED.). (1976). PLUTONIUM: FACTS: INFER-ENCES. SUMMARY REPORT. EPRI-EA-43-SY. 19PP.

ENEA/OECD. (1971). RADIOACTIVE WASTE MANAGEMENT IN WESTERN EUROPE, 126PP.

HAMILTON, E.I. (1970). THE CONCENTRATION OF URANIUM IN AIR FROM CONTRASTED NATURAL ENVIRONMENTS. HLTH. PHYS. 19, 511-520.

JACOBSON, L., OVERSTREET, R. (1948). THE UPTAKE BY PLANTS OF PLUTONIUM AND SOME PRODUCTS OF NU-CLEAR FISSION ADSORBED ON SOIL COLLOIDS. SOIL SCI. 65, 129-234.

MCHENRY, J.R., RHODES, D.W., ROWE, P.P. (1956). CHEMI-CAL AND PHYSICAL REACTIONS OF RADIOACTIVE LIQUID WASTES WITH SOILS. USAEC, PROC. SEMINAR ON SANITARY ENGINEERING ASPECTS OF THE ATOMIC ENERGY IN-DUSTRY. PP 170-190. TID-7517 (PT 1A).

MEANS, J.L., CRERAR, D.A., BORCSIK, M.P. (1978). RADIO-NUCLIDE ADSORPTION BY MANGANESE OXIDES AND IMPLICATIONS FOR RADIOACTIVE WASTE DISPOSAL. NATURE 274, 44-47.

MULLER, R.N. (1978). CHEMICAL CHARACTERIZATION OF LOCAL AND STRATOSPHERIC PLUTONIUM IN OHIO SOILS. SOIL SCI. 125, 131-136.

MULLER, R.N. & TISUE, G.T. (1977). PREPARATIVE-SCALE PU238, PU239, PU240, FOREST, TREE, SOIL, ORGANIC SIZE FRACTIONATION OF SOILS & SEDIMENTS AND AN MATTER, USA, REPROCESSING (PLANT), GASEOUS EFFLU- APPLICATION TO STUDIES OF PLUTONIUM GEOCHEMISTRY. ENT, FALLOUT, DATA, SPATIAL VARIATION, LEVEL, MOVE- SOIL SCI. 124(4), 191-198. (BIOL. ABSTR. 65(4), ND. 24331).

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