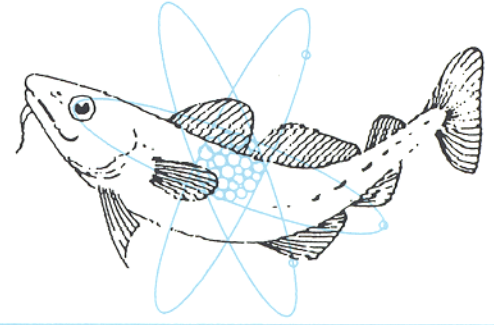


**AQUATIC ENVIRONMENT
MONITORING REPORT**
Number 32



**A review of radioactivity in the Irish Sea
A report prepared for the Marine
Pollution Monitoring Management Group**

P.J. Kershaw, R.J. Pentreath, D.S. Woodhead and G.J. Hunt



Directorate of Fisheries Research
Lowestoft, 1992

MINISTRY OF AGRICULTURE, FISHERIES AND FOOD
DIRECTORATE OF FISHERIES RESEARCH

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1. INTRODUCTION

The principal sources of radioactive waste discharges, containing artificial radionuclides, to the Irish Sea and all UK coastal waters, are related to power generation and the nuclear fuel cycle (Figure 1). Discharges are dominated by those from the Sellafield nuclear fuel reprocessing plant (Table 1). By comparison, other sources such as hospital, industrial and research users are rather small. It follows that most emphasis in this report, has been placed on describing our current understanding of the consequences of the Sellafield discharges.

The main substance of this report was prepared by R. J. Pentreath (1985) as a contribution to the IAEA TECDOC-329 on the 'Behaviour of radionuclides released into coastal waters' (IAEA, 1985). The original paper has been considerably extended and edited, and the present report attempts to represent the current state of knowledge on radioactivity in the Irish Sea. In general, greater space has been devoted to

earlier work, up to the mid-1980s, which provides a background, and historical perspective, to the later studies. The latter are dealt with more concisely and with appropriate reference to the literature which should be consulted when greater detail is required. Any omissions, which are regretted, are the responsibility of the principal author (P. J. Kershaw) who would welcome comments. The SI system has been used to present radiological data (i.e. Bq, Gy, Sv) with appropriate conversions made when describing earlier studies which reported in non-SI units (i.e. Ci, rad, rem).

2. SELLAFIELD DISCHARGES

2.1 Controls

The British Nuclear Fuels plc (BNFL) Sellafield Site (formerly referred to as Windscale) is situated on the Cumbrian coast of England (Figure 1). The site

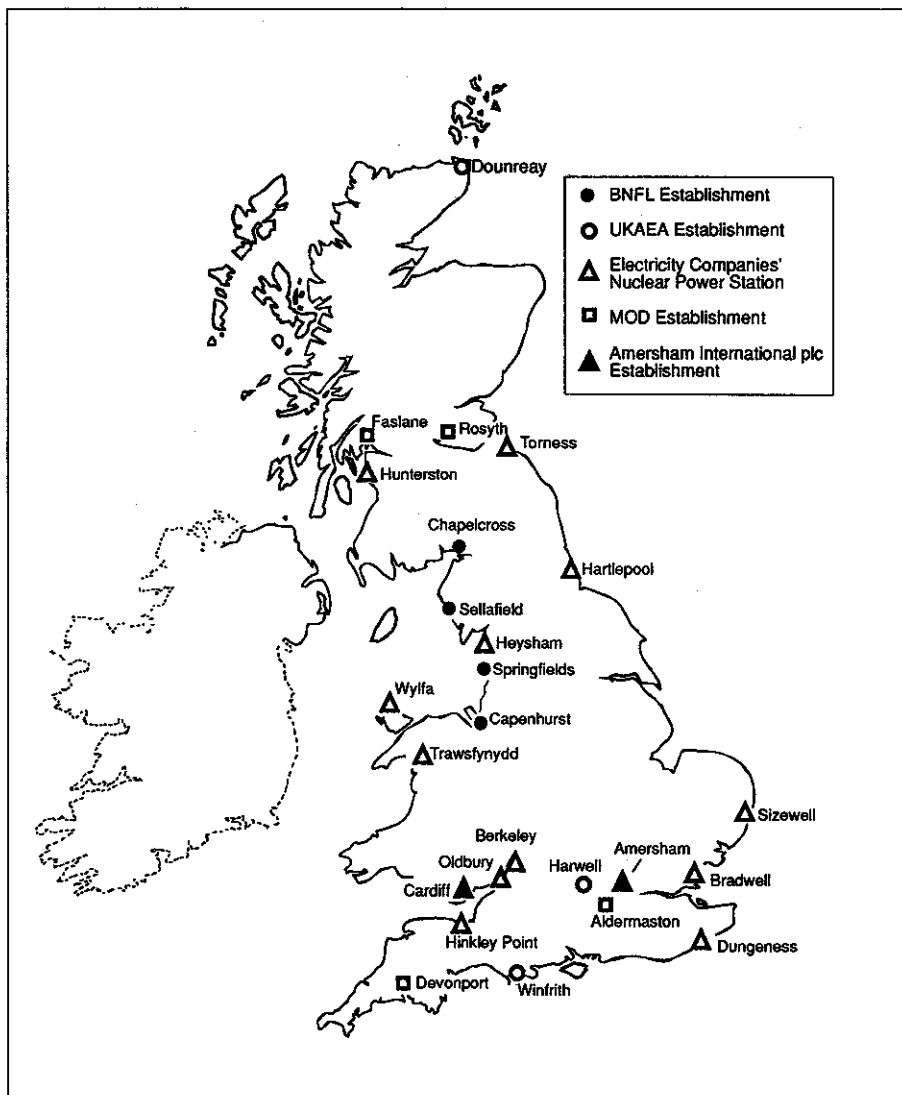


Figure 1. Main UK sources of artificial radionuclides

Table 1. Principal discharges of liquid radioactive waste from UK nuclear establishments in 1990 (from: MAFF, 1992)

Establishment	Radioactivity	Discharge limit (annual equivalent), TBq	Discharges during 1990	
			TBq ¹²	% of limit
British Nuclear Fuels plc				
Sellafield Sea pipelines ²	Total alpha	10	2.16	22
	Total beta	500	70.93	14
	Tritium	3500	1698.62	49
	Carbon-14	4	1.97	49
	Cobalt-60	8	0.17	2.1
	Strontium-90	35	4.22	12
	Zirconium-95+Niobium-95	180	6.82	3.8
	Technetium-99	10	3.82	38
	Ruthenium-106	170	16.54	9.7
	Iodine-129	0.4	0.11	28
	Caesium-134	10	1.15	12
	Caesium-137	110	23.46	21
	Cerium-144	22	2.01	9.1
	Plutonium-alpha	7	1.14	16
	Plutonium-241	170	31.61	19
	Americium-241	3	0.75	25
Seaburn sewer	Total activity	0.148	1.3 10 ⁻³	0.9
Springfields	Total alpha	13.32	0.2	1.5
	Total beta	444	92	21
Chapelcross	Total alpha	0.1	5 10 ⁻⁴	0.5
	Total beta ¹	25	0.11	<1
	Tritium	5.5	0.28	5.1
Capenhurst				
Rivacre Brook	Uranium	0.02	3 10 ⁻³	15
	Uranium daughters	0.02	0.01	50
	Non-uranic alpha	0.003	1.3 10 ⁻⁴	4.3
	Technetium-99	0.1	6.6 10 ⁻³	6.6
Meols outfall	Technetium-99	0.148	NIL	NIL
United Kingdom Atomic Energy Authority				
Winfrith	Tritium	650	39.2	6.0
	Cobalt-60	10	1.2	12
	Zinc-65	6	0.19	3.2
	Total alpha	0.3	4 10 ⁻³	1.3
	Other radionuclides	80	2.6	3.3
Harwell	Total activity ^{1,3}	8.88	0.17	1.9
	Tritium	8.88	1.0	12
Dounreay	Total alpha ⁴	0.75	0.022	2.9
	Total beta	110	4.3	3.9
	Tritium	130	0.3	<1
	Cobalt-60	1	0.023	2.3
	Strontium-90	12	1.3	11
	Zirconium-95+Niobium-95	6	0.01	<1
	Ruthenium-106	12	0.34	2.8
	Silver-110m	0.4	0.01	<2.5
	Caesium-137	50	2.2	4.4
	Cerium-144	12	0.038	<1
	Plutonium-241	15	0.72	4.8
Curium-242	1	0.019	1.9	
Nuclear Electric plc				
Berkeley	Total activity ¹	7.4	0.33	4.4
	Tritium	55.5	1.35	2.4
Bradwell	Total activity ¹	7.4	0.32	4.4
	Zinc-65	0.185	9.7 10 ⁻⁴	<1
	Tritium	55.5	1.4	2.5
Dungeness				
'A' Station	Total activity ¹	7.4	0.39	5.3
	Tritium	74	0.71	<1
'B' Station	Total activity ^{1,5}	4	9.1 10 ⁻³	<1
	Sulphur-35	25	0.05	<1
	Tritium	650	7.2	1.1
Hartlepool	Total activity ^{1,5}	4	0.02	<1
	Sulphur-35	7.5	0.92	12
	Tritium	1850	166	9
Heysham				
Station 1	Total activity ^{1,5}	4	0.058	1.4
	Sulphur-35	7.5	0.4	5.3
	Tritium	1850	157	8.5
Station 2	Tritium	1200	45.2	3.8
	Sulphur-35	7	0.073	1
	Cobalt-60	0.036	7 10 ⁻⁶	<1
	Other radionuclides	0.45	0.011	2.4

Table 1. Continued

Establishment	Radioactivity	Discharge limit (annual equivalent), TBq	Discharges during 1990	
			TBq ¹²	% of limit
Hinkley Point⁶				
'A' Station	Total activity ^{1,5}	7.4	0.39	6.2
	Sulphur-35	3.7	0.23	7.4
	Tritium	74	0.74	1.2
	Total activity ^{1,7}	1	0.053	-
	Caesium-137	1.5	0.084	-
	Tritium	25	0.17	-
'B' Station	Total activity ^{1,5}	3.7	0.03	< 1
	Sulphur-35	22.2	1.2	6.6
	Tritium	666	231	42
	Total activity ^{1,5,8}	0.25	7.5 10 ⁻³	18
	Sulphur-35	2.0	0.39	-
	Cobalt-60	0.035	2.6 10 ⁻⁴	-
Tritium	650	64	-	
Oldbury	Total activity ¹	3.7	0.43	12
	Tritium	74	1.7	2.3
Sizewell	Total activity ¹	7.4	0.43	5.8
	Tritium	111	5.0	4.5
Trawsfynydd	Total activity ¹	1.48	0.33	22
	Caesium-137	0.259	0.041	16
	Tritium	74	2.5	3.4
Wylfa	Total activity ¹	2.405	0.072	3.0
	Tritium	148	5.4	3.6
Scottish Nuclear Ltd				
Hunterston				
'A' Station	Total activity ¹	7.5	0.32	4.3
	Tritium	48	0.52	1.1
'B' Station	Total activity ^{1,5}	3.7	0.05	1.4
	Sulphur-35	26	2.5	9.6
	Tritium	1480	353	24
Tomess	Tritium	1200	82	6.8
	Sulphur-35	10	0.081	< 1
	Cobalt-60	0.05	2.9 10 ⁻⁵	< 1
	Beta activity ^{1,5,8}	0.45	1.8 10 ⁻³	< 1
	Total alpha	4.5 10 ⁻³	8 10 ⁻⁶	< 1
Ministry of Defence (Procurement Executive)				
Aldermaston	Total activity ^{1,3}	5.8	0.045	< 1
	Tritium	5.8	0.60	10
Ministry of Defence (Navy Department)				
Devonport ⁹	Total activity ^{1,8}	2 10 ⁻³	9 10 ⁻⁶	< 1
	Cobalt-60	0.016	1.4 10 ⁻³	8.8
	Tritium	0.12	0.049	41
Faslane	Total activity ¹	0.037	8.4 10 ⁻⁵	< 1
Rosyth ¹⁰	Beta activity ^{1,8}	0.01	2 10 ⁻⁴	2.0
	Cobalt-60	0.055	5 10 ⁻⁴	< 1
	Tritium	0.01	5.6 10 ⁻⁴	5.6
	Total alpha	1 x 10 ⁻⁶	6 x 10 ⁻⁷	60
Amersham International plc				
Amersham	Total activity ^{1,3}	2.7	1.07	40
	Tritium	14.8	0.032	< 1
Cardiff	Beta/gamma activity ¹¹	0.096	0.022	23
	Carbon-14	2	1.57	79
	Tritium	1400	609	44

¹ Excluding tritium

² Authorisation was varied with effect from 1 January 1990

³ Authorisation of agreement specifies a control formula in which the total effective activity is calculated to allow for the relative radiotoxicities of different nuclides. The sums of the actual discharges were lower than the values indicated

⁴ Excluding curium-242

⁵ Excluding sulphur-35

⁶ Authorisation was revised with effect from 1 November 1990. For each station the first block of data relates to the period 1 January to 31 October 1990; the second block of data relates to the period 1 November to 31 December 1990. '% of limit' refers to discharges during the first ten months of the year as a percentage of the equivalent limit for ten months of a year

⁷ Excluding caesium-137

⁸ Excluding cobalt-60

⁹ The operator of this site is Devonport Management Ltd

¹⁰ The operator of this site is Babcock Thorn Ltd

¹¹ Excluding tritium, carbon-14 and radioisotopes of calcium and strontium

¹² Some discharges are upper estimates because they include 'less than' data derived from analyses of effluents at limits of detection

contains the Windscale reprocessing plant and the Calder Hall power station and, prior to April 1971, was operated by the United Kingdom Atomic Energy Authority (UKAEA). The principal activities of the Sellafield Site which produce low-level radioactive waste discharges are the reprocessing of irradiated fuel and the conditioning and storage of nuclear materials and radioactive wastes. There is an adjacent site, the Windscale Laboratory operated by the UKAEA, which produces a minimal contribution to the discharges.

Low-level liquid wastes from the Sellafield Site are discharged, under authorisation, into the eastern Irish Sea by a pipeline which extends some 2.1 km seaward of the low water mark and ends about 20 m below the surface of the water. The discharges are subject to controls under the Radioactive Substances Act 1960 (Great Britain - Parliament, 1960) which requires that, in England, such discharges are authorised jointly by the Department of the Environment (DOE) (formerly by the Ministry of Housing and Local Government, which became part of the Department of the Environment in 1970) and the Ministry of Agriculture, Fisheries and Food (MAFF), after consultation with appropriate local authorities. The Authorising Departments apply a regime of controls which includes appreciation of the dose limitation philosophy of the International Commission on Radiological Protection (ICRP); this includes the concept of optimisation of practices to ensure that 'all exposures shall be kept as low as reasonably achievable ...' (ALARA), subject to compliance with appropriate dose limits.

The most important low-level radioactive wastes arise both in water used to purge the cooling ponds in which spent fuel elements are kept, and from the reprocessing plant whose low-level liquid wastes are collected and neutralised in 'sea tanks' before being discharged around high water.

2.2 Rates of discharge

Predictions about the dispersion and fate of low-level radioactive waste in the Irish Sea were made before disposal commenced. By 1952, sufficient wastes had accumulated to allow small-scale, experimental discharges to take place to test these initial assumptions (Fair and Maclean, 1956). Some 370 TBq in total of radioactivity was discharged in 1952 and, in 1953, the discharge was approximately 74 TBq a month (Howells, 1966). New plant was commissioned for the separation of irradiated uranium magnox fuel in 1964. This plant allowed greater decontamination of Pu and U from fission products to be obtained in one step by using TBP (tributyl phosphate) throughout, instead of a two-step process using Butex (dibutyl carbitol) and

TBP which had been used previously (Williams and Davidge, 1962). Details of the major radioactive components of the effluent have been available since 1960 with the most comprehensive analysis available from 1978 onwards. Annual discharge figures have been published, in recent years, by the Environment Departments (Department of the Environment, 1991; Scottish Development Department, 1990) and the operator (BNFL, 1979-1989). The quantities of radionuclides discharged have changed markedly with time (Figure 2). The quantities of some of the shorter-lived fission product nuclides in the effluent, such as ^{95}Zr , ^{144}Ce and ^{106}Ru , have steadily declined since the early 1970s. Discharges of ^{134}Cs and the longer-lived ^{137}Cs reached peak values from 1974 to 1978. This increase was due to a prolonged cessation of reprocessing in 1974 and increased residence time of magnox fuel in the storage ponds. This resulted in an increase in the corrosion of the cladding and a subsequent increase in the concentrations of soluble radionuclides in the cooling pond waters. Measures were taken to control corrosion rates by chemical treatment of the pond water and, in 1976, zeolite skips were introduced to reduce the ^{137}Cs concentrations as an interim measure (Handyside *et al.*, 1982) until a new effluent treatment plant (SIXEP: Site Ion-Exchange Plant) came into operation in 1985.

Discharges of the major transuranium nuclides have also fluctuated (Figure 2) with peak values of ^{241}Am from 1971 to 1975, and of ^{241}Pu from 1970 to 1980. The Pu-alpha discharges were greatest in 1973. Data on ^{239}Pu , separate from $^{239,240}\text{Pu}$, have been available from 1978, since which time the ratio of $^{239,240}\text{Pu}/^{238}\text{Pu}$ has varied from about 5.5 to 2.5 on a month-to-month basis (Pentreath *et al.*, 1984). However, analyses of surface sediment samples from Newbiggin, in the Esk Estuary, indicated that this ratio fell from about 20 in 1966 to about 5 or less after 1972 (Hetherington, 1978). This interpretation was supported by a recent study (Kershaw *et al.*, 1990(a)) of undisturbed sediment cores from Senhouse Dock, Maryport. A chronology was established by comparing a variety of radionuclide concentrations and isotope ratios in the core with the available information on decay-corrected discharges. This allowed an estimate to be made of the quantity of ^{238}Pu discharged in the period 1959 to 1977 (94 ± 8 TBq) for which no discharge data exist (Figure 3).

Discharges of Pu and Am are due to be reduced further from 1993 onwards, with the commissioning of the Enhanced Actinide Removal Plant (EARP). Conversely, the quantities of ^{99}Tc , ^{129}I , ^{60}Co and ^{14}C will increase as a result of both the processing of medium-level radioactive liquors (presently being stored on site) and the planned starting-up of the Thermal Oxide Reprocessing Plant (THORP).

2.3 Chemical forms of nuclides

In view of the many processes which occur on site, it is not easy to predict the chemical form of any specific radionuclide, nor to assume that this would remain constant over prolonged periods of time. Nevertheless, some generalisations can be made, particularly in view of the fact that the sea tank effluent is neutralised to pH 8 to 9 by the addition of ammonia solution prior to discharge, and contains some iron which would be precipitated out as ferric hydroxide.

The chemical nature of the effluent has not been studied extensively but some data exist for the transuranium nuclides (Pentreath *et al.*, 1984). Results obtained during 1982 are given in Table 2 and these are assumed to be typical of routine releases. Some 99% of the Pu (α), ^{241}Am and $^{243/244}\text{Cm}$ in the sea tanks, and about 60% of the ^{237}Np , were associated with particulate ($> 0.22 \mu\text{m}$) material. The percentage of Pu (α) in the pond water effluents retained by filtration was somewhat lower, and that of ^{237}Np was lower still. There were also differences in chemical speciation, with reduced (III + IV) Pu (α) predominating in the

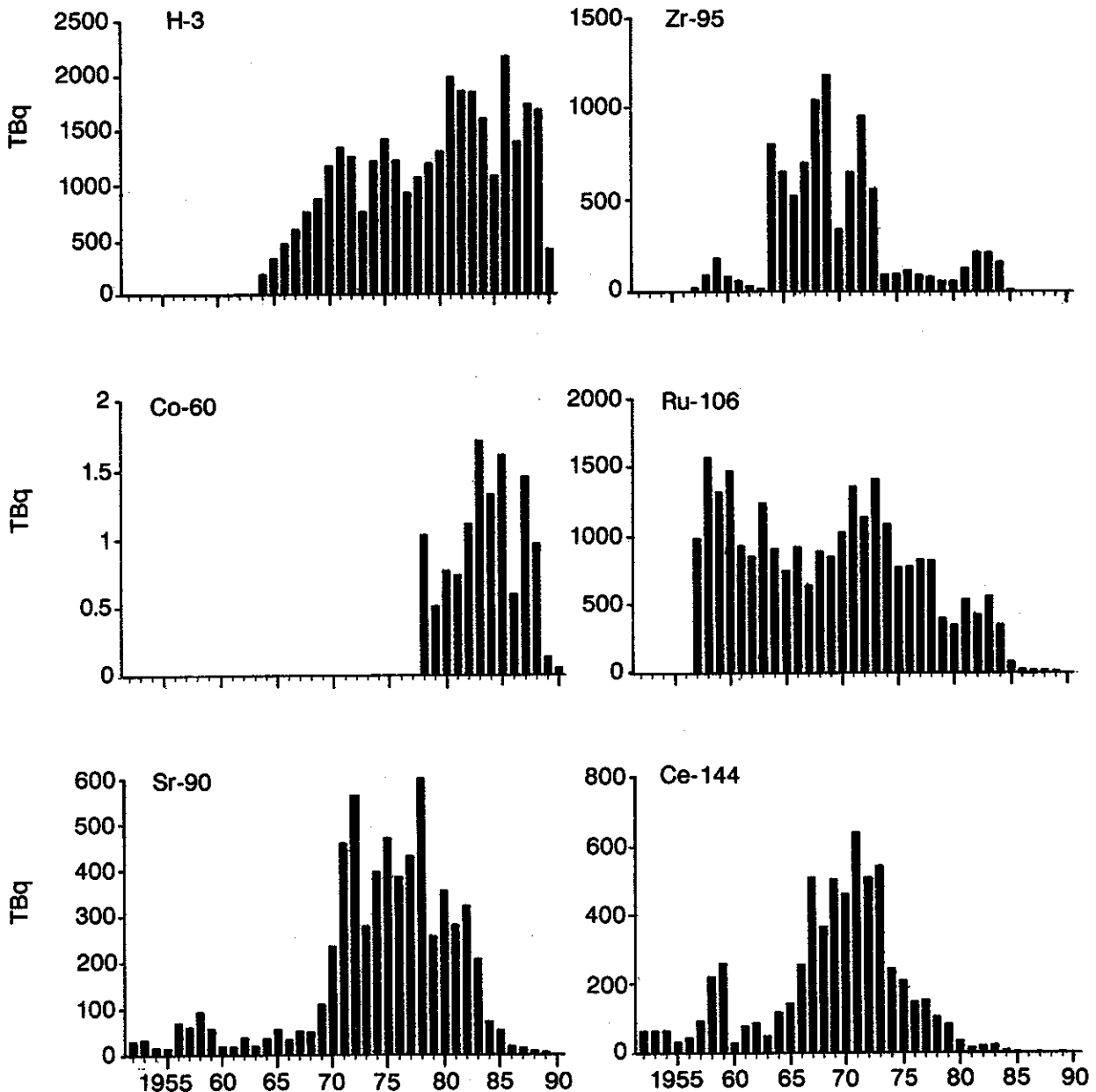


Figure 2. Annual discharges to sea from BNFL Sellafield

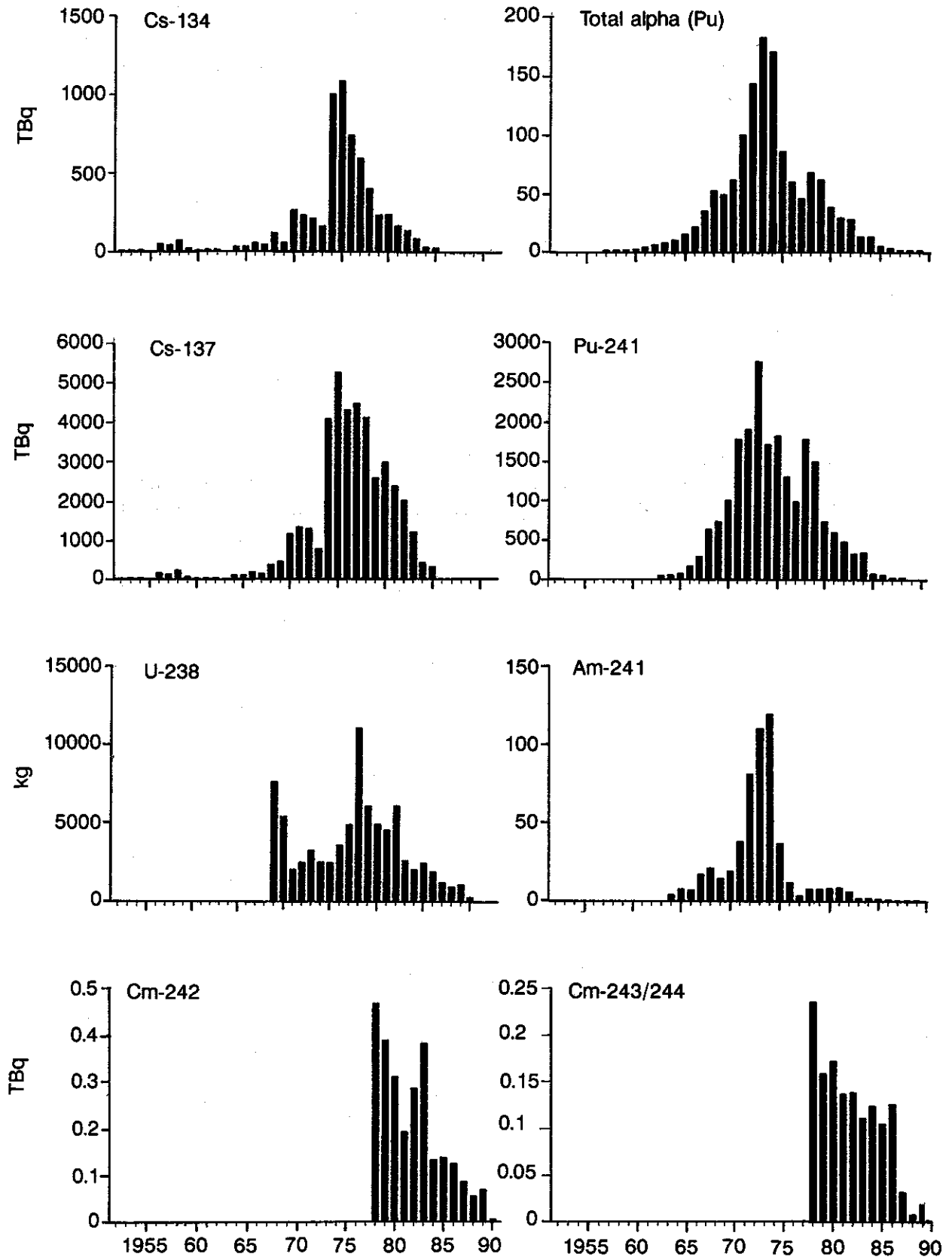


Figure 2 continued. Annual discharges to sea from BNFL Sellafield

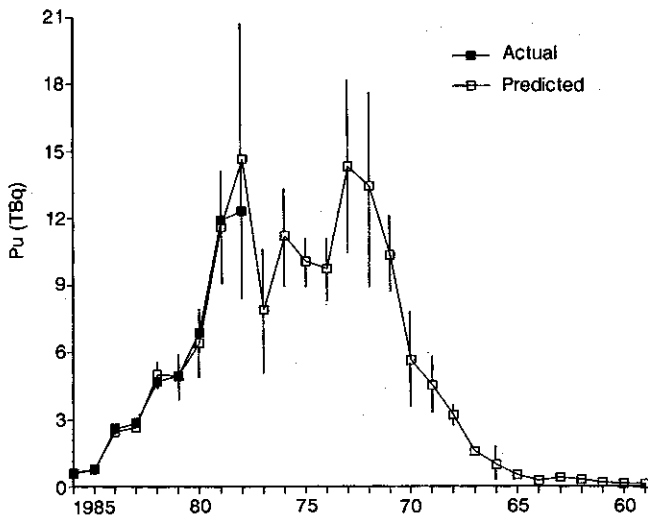


Figure 3. Annual discharge of ^{238}Pu (TBq year^{-1}) from BNFL Sellafield: reported values (closed squares) and estimated values (open squares) from a dated core (from: Kershaw *et al.*, 1990(a))

seawater effluent and oxidized (V + VI) Pu (α) predominating in the filtrate of the effluent from the cooling ponds. In view of the small contribution (< 10%) of these pond effluents to the total Pu (α) discharges, however, only about 1% of the total would appear to be in an oxidised form. In contrast, only 5% of the ^{237}Np in the filtrate was present in the reduced NP (IV) form so that, in total, about half of the combined ^{237}Np discharges are in the oxidised form. The nuclides of Am and Cm were present only in the Am (III) and Cm (III) forms.

The effluent also contains 'hot particles' (Pentreath *et al.*, 1984) identified as discrete clusters of α -tracks by CR-39 track-etching techniques (Hamilton and Clifton, 1981). Such particles, clearly having an effluent origin, are also evident in environmental samples (Hamilton, 1985; Hamilton *et al.*, 1991). Their contribution to the total quantities discharged is unknown, but the evidence of Hamilton and co-workers, working in the Esk Estuary and offshore, would suggest that significant numbers of such particles persist in the environment for several months, at least, before dissolving with some being preserved in rapidly accumulating estuarine sediments (Hamilton, 1981; Hamilton and Clifton, 1980). It has been estimated that about 10% of the Pu (α) in offshore surface sediments consists of hot-particles (Kershaw *et al.*, 1986(a)). Such calculations depend on the criteria used to define a 'hot particle'.

Table 2. Characteristics of transuranium nuclides in Sellafield liquid-effluent samples taken in 1982 (errors based on $\pm 1\sigma$ propagated counting errors^(a)) (from: Pentreath *et al.*, 1984)

Percentage	Sea tanks Filtrate	Particulate	Pond water Filtrate	Particulate
^{238}Pu	1.3 ± 0.3	98.7 ± 3.6	12.0 ± 1.3	88.0 ± 5.4
$^{239,240}\text{Pu}$	1.4 ± 0.1	98.7 ± 2.2	13.2 ± 0.6	86.8 ± 2.6
^{241}Am	1.6 ± 0.2	98.4 ± 3.9	2.0 ± 0.2	98.0 ± 4.2
$^{243,244}\text{Cm}$	0.0	100.0	4.1 ± 1.4	95.9 ± 15.9
^{237}Np	38.4 ± 3.6	61.6 ± 6.4	88.2 ± 47.1	11.8 ± 5.1
$^{239,240}\text{Pu}/^{238}\text{Pu}$	3.7 ± 1.0	3.5 ± 0.1	4.2 ± 0.4	3.8 ± 0.1
$^{239,240}\text{Pu}$ in each fraction as Pu (V/VI)	2.2 ± 7.3	1.4 ± 0.5	96.0 ± 3.4	1.7 ± 0.4
^{237}Np in each fraction as Np (IV)	4.9 ± 3.7	88.0 ± 8.0	0.0	0.0

^(a)Larger counting errors are due to short counting times

3. THE IRISH SEA

3.1 Physical oceanography

The Irish Sea is a semi-enclosed body of water, connecting with the Atlantic Ocean via St Georges Channel in the south and via the narrower North Channel, Clyde Sea and Malin Shelf Sea to the north. The area is effectively sub-divided into two principal regions, to the east and west of the Isle of Man. The western portion consists of a deep channel (>100 m) whereas depths in the eastern Irish Sea seldom exceed 50 m, and between the Isle of Man and Cumbria seldom exceed 30 m (Figure 4).

The physical oceanography of the Irish Sea has been quite extensively studied and modelled, the most recent reviews being those of Howarth (1984), Dickson (1987) and Dickson and Boelens (1988). Atlantic water enters the Irish Sea from both the North Channel and St Georges Channel (Figure 4), the latter predominating, and this is reflected in the annual mean surface isohalines (Bowden, 1980). The eastern Irish Sea's salinity is markedly affected by freshwater input, which considerably exceeds that of precipitation minus evaporation. The sea surface temperature has a large annual variation, with a minimum in February and a maximum in August. The annual range off the Cumbrian coast exceeds 10°C (~5°C to ~16°C) and weak stratification may develop in the eastern Irish Sea during the summer — except in Liverpool Bay, where the water column remains well-mixed because of the shallow depths and relatively strong tidal currents (Pingree and Griffiths, 1978). Some stratification can occur in the winter and spring, however, due to freshwater input (Jones and Folkard, 1971).

Tides dominate the dynamics of the Irish Sea, and these have been extensively studied and modelled (e.g. Mungall and Matthews, 1978). They are propagated in the Atlantic, with large spatial variations in the amplitude of the tidal currents. Low-frequency currents are created by meteorological conditions, spatial variations in water density, and non-linear tidal effects. The review by Howarth (1984) concluded that the water off Sellafield tended to oscillate, flowing southeastwards parallel to the coast during weak winds, and north-westwards during storms. It eventually leaves the eastern Irish Sea, via either the south or north of the Isle of Man, and thence through the North Channel.

A number of two-dimensional and three-dimensional hydrodynamic models of the Irish Sea have been developed (Hunter, 1972; Horwood, 1974; Horwood and Bedwell, 1978; Pingree and Griffiths, 1979; Heaps, 1973, 1974, 1979; Heaps and Jones, 1975, 1977, 1979; Davies and Jones, 1992). In addition, several box models have been developed specifically to describe the distribution of radionuclides discharged by Sellafield into the Irish Sea and beyond (Camplin *et al.*, 1982; Hallstadius *et al.*, 1987; Gurbutt *et al.*, 1988; Jefferies and Steele, 1989; Gurbutt and Kershaw, 1989; Howarth, 1989; section 9).

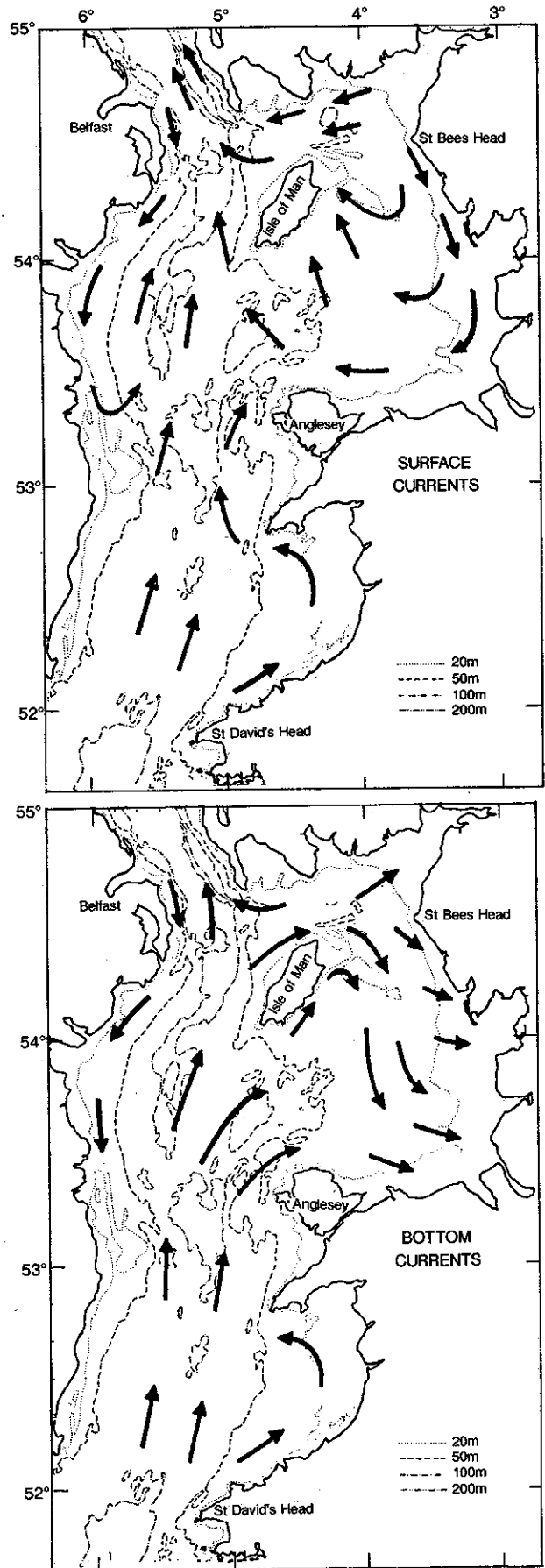


Figure 4. Irish Sea bathymetry, showing surface and bottom mean residual currents (from: Dickson, 1987)

3.2 Sediment distributions

The geological structure of the Irish Sea has been studied in some detail (Wright *et al.*, 1971; Mitchell, 1972; Bott, 1968; Dobson, 1977(a) and (b)). The area consists of several sedimentary basins, formed by faulting, containing Carboniferous and Permo-Triassic rocks (Smith *et al.*, 1980; Williams *et al.*, 1981) which were deeply eroded by successive glacial advances. These have been overlain by tills and boulder clays.

The pattern of sedimentation in the northern Irish Sea during the Flandrian (Holocene) period from 10,000 years B.P. (Before Present) to the present day has been the subject of much discussion, with particular attention being paid to the fate of the finer-grained sediments (Belderson, 1964; Belderson and Stride, 1969; Cronan, 1969; Pantin, 1977, 1978, 1991; Mauchline, 1980; Williams *et al.*, 1981; Stride, 1982; Johnson, 1983; Kirby *et al.*, 1983; Kershaw, 1986, Kershaw *et al.*, 1988(a)). The most comprehensive accounts of the distribution and origin of Quaternary sediments in the Irish Sea are given by Pantin (1977, 1978) with Dobson (1977(a)(b)) providing a summary of the underlying geological structure.

Muddy sediments are confined to two main areas: a belt of muds and muddy sands parallel to the Cumbrian coast extending into Liverpool Bay in the south and across the mouth of the Solway Firth to Wigtown Bay in the north, and a relatively deep (~100 m) basin lying between the Isle of Man and the coast of Ireland (Figure 5). Both areas coincide with zones of relatively weak tidal currents (Belderson, 1964; Belderson and Stride, 1969; Howarth, 1984). The strongest tidal currents occur north and south of the Isle of Man and are associated with coarser sands and gravels. Finer grained material has been winnowed from these areas (Eden *et al.*, 1973) and there is evidence, from observations of sand waves, of an easterly movement of sand towards the eastern mud belt (Belderson and Stride, 1969; Stride, 1982). The transition from coarse sands to fine-grained muds is quite abrupt, compared with similar facies changes in the bottom sediments to the west of the Netherlands in the southern North Sea (Stride, 1982).

The hydrological and sedimentological evidence (Belderson, 1964; Belderson and Stride, 1969; Cronan, 1969; Pantin, 1977, 1978, 1991; Mauchline, 1980) suggest that both areas of muddy sediments are zones of active sedimentation. This hypothesis was challenged in a review by Kirby *et al.* (1983) who concluded that much of the evidence to support the existence of present-day accretion was circumstantial and they doubted whether there were sufficient sources of fine sediment to support continued sedimentation. There is limited evidence, from ¹⁴C dating, of 'recent' net sedimentation in the NE Irish Sea (within the past few hundred years) but at a low rate of accumulation (~0.1 mm year⁻¹, Kershaw *et al.*, 1988(a)).

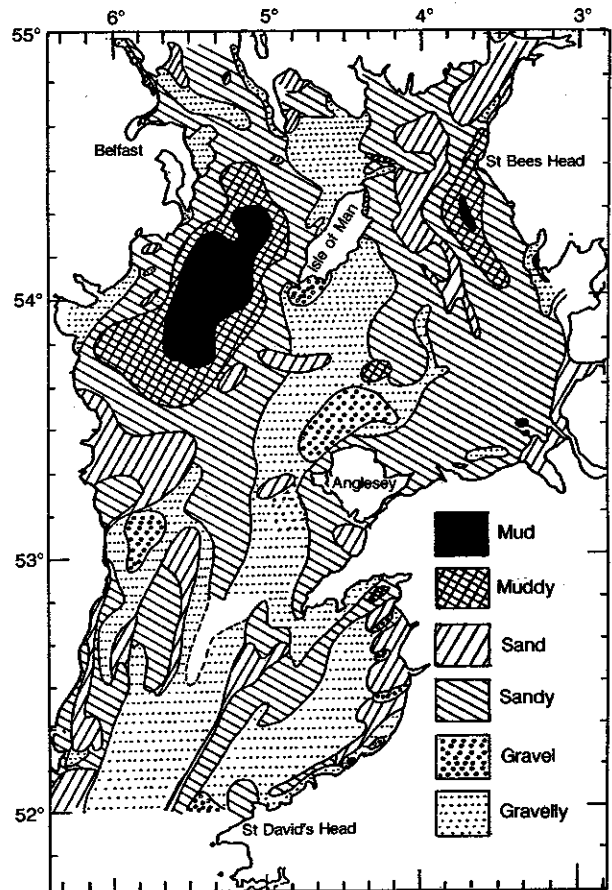


Figure 5. Distribution of fine-grained sediments in the Irish Sea

3.3 Fisheries

The Irish Sea supports a varied commercial fishery, the history of which has been reviewed by Brander (1977). The quantities of fish landed, and the proportions taken by different countries, are published annually by the International Council for the Exploration of the Sea (ICES), from which the data in Table 3 have been obtained (ICES, 1990). As can be seen, the major fish landed, in terms of weight, are herring, whiting and cod for the area as a whole. There are marked spatial variations, however, but the principal fish species caught in the Sellafield discharge area is the plaice, which also constitutes the major species landed at the nearest fishing port, Whitehaven.

Aspects of the radioecology of the area around Sellafield have been presented in a large number of publications (see section 6). The first general survey and overall evaluation was reported by Mauchline (1963) and Mauchline and Templeton (1963).

Table 3. Weight (t) of main demersal, pelagic and shellfish species taken in the Irish Sea by all countries in 1987 (from: ICES, 1990)^(a)

Catch	Tonnes
Demersal species	
Brill	252
Dab	411
Flounder	183
Lemon sole	314
Megrim	289
Plaice	5572
Sole	2041
Turbut	273
Cod	13183
Haddock	1287
Hake	1318
Ling	288
Pollack	655
Saithe	1526
Whiting	11684
Conger eel	214
Gurnards	199
Monk	1182
Picked dogfish	7112
Dogfish + hounds	888
Rays + skates	5332
Pelagic species	
Horse mackerel	27
Herring	11177
Sprat	41
Mackerel	181
Shellfish	
Edible crab	896
Crawfish	4
Lobster	80
Norway lobster	10306
Crangonid shrimps	483
Whelks	450
Periwinkles	349
Oysters	51
Mussel	5471
Escallop	2572
Queen scallop	9857
Cockles	18459
Squids	169

^(a)Scientific names, where appropriate, are given in the reference

4. BEHAVIOUR OF RADIONUCLIDES IN SEA WATER

4.1 Fission and neutron-activation products

Radionuclides of Cs (¹³⁴Cs and ¹³⁷Cs) have been studied in considerable detail. During the period 1963 to 1966, observations were made off the Cumberland coast (Preston *et al.*, 1971), with samples being taken quarterly from a network of up to 25 stations covering an area of 10³ km². These data showed that the ¹³⁷Cs concentration isopleths close to the shore were elliptical and parallel to the coast, with their centres displaced approximately 1.5 km in a southerly direction; further offshore the contours were displaced to the

north. Since 1968, the inventories of ¹³⁷Cs in filtered sea water have been examined over much larger areas of the Irish Sea. The distributions in 1968 and 1969 were reported by Preston *et al.* (1972), and all of the earlier data were reviewed by Jefferies *et al.* (1973).

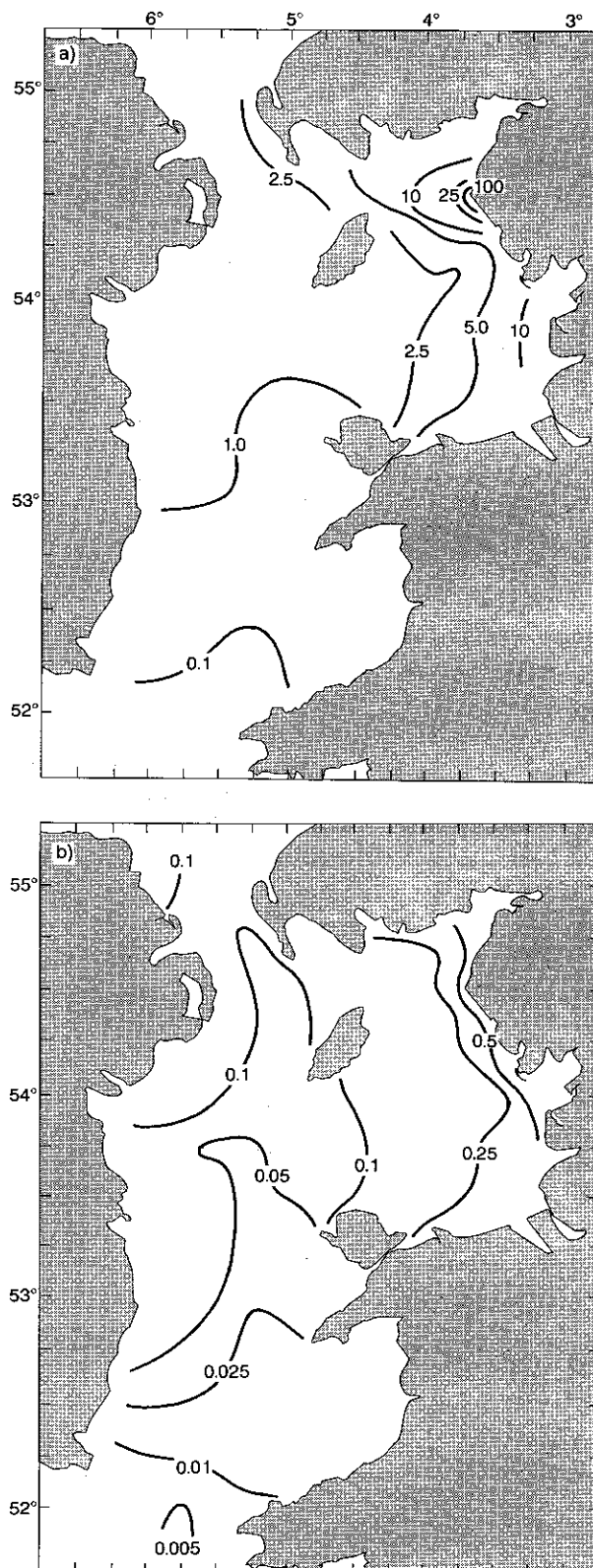


Figure 6. Distribution of ¹³⁷Cs (Bq kg⁻¹) in filtered sea water in the Irish Sea for the years (a) 1977; and (b) 1988 (from: Hunt, 1979, 1989)

The detailed studies of Jefferies *et al.* (1982) for the period 1970 to 1978 showed that a release rate of 1 Ci day^{-1} (37 GBq day^{-1}) resulted in a mean annual concentration of 4.2 pCi l^{-1} (155 m Bq l^{-1}) in the immediate vicinity of the discharge point, with an approximate 30-fold reduction in the North Channel, and at least a 300-fold reduction in the southern entrance. The residence half-times of water in the Irish Sea, based on the Cs data, were estimated as being of the order of 1 year between 1970 and 1976, but considerably less than 1 year for the period from 1976 to May 1978. The cause of this apparent change is unknown.

In recent years, the distribution of ^{137}Cs in the Irish Sea and certain adjacent waters has been determined annually and is illustrated in MAFF's Aquatic Environment Monitoring Report Series (Hunt, 1979-1985(a), 1976-1989; MAFF, 1990, 1992, in press). The very large quantities of Cs data, covering a period of several decades, have been compiled into a coherent data set which is being published in MAFF's Data Report Series (Camplin and Steele, 1991; Baxter, 1990; Baxter and Camplin, in press (a), (b), (c); Baxter *et al.*, 1992). The considerable reduction in Cs discharges since the mid-1970s (Figure 2) resulted in a decrease in water concentrations, which can be demonstrated by comparing the distribution in 1977 with that in 1988 (Figure 6). There is some field evidence that nearshore ^{137}Cs water concentrations, since the early 1980s, have not reduced as quickly as the reduction in the discharge would have implied (Table 4) (Hunt and Kershaw, 1990). This suggests that re-mobilisation of ^{137}Cs from the subtidal sediments is occurring as the system re-equilibrates - a hypothesis supported by model calculations and some limited experimental work (Kershaw *et al.*, 1990(b)) and a study of the evolution of the $^{137}\text{Cs}/^{241}\text{Am}$ ratio in surface sediments since 1978 (McCartney *et al.*, in press).

The overall decline in Cs concentrations in the Irish Sea and other UK waters was interrupted in April 1986 following the Chernobyl reactor accident. The signal

could be detected both as an overall increase in radionuclide concentrations of ^{134}Cs , ^{137}Cs , ^{110}Ag , ^{131}I , $^{129\text{m}}\text{Te}$ and ^{103}Ru , and a significant increase in the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio. However, levels in nearshore waters, as well as in seaweed and molluscs, decreased rapidly in the Irish Sea (Camplin *et al.*, 1986) (Figure 7). The effects were more marked and longer lasting in the North Sea, with run-off via the Baltic Sea masking the previous distribution pattern based on the Sellafield discharges (Mitchell and Steele, 1988; Nies, 1988).

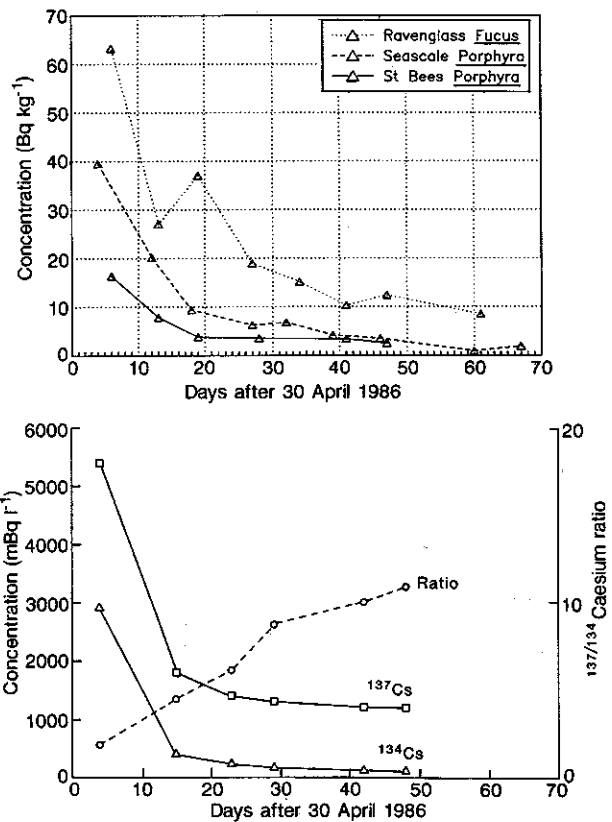


Figure 7. Concentration of ^{134}Cs and ^{137}Cs (Bq kg^{-1}) in shoreline water at Southerness during 1986, showing the declining influence of contamination from the Chernobyl reactor accident (from: Camplin *et al.*, 1986)

Table 4. Remobilisation of ^{137}Cs from the bed of the Irish Sea (from: Hunt and Kershaw, 1990)

Year	Measured inventory in sea water ^a (TBq)	Inventory without remobilisation ^b (TBq)	Remobilised activity ^a (TBq)
1983	1600 ± 500	1450	150 ± 500
1984	1100 ± 400	680	420 ± 350
1985	680 ± 230	420	260 ± 230
1986	420 ± 140	110	250 ^c ± 140
1987	290 ± 100	36	220 ^c ± 100
		Total	1300 ^c ± 700

^aErrors represent ±1 standard deviation based on propagated sampling uncertainties

^bBased on procedures described in Hunt, 1985(b)

^cAfter subtraction of about 100 TBq, apportioned between 1986 and 1987 to allow for input from Chernobyl

This labelling of sea water by Cs radionuclides from Sellafield has provided excellent opportunities to study the hydrography of the area as a whole and of adjacent waters (Wilson, 1974; Baxter *et al.*, 1979; Livingstone *et al.*, 1982; Prandle, 1984; Nies, 1988, 1990; Kautsky, 1988, 1989; Bradley *et al.*, 1988). The waters leaving the Irish Sea pass through the Clyde Sea, and the rate of exchange of the water, using ^{137}Cs data, has been estimated to be approximately 4.5 months (Baxter *et al.*, 1979). From the Clyde Sea, ^{137}Cs can be traced along the western Scottish coast and eastwards into the northern North Sea (Bradley *et al.*, 1988). A number of studies have been made of the distributions of ^{137}Cs (Figure 8) and of ^{90}Sr and other radionuclides, in the North Sea (Hunt, 1989; Kautsky and Murray, 1981; Kautsky, 1988; Nies, 1988, 1990). Studies have also been made on the concentrations in the Baltic Sea (Kautsky and Eicke, 1982(a)), and on the extent of contamination along the Norwegian coast (Kautsky, 1989) and in the north-eastern Atlantic (Kautsky, 1989).

Other fission and activation products have not been studied to the same extent as ^{137}Cs with respect to their behaviour in sea water. Hetherington and Jefferies (1974) concluded that ^{106}Ru , ^{144}Ce , ^{95}Zr , ^{95}Nb were more rapidly removed to the sediments than Cs. They also concluded that out of the total inventory of each radionuclide in the immediate vicinity of Sellafield, approximately 60% of the ^{137}Cs and more than 95% of

the ^{106}Ru , ^{144}Ce , ^{95}Zr and ^{95}Nb were retained in the seabed compartment. The uncertainties in such calculations are considerable. The significant differences in half-life make it difficult to directly compare the behaviour of these radionuclides.

^{99}Tc , present as pertechnetate (TcO_4^-) in aerobic conditions (Sparkes and Long, 1988), behaves conservatively in sea water with a distribution coefficient (Kd - radionuclide concentration per unit mass particulate (Bq kg^{-1})/radionuclide concentration per unit volume sea water (Bq l^{-1})) of <10 (cf. Cs, Kd $\sim 10^3$; Pu, Kd $\sim 10^5$). This should make it an ideal tracer of water movements, although relatively few studies have been reported (Pentreath *et al.*, 1980). ^{99}Tc from Cap de La Hague is currently being used to investigate water movements in the English Channel and the southern North Sea in an international collaborative programme under the EC Marine Science and Technology Programme.

The likely increased discharge of ^{99}Tc from Sellafield, resulting from the commissioning of the EARP and THORP plants (see section 2), from 1993 onwards will provide an opportunity to use this tracer to study water movements, and validate transport models, within the Irish Sea, and exchange in the North Channel, Malin Shelf and along the Scottish coastal current.

^{14}C discharged from Sellafield, with a similar distribution pattern to ^{137}Cs , has caused an approximate doubling of the current ambient concentrations in the eastern Irish Sea (Begg *et al.*, 1991). There are four main carbon reservoirs in the marine environment: dissolved inorganic and organic (DIC and DOC); and, particulate inorganic and organic (PIC and POC). It is the DIC which has been used most frequently in tracer studies.

The distribution of the most soluble radionuclide, ^3H , has been studied in detail, particularly during the period 1968 to 1974 (Hetherington and Robson, 1979). It has been shown that, for a release rate of 1 Ci day^{-1} (37 GBq day^{-1}), the water concentration in the immediate vicinity of the discharge point is about 5 pCi l^{-1} (185 mBq l^{-1}). At greater distances, however, such normalised interpretations of the data have indicated both southerly and northerly dispersion.

4.2 Transuranium radionuclides

A considerable effort has also been expended on studying the behaviour of the transuranium nuclides. It has been shown (Nelson and Lovett, 1978; Lovett and Nelson, 1981; Pentreath *et al.*, 1985) that $^{239,240}\text{Pu}$ exists in filtered Irish Sea water in a mixture of oxidation states. More than 90% of the $^{239,240}\text{Pu}$ in the filtrate exists in the oxidised form, as determined by selective precipitation, and co-precipitation with hydrated

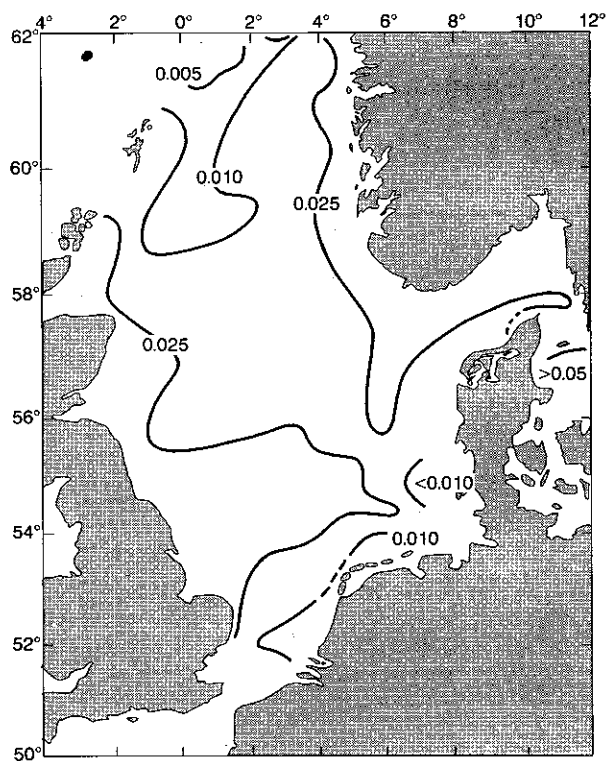


Figure 8. Distribution of ^{137}Cs (Bq kg^{-1}) in filtered sea water in the North Sea for the year 1988 (from: Hunt, 1989)

silicon dioxide (Nelson and Orlandini, 1979) suggests that Pu (V) predominates. The precise nature of ^{241}Am is unclear but it is thought to exist almost entirely as Am (III). The ^{237}Np , away from the vicinity of the discharge area, is considered to exist in solution as Np (V) (Pentreath and Harvey, 1981).

The concentrations of $^{239,240}\text{Pu}$ and ^{241}Am in Irish Sea water differ from year to year. Data for filtrate surface water in 1979 are shown in Figure 9 (Pentreath *et al.*, 1984). The overall pattern of distribution was not dissimilar from that of previous years (Hetherington *et al.*, 1975). In 1985, when the next major survey was undertaken, concentrations of both radionuclides in the coastal region had been reduced by about a factor of three. This was attributed to the decrease in discharges (Figure 2).

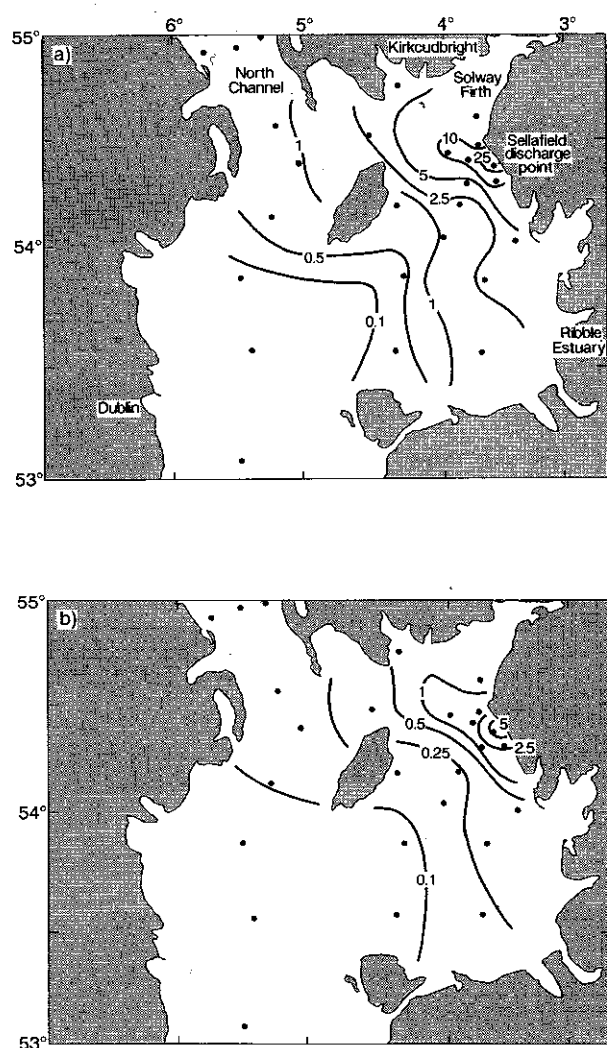


Figure 9. Distribution of (a) $^{239,240}\text{Pu}$ and (b) ^{241}Am (mBq l^{-1}) in filtered sea water in the Irish Sea for the year 1979 (from: Pentreath *et al.*, 1984)

There has been less agreement on the results from shoreline sampling. Hunt (1985(b)) reported a slight decrease at St Bees over the period 1980 to 1983 in the Pu (α) and ^{241}Am in filtrate and particulate. However, McKay and Pattenden (1989) presented data from a larger number of sites along the Cumbrian coast over the same period which showed marked variability but no consistent trend in concentrations. The particulate fraction contains a much higher proportion of the total activity than the filtrate, in marked contrast to waters further offshore. Suspended loads in these inshore waters are highly variable, spatially and temporally, and are generally significantly higher than loads offshore ($\sim 100 \text{ mg l}^{-1}$, as against $1\text{--}10 \text{ mg l}^{-1}$; McKay and Pattenden, 1989). The transuranium nuclides are dispersed throughout the Irish Sea including Irish waters (Mitchell, *et al.*, 1987; 1988; 1991; Sanchez-Cabeza, 1989) and are also transported into the North Sea with their distributions in total sea water samples being described in a number of papers (Murray *et al.*, 1978, 1979; Kautsky and Murray, 1981; Kautsky and Eicke, 1982(b)).

5. ASSOCIATION WITH SEDIMENTS

5.1 Suspended particulates

Suspended loads in the Irish Sea typically lie within the range of 1 to 10 mg l^{-1} . Samples of sea water taken by MAFF and others for radiochemical analysis are routinely filtered (0.22 or $0.45 \mu\text{m}$), and this immediately gives an indication of the extent to which different nuclides are adsorbed to suspended particulates. Direct estimates of distribution coefficients have been obtained for the transuranium nuclides (Pentreath *et al.*, 1984; Kershaw *et al.*, 1986(b)), and some of these are given in Table 5. The order is that of $\text{Am} > \text{Cm} > \text{Pu} > \text{Np}$, although the Pu value will certainly be that for a mixture of oxidation states, it having been shown that the Pu (III + IV) Kd values are of the order of 10^6 whereas those for Pu (V + VI) are less than 10^4 (Nelson and Lovett, 1978). The extent to which any change in oxidation state with distance (or time) affects Kd values is not entirely clear, an effect which may apply to Np as well as to Pu (Pentreath and Harvey, 1981).

The particulate/filtrate cut-off at 0.22 or $0.45 \mu\text{m}$ is operationally-defined. Recent studies have begun to investigate colloidal associations in the filtrate fraction using a variety of ultra-filtration techniques (K. S. Leonard, pers. comm.) and aluminium oxide beds (P. I. Mitchell, pers. comm.) with a view to identifying the physical and chemical (organic/inorganic) controls on radionuclide behaviour within these size ranges (≥ 1000 Daltons).

Table 5. *Kd* values obtained from sea water samples taken within the Irish Sea: \bar{x} = mean, s_x = standard error, n = number of samples (from: Pentreath *et al.*, 1984)

Year		Radionuclide			
		^{239,240} Pu	²⁴¹ Am	^{243,244} Cm	²³⁷ Np
1977	\bar{x}	3.1×10^5	2.4×10^6	-	-
	s_x	0.8×10^5	0.4×10^6	-	-
	n	15	15	-	-
1978	\bar{x}	2.8×10^5	2.2×10^6	1.5×10^6	-
	s_x	0.4×10^5	0.3×10^6	0.5×10^6	-
	n	21	21	6	-
1979	\bar{x}	3.7×10^5	1.9×10^6	1.6×10^6	-
	s_x	0.4×10^5	0.2×10^6	0.4×10^6	-
	n	28	28	8	-
1980	\bar{x}	-	-	-	1.1×10^4
	s_x	-	-	-	0.4×10^4
	n	-	-	-	8

Kd values derived from pCi kg⁻¹ dry weight suspended load/pCi l⁻¹ of sea water, filtered through a 0.22 µm filter

The resuspension of sediment by tides and waves and the subsequent transport in suspension appear to be significant factors influencing the northwards migration of particle-reactive radionuclides to the Solway coast (MacKenzie *et al.*, 1987) and the overall distribution of radionuclides in surface sediments (McCartney *et al.*, in press; Jones *et al.*, 1984).

Information on the controls of scavenging by suspended particulate material, and sea-bed sediments, has been obtained by studying the disequilibria in the distribution of the naturally-occurring radionuclide ²³⁴Th (particle-reactive, $K_d \sim 2 \times 10^6$; $t_{1/2} = 24$ days) and its parent ²³⁸U (conservative in sea water, $K_d \sim 1 \times 10^3$; $t_{1/2} = 4.5 \times 10^9$ years) (Kershaw and Young, 1988; Kershaw *et al.*, 1988(c)). This has demonstrated the importance of water depth, suspended load, water column stratification, tide- and wave-induced resuspension, and sediment grain size on the scavenging of dissolved forms and their incorporation into the sea bed. A combination of these factors leads to the enhanced scavenging of ²³⁴Th near the Cumbrian coast, providing a partial explanation of the relatively high inventories of Pu and Am observed to the north of the Sellafield outfall (sub-section 5.2).

In the course of this study, it became apparent that there are additional sources of naturally-occurring radionuclides in this region. The principal additions appear to be related to the discharges from a phosphogypsum plant at Whitehaven (McCartney *et al.*, 1990; section 7).

5.2 Sea-bed sediments and environmental inventories

The sea-bed sediments of the north-eastern Irish Sea represent a significant temporary sink for many of the radionuclides discharge by Sellafield. Radionuclide distributions are strongly influenced by distance from the source, the time dependent variations in the quantities discharged and the nature of the sediments.

Derived K_d values reflect sediment type (Hetherington and Jefferies, 1974). Data derived from analyses of offshore surface mud for Pu and Am are given in Table 6 (Pentreath *et al.*, 1980). Values for ⁹⁵Zr/⁹⁵Nb, ¹⁰⁶Ru and ¹³⁷Cs, similarly derived for both silt and sand at a shoreline site, are given in Table 7 (Jefferies, 1968). The distribution of radionuclides with depth in the sediments in offshore areas, and 'half-depth' values have been calculated for ⁹⁵Zr/⁹⁵Nb, ¹⁴⁴Ce, ¹⁰⁶Ru, ¹³⁴Cs and ¹³⁷Cs (Hetherington and Jefferies, 1974). Inventories for these nuclides in the immediate offshore area have been calculated from estimates of the 'half-depths', K_d values, and water- volume-to-sediment ratios. Over 95% of the ⁹⁵Zr/⁹⁵Nb, ¹⁰⁶Ru and ¹⁴⁴Ce were considered to reside in the sea bed and, in view of their short half-lives, their potential mobility offshore has not received detailed attention. For the longer-lived transuranium nuclides, however, an understanding of their long-term behaviour in the sea bed is necessary in order to make estimates of collective dose commitments (section 8).

Table 6. K_d for sediment from the surface of muddy areas of the Irish Sea; \bar{x} = mean; s_x = standard error of the mean; n = number of samples (from: Pentreath *et al.*, 1980)

Time of collection		Radionuclide	
		$^{239,240}\text{Pu}$	^{241}Am
January 1976	\bar{x}	1.4×10^5	1.3×10^6
	s_x	0.4×10^5	0.2×10^6
	n	7	6
September 1977	\bar{x}	1.3×10^5	1.4×10^6
	s_x	0.6×10^5	0.5×10^6
	n	5	5

K_d values derived from Bq kg⁻¹ dry weight/Bq l⁻¹ of 0.22 μm filtered sea water

In view of the K_d values indicated in Table 6, it would be expected that the majority of Pu and Am discharged would be associated with the offshore sea bed, and estimates of this fraction were initially made on the basis of sea-bed core data (Hetherington *et al.*, 1975, 1976; Hetherington, 1976(a)). The most readily made calculations, however, in view of the complex nature of the sea bed and the relationship between concentration and particle size (Hetherington, 1976(a)), are those which estimate the rates of loss of the radionuclides from the Irish Sea. Estimated total inventories of $^{239,240}\text{Pu}$ in the northern Irish Sea (north of 53°20'N) for the years 1973 to 1979 are given in Table 8, based on the total in the filtrate and the average percentile values for filtrate as a fraction of the total in the sea water (Pentreath *et al.*, 1984). If it is assumed that this body of water resides in the area with a half-time of about one year, then the quantity leaving per year can

Table 7. Estimates of fission product K_d values (from: Jefferies, 1968)

Radionuclide	Surface silt	Surface sand
$^{95}\text{Zr}/^{95}\text{Nb}$	1.5×10^4	5.0×10^2
^{106}Ru	1.4×10^4	4.0×10^2
^{137}Cs	0.8×10^3	0.6×10^2

be estimated and compared with the quantities discharged in the preceding twelve months. There is a considerable range in the values estimated in this way, and the possible effect of suspended load can also be seen.

Table 8. Estimated inventories of $^{239,240}\text{Pu}$ in the waters of the northern Irish Sea and the calculated rates of loss from the area relative to the discharge (from: Pentreath *et al.*, 1984)

Cruise date	Filtrate inventory (<0.22 μm) (TBq)	% $^{239,240}\text{Pu}$ in filtrate (mean value)	Total inventory (TBq) months' discharge	Loss through North Channel as % of previous 12
July 1973	2.18	57	3.82	4
July 1974	1.33	62	2.15	4
July 1975	2.55	52	4.90	9
January 1976	2.48	30	8.27	17
Sept. 1977	1.59	56	2.84	6
May 1978	1.63	54	3.02	6
April 1979	1.26	55	2.29	3

A more direct method of estimating the rate of loss is that of taking the concentration of $^{239,240}\text{Pu}$ in surface waters of the North Channel and, assuming a flow rate out of the channel of 5 km^3 per day, estimating the annual loss compared with the annual discharge rate (Pentreath *et al.*, 1984). Unfortunately, the extent to which surface water is representative of the total water column in the channel is not clear; limited data available suggest that calculations made on this basis may overestimate the rate of loss. Nevertheless, both methods give similar estimates, which indicate that some 3 to 17% of each year's discharge (average 7%) will be removed from the Irish Sea. A third method, by relating the concentrations relative to ^{137}Cs , whose fluctuations in the area have been more closely studied, gives similar results (Pentreath *et al.*, 1984). In view of the similar K_d values for ^{241}Am (and for isotopes of Cm), it may be assumed that the results of these approximate calculations also apply to those radionuclides. The distributions of $^{239,240}\text{Pu}$ and ^{241}Am within the sea bed reflect the distribution of fine sediments. But there are other factors to take into account, including the practice of discharging from the sea tanks on or about high water with the tide ebbing to the north. The initial distributions will also depend on the local water movements, which are known to be wind-driven and variable in the immediate vicinity of the pipeline, and on the suspended load. The total quantities of $^{239,240}\text{Pu}$ and ^{241}Am estimated to be lying within the upper 30 cm of the sea bed, as of 1977/1978, from analysis of sediment cores, are shown in Figure 10. Isopleth intervals have been chosen assuming an exponential decrease in concentration with distance. The precise depth of cores at different sites influences the results, as do their locations relative to changes in the nature of the sea bed. The data are therefore no more than an approximation, but they do indicate that at least 240 TBq of $^{239,240}\text{Pu}$ and 290 TBq of ^{241}Am were associated with the sea bed within a coastal strip approximately 30 km wide, running from Kirkcudbright Bay in the north to the Ribble estuary in the south. The quantities outside this zone amounted to about 40 TBq of $^{239,240}\text{Pu}$ and 50 TBq of ^{241}Am . Approximately 20% of the ^{241}Am was estimated to have arisen from *in situ* decay of ^{241}Pu .

The detailed distribution of γ -emitting radionuclides in sub-tidal sediments has been described by Miller *et al.* (1982) and Jones *et al.*, (1988). The distributions of ^{137}Cs , ^{106}Ru and $^{95}\text{Zr} + ^{95}\text{Nb}$ were similar, with contours of equal activity running parallel to the coast, displaced north of the outfall (Figure 11). The authors concluded that the pattern was largely a response to the transport of particle-associated effluent, an argument which is supported by other studies using isotope ratios (Mackenzie *et al.*, 1987; Baxter *et al.*, 1987). A recent re-interpretation of existing, extensive, DFR data sets on radionuclide concentrations in surface sediments, from samples collected in the period 1968 to 1988 has confirmed the existence of the northerly footprint

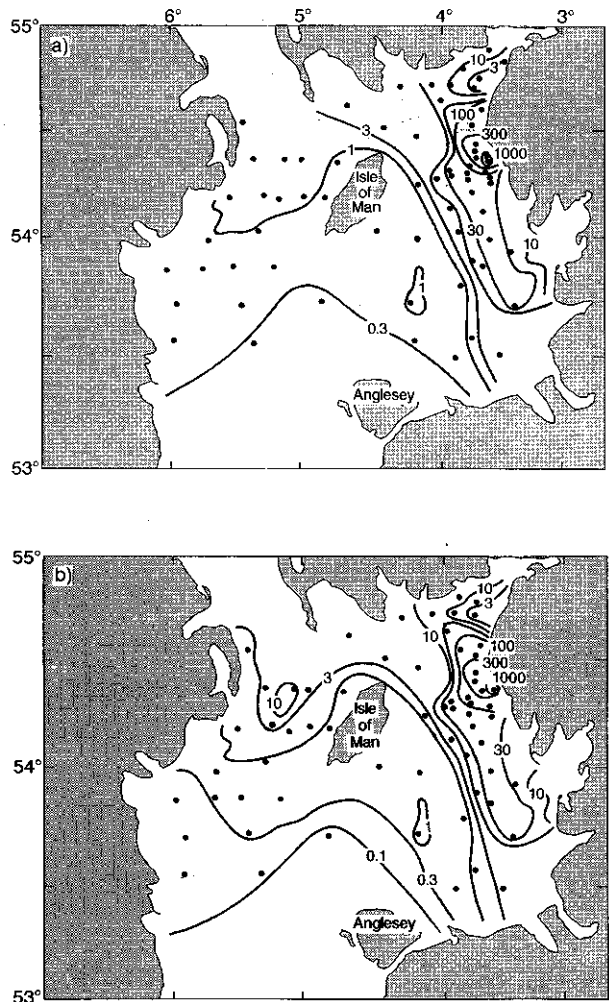


Figure 10. Distribution of (a) $^{239,240}\text{Pu}$ and (b) ^{241}Am (kBq m^{-2}) in sea-bed sediments in the Irish Sea for the years 1977/1978 (from: Pentreath *et al.*, 1984)

(McCartney *et al.*, in press). It is seen most obviously when the effects of grain-size on radionuclide concentrations are diminished by plotting isotope ratios, such as $^{137}\text{Cs}/^{241}\text{Am}$ or $^{241}\text{Am}/^{40}\text{K}$. Recent hydrodynamic modelling has indicated that persistent northerly flows of bottom water are set up parallel to the Cumbrian coast during strong northerly and westerly gales, because of topographic constraints on the wind-driven flow (Davies and Jones, 1992). Westerly gales will also result in the greatest wave-induced resuspension. It is evident that a mechanism exists which could result in the episodic movement northwards of particle-associated radionuclides, away from the vicinity of Sellafield. Model developments are being linked to a major field programme addressing the problem of cohesive sediment dynamics (COSEDS).

Pentreath *et al.*, (1984, 1986) were able to account for 65% of the reported ' $^{239,240}\text{Pu}$ ' discharge by taking into account the sea bed and sea water inventories and the percentage loss through the North Channel. This

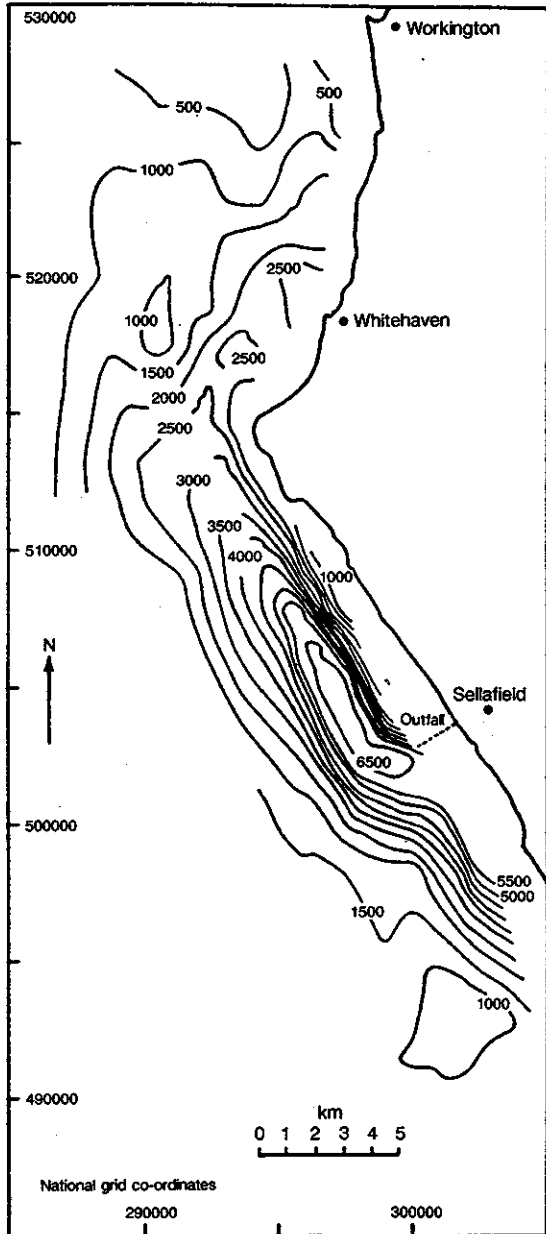


Figure 11. Distribution of ^{137}Cs (Bq kg^{-1}) in sea-bed sediments in the eastern Irish Sea for the year 1982 (from: Jones *et al.*, 1988)

calculation used the total Pu (α) discharge prior to 1978, with an assumption that it comprised $^{239,240}\text{Pu}$ only. The estimate of the 1959-1978 ^{238}Pu discharge provided by Kershaw *et al.*, (1990(a)), amounting to 18% of the total Pu (α), allowed the revision of the total environmental inventory of $^{239,240}\text{Pu}$ (sea bed and sea water inventories plus percentage loss through the North Channel) to 74% of the discharge. At least part of the 'missing' 26% is associated with estuarine and intertidal deposits. (Eakins, *et al.*, 1988, 1990) (Table 9).

A further major survey of radionuclide distributions in sediments took place in 1983 (Woodhead, 1988). This confirmed the overall distribution pattern in the north-eastern Irish Sea, but also provided greater resolution near the discharge pipe and an approximate description

of the vertical distribution (Figure 12). The latest survey of the subtidal sediments was conducted by DFR in 1988, with sampling of nearshore and intertidal sediments along the Cumbrian coast in 1989. The results of this study are still being worked up but first indications are that radionuclide concentration contours run parallel to the coast offshore of the surf zone (i.e. radionuclide concentrations in sediment decrease shorewards) (McCartney *et al.*, in press).

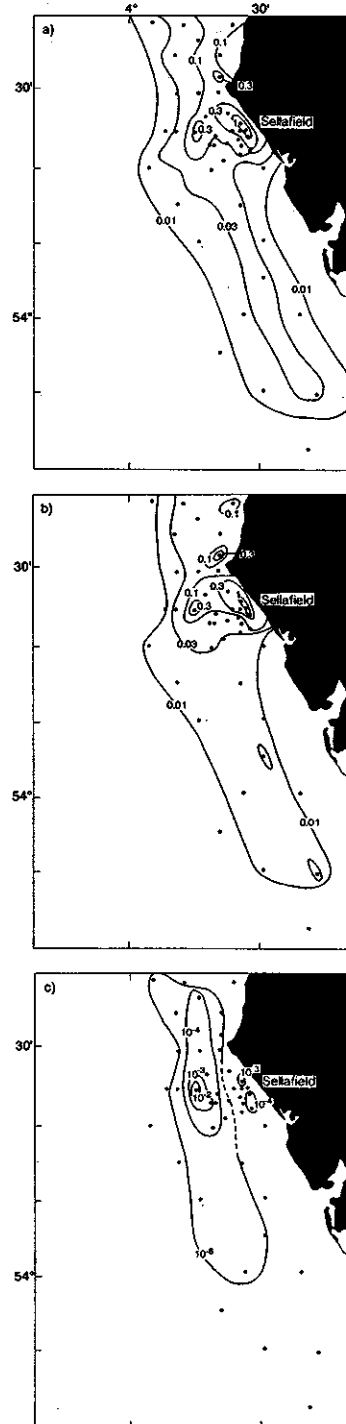


Figure 12. Distribution of $^{239,240}\text{Pu}$ (mBq m^{-2}) in sea-bed sediments in the eastern Irish Sea for the year 1983 (from: Woodhead, 1988): (a) total core; (b) at depths > 5 cm; (c) at depths > 71 cm

5.3 Sea-bed processes

It might be expected that the sediments, specifically sediment cores, could provide an unambiguous historical record of past discharges, from which inferences could be drawn, for example, about the fundamental properties of elements which are essentially 'new' to the marine environment (e.g. Np, Pu, Am, Cm) or the relative mobilities of common elements (e.g. Co) in shallow, coastal seas. There have been several attempts to match core profiles with Sellafield discharges in sediments from the Esk estuary, some 10 km south of the pipeline (sub-section 5.4). These have made use of $^{239,240}\text{Pu}/^{238}\text{Pu}$ ratios (Hetherington, 1978; Stanners and Aston, 1981; Aston and Stanners, 1981(a)) to estimate sediment accumulation rates, sometimes in combination with measurements of fission products (Stanners and Aston, 1984). Some of the uncertainties in using isotope ratios for this purpose, in the Esk or at other intertidal sites, have been pointed out by MacKenzie and Scott (1982). Most notable are the lack of ^{238}Pu discharge data prior to 1978 and the mixing of sediment contaminated at different times. This latter point was addressed by Aston *et al.* (1985) in an extensive survey of ^{238}Pu and $^{239,240}\text{Pu}$ in intertidal sediments. The most comprehensive, and successful, studies undertaken have been by Hamilton and co-workers (Clifton and Hamilton, 1982; Hamilton, 1983; Hamilton and Clarke, 1984) who stressed the importance of understanding the dynamics and variability of the whole system in order to place the results of radionuclide analyses in context.

Attempts to adopt a similar approach to that of the subtidal sediments have met with limited success. Radionuclide concentration and ratio profiles in sea-bed cores are frequently quite 'irregular', with little apparent correlation with the discharge history, and provide numerous examples - on the basis of $^{239,240}\text{Pu}/^{238}\text{Pu}$ ratios - of 'older' sediment overlying 'younger' deposits. Indeed, variations in concentration, by as much as a factor of 100, are observed in the $^{239,240}\text{Pu}$ concentration from subsamples at the same level within one core, (Pentreath, 1987). This does not necessarily represent large-scale erosion and redeposition of sea bed sediments to depths of tens of centimetres, but does represent the effects of sediment mixing by benthic fauna. This was first suggested by Pentreath *et al.*, (1980) and was subsequently evaluated and described in a series of studies (Kirby *et al.*, 1983; Kershaw *et al.*, 1983, 1984; Jensen and Crawford, 1984; Swift and Kershaw, 1986; Pentreath, 1987; Kershaw *et al.*, 1988(a)(b); Woodhead, 1988). Below 5 cm, mixing appears to be dominated by 2 species, the Echiuran worm *Maxmülleria lankesteri* and the Thalassinid shrimp *Callinassa subterranea* (Kershaw *et al.*, 1984; Swift and Kershaw, 1986). The former is

frequently found at depths of 30 to 40 cm and the latter has been observed down to 150 cm. *M. lankesteri* feeds by extending its proboscis onto the sediment surface and ingesting sediment particles (D. Hughes, pers. comm.). A proportion of this material is defaecated within the burrow, becoming incorporated in the burrow lining, leading to enhanced concentrations of $^{239,240}\text{Pu}$ and ^{241}Am , and 'recent' $^{239,240}\text{Pu}/^{238}\text{Pu}$ ratios in the burrow lining relative to adjacent sediment at the same depth (Kershaw *et al.*, 1984). *C. subterranea* excavates extensive burrow systems and transports sediment to the surface, effectively burying freshly deposited particles and also providing fresh surfaces for adsorption (J. Atkinson, pers. comm.). Both animals bring about the net transfer of sediment. In addition there are several species which also bring about a transfer of water or sediment, or which cause disruption to the primary sedimentary fabric (Swift, in press (a)). The bioturbation is both extensive and heterogenous, a combination of 'conveyor-belt' and 'biodiffusive' mixing. This is responsible for the great variety of vertical profiles of Sellafield radionuclides which has been observed in cores from the area (e.g. Pentreath *et al.*, 1980; Kirby *et al.*, 1983; Pentreath, 1987); it is very difficult to quantify this mixing process, other than on the basis of interpreting individual core profiles (Gurbutt and Kershaw, 1987). Recent evidence, provided by the ^{14}C dating of *Turritella communis* shells, suggests that the sub-tidal sediments are accumulating very slowly in the north-eastern Irish Sea ($\sim 0.1 \text{ mm year}^{-1}$, Kershaw *et al.*, 1988(a)(b)) and some intrabasinal sediment erosion and deposition is likely to occur; this will result in the mixing of relatively 'new' and 'old' sediment contamination, further blurring the sediment record (Aston *et al.*, 1985).

In addition to bioturbation, chemical conditions within the sea bed are known to exert an influence on the filtrate/sediment partitioning behaviour of redox-sensitive radionuclides such as Pu and Np (Nelson and Lovett, 1981; Harvey, 1981(a)(b); Harvey and Kershaw, 1982; Sholkovitz, 1983; Kershaw *et al.*, 1986(b); Malcolm *et al.*, 1990(a)(b)). The measurement of K_d values within the sea bed presents certain problems not encountered in water-column studies. The microbial oxidation of organic matter brings about fluctuations in the redox potential and hence influences the depth distribution of redox-sensitive components, such as iron, manganese and certain radionuclides. It is important that the measurements accurately reflect *in-situ* chemical conditions, rather than conditions which may develop during sample collection and handling, particularly if relatively reducing sediments are brought into contact with atmospheric oxygen. The problem is compounded by the relatively large quantities of pore-water required for accurate radionuclide speciation determinations (up to one litre).

Significant quantities of Pu (V + VI) (~60%) in interstitial water are found in the upper 2-5 cm of sediment near the Sellafield pipeline (Kershaw *et al.*, 1986(b); Malcolm *et al.*, 1990(b)), but not at sites further offshore in muddier sediments (Figure 13). This appears to be due to the presence of a large population (500-1000 individuals m⁻²) of the burrowing Ophiuroid *Amphiura filiformis* at the former site, resulting in enhanced bio-irrigation of the sediment by the oxygenated overlying water. K_d values for Np and Am are within the range measured in sea water.

Attempts have been made to quantify the rate of mixing by bioturbation using measurements of naturally-occurring radionuclides from the ²³⁸U decay series. The modelling of excess ²³⁴Th profiles, which approach an exponential form, in the upper 5-6 cm yields biodiffusion rates in the region of 40 cm² y⁻¹ (Kershaw *et al.*, 1988(c)). But, excess ²¹⁰Pb profiles, reaching to depths of more than 35 cm, integrate the effects of biodiffusion and conveyor-belt advective mixing, giving apparent biodiffusion rates which vary by over 2 orders of magnitude. A more

sophisticated approach is required for modelling ²¹⁰Pb/²²⁶Ra in these sediments (Swift and Kershaw, 1986; Gurbutt and Kershaw, 1987).

5.4 Intertidal and estuarine environments

The interest in the contaminated offshore area is primarily because of its potential to act as a significant long-term sink, or continuing source, of long-lived nuclides in the future. Contaminated estuaries, and the shore-line in general, are important areas because of the public's direct contact with them.

The intertidal zone, when exposed to wave activity, is dominated by sandy deposits. Consequently radionuclide concentrations are relatively low but the dynamic nature of this environment results in a large volume of sediment becoming contaminated, with uniform mixing of beach sands to the base of wave activity being common.

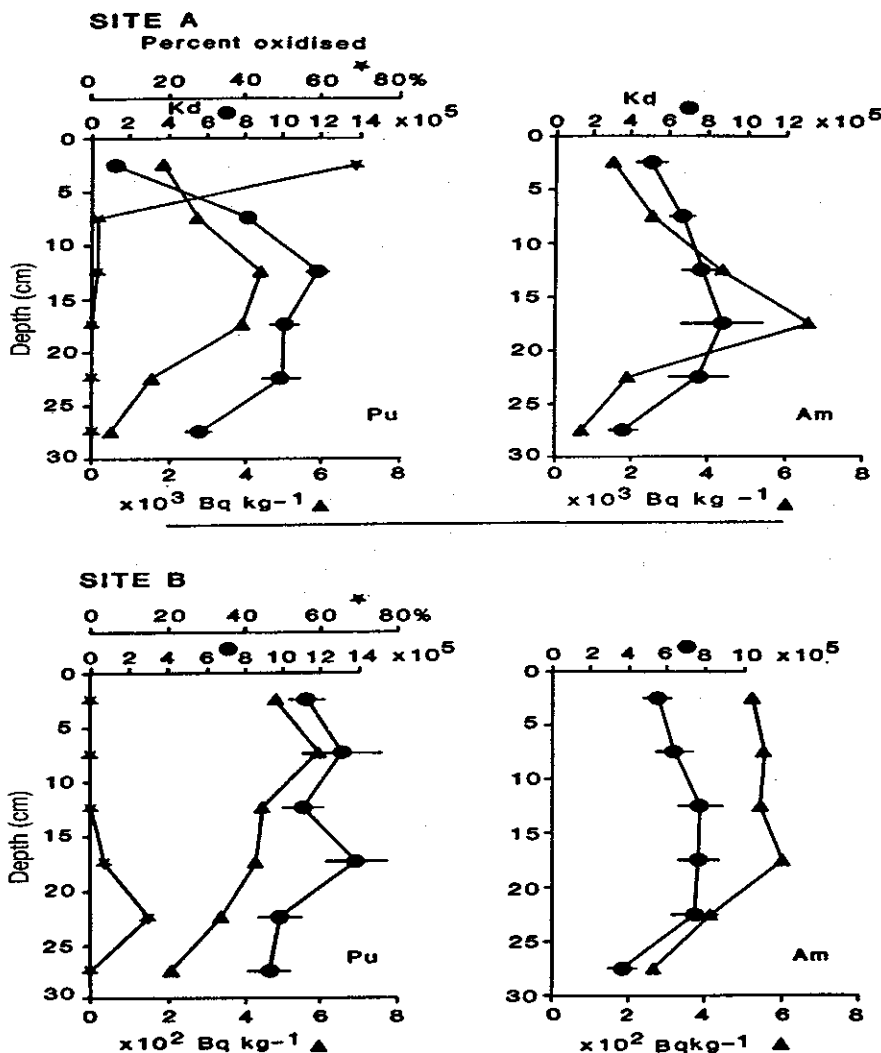


Figure 13. Depth distribution of the percentage of oxidised ^{239,240}Pu (stars), and the concentrations (triangles) and K_d values (circles) of ^{239,240}Pu and ²⁴¹Am at two sites in the eastern Irish Sea. (from: Kershaw *et al.*, 1986(b))

Inventory estimates in intertidal sediments for $^{239,240}\text{Pu}$ and ^{241}Am of 4.2 TBq and 7 TBq respectively, have been made by Eakins *et al.*, (1988, 1990) for the years 1983-84, in a zone extending from Silloth to Walney Island (Table 9). Estimates have also been made for the Duddon Estuary and Morecambe Bay, although the uncertainties are higher because of the small number of cores collected. A further assessment is underway (J. Howorth, pers. comm.). Significant quantities of artificial radionuclides are also found along the southern Scottish coast. For example, Garland *et al.* (1989) reported inventories of $^{239,240}\text{Pu}$ and ^{241}Am and ^{137}Cs in Wigtown Bay of 1.9, 2.9 and 11 TBq respectively.

An additional source of beach contamination resulted from the accidental discharge of radioactive solvent and 'crud' in November 1983 (HSE, 1984; DOE, 1984). In gross terms the incident did not add significantly to the contamination arising from routine plant operations. But highly contaminated debris was discovered along the strandline, consisting of a wide variety of natural (e.g. seaweed, feathers, shells) and man-made (mostly plastic) materials (MAFF, 1983, 1984; DOE/MAFF, 1985). Detailed examination (Woodhead *et al.*, 1985) of some of the more active items allowed a proportion to be ascribed to the 1983 incident, however, several items, on the basis of their radionuclide signature, pre-dated the incident. Their origin is not known.

The general spatial distribution of radionuclides at a number of shore-line locations, temporal trends in

concentrations and the γ -dose rates arising from them, have been reported on in the Aquatic Environment Monitoring Reports issued by MAFF (see section 8).

An estuarine complex to the south of Sellafield, the Esk Estuary, has received detailed study over a period of many years. The estuary receives waters from the rivers Irt, Mite and Esk. Early surveys indicated that the major fraction of the radioactivity in the estuary was associated with fine flocculated material (Templeton and Preston, 1966) and detailed studies were made on the relationship between radionuclide concentration and particle size, rates of penetration into the estuarine sediments, and on the estimated transit times between Sellafield and the Ravensglass area. The latter were considered to be approximately 1 month for ^{106}Ru and ^{144}Ce , and 2 to 3 months for $^{95}\text{Zr}/^{95}\text{Nb}$. It was thought, however, that such relatively fast times could have resulted in coincidental timing between discharge and favourable transport conditions (Hetherington and Jefferies, 1974).

Later studies in the area have been diverse in their nature and content. Aston and Stanners (1979) used multi-radionuclide discharge patterns and sediment profiles within the River Esk to estimate net sedimentation rates. Other studies were made to identify the extent to which ^{137}Cs , ^{144}Ce , ^{106}Ru and $^{95}\text{Zr}/^{95}\text{Nb}$ varied within the Rivers Irt and Esk (Aston and Stanners, 1982(a)), and the relationship between variations across transects within the estuary (Aston and Stanners, 1982(b)).

Table 9. Total deposits of actinides in the intertidal regions of western Cumbria (from: Eakins *et al.*, 1988)

Region	Intertidal area	Total (km ²)	Deposit in sand/mud			Total deposit in region				
			Sand/mud (km ²)	^{238}Pu (kBq m ⁻²)	$^{239,240}\text{Pu}$ (kBq m ⁻²)	^{241}Am (kBq m ⁻²)	^{238}Pu (TBq)	$^{239,240}\text{Pu}$ (TBq)	^{241}Am (TBq)	
A	Silloth - Mawbry	10.64	8.27	9	53	73	0.07	0.44	0.60	
B	Mawbry - Maryport	6.23	3.81	17	97	130	0.06	0.37	0.49	
C	Maryport - Workington	3.56	1.31	21	97	131	0.03	0.13	0.17	
D	Workington - St Bees Head	1.99	1.61	8	40	55	0.01	0.06	0.09	
E	St Bees Head - Counderton	1.29	0.80	35	167	262	0.03	0.13	0.21	
F	Counderton - Sellafield	2.51	1.25	50	227	476	0.06	0.29	0.60	
G	Sellafield - NGR 042000	1.78	1.42	50	242	390	0.07	0.34	0.56	
H	NGR 042000 - Drigg Point	3.68	3.23	68	323	718	0.22	1.04	2.32	
I	Drigg Point - Annaside (Excluding the Esk Estuary)	4.60	2.96	40	194	265	0.11	0.57	0.79	
J	Annaside - Kirksanton	2.70	1.82	14	68	96	0.03	0.13	0.17	
K	Kirksanton - Haverigg	4.41	4.31	14	63	87	0.06	0.27	0.37	
L	Walney Island (west facing, North End - SE Point)	8.46	4.56	21	100	146	0.10	0.46	0.67	
							Total	0.85	4.22	7.04
	Duddon Estuary (mean of 2 cores)	32.2	31.9	10	47	58	0.30	1.52	1.85	
	Morecambe Bay (mean of 5 cores)	215	210	10	51	74	2.2	10.8	15.6	

As is to be expected, the transuranium elements have also been examined in detail. The results of analyses published by Hetherington (1976(a)), relating to the estuary from 1966 to 1973, have been useful because, assuming a relatively short transit time from Sellafield, the Pu ratios in surface sediments can be interpreted as approximately representative of those of the recent discharge during this period. These ratios were used to estimate rates of sedimentation within the estuary (Hetherington, 1976(a), 1978; Hetherington *et al.*, 1975). More recent work by Aston and Stanners (1981(a), 1982(c)) concluded that there may actually be a considerable time-lag between Pu and Am discharge and their subsequent deposition within the estuary. Estimates of transit times differ according to the methods used, and for short-lived radionuclides, the hydrodynamic conditions which can vary considerably (Hamilton, 1983; Hamilton and Clarke, 1984; Howarth, 1984; Stanners and Aston, 1984).

Estuaries are very complex systems; and Clifton and Hamilton (1982) have stressed the necessity of taking this into account when discussing the distribution of radionuclides within them. Radionuclide distributions are closely related to sedimentary processes (Kelly and Emptage, 1992). The preservation of the historical record of the Sellafield effluent in sediment cores is dependent upon the processes of accretion, erosion and remixing, and these vary both spatially and temporally within the Esk Estuary. Thus, estimates of sedimentation rates, ranging from 0.5 up to 6 cm year⁻¹ (e.g. Stanners and Aston, 1984), may be real, but limited in geographical extent and, perhaps, short lived. In some areas of the estuary erosion has occurred leading to the exposure of sediments contaminated many years before, whilst in other areas the sediments appear to have a residence time of 10s or 100s of years (Emptage, 1992; E. I. Hamilton, pers. comm.). Sedimentation processes in the Esk, and their influence on radionuclide distributions and dose rates, have been comprehensively described and quantified in a recent study (Emptage, 1992).

The Esk Estuary is not the only one to have been studied (Lowton *et al.*, 1966). Stanners and Aston (1981) presented more recent data for ¹³⁴Cs, ¹³⁷Cs and ¹⁰⁶Ru in a number of shore-line and estuarine areas and Aston *et al.*, (1981) and Assinder (1983) described Pu in the Wyre Estuary. The distribution of ¹³⁷Cs in surface intertidal sediments in the Solway Firth has been reported by Jones *et al.*, (1984), using a hovercraft-mounted γ -spectrometer. The water and sediments of the Ribble Estuary contain artificial radionuclides from Sellafield and appreciable quantities of thorium isotopes. These latter radionuclides are discharged from the BNFL Springfields' works arising from the manufacture of fuel elements and the production of uranium hexafluoride. Highest concentrations occur in areas of fine sediment deposition (MAFF,

1992) and there is some evidence of transfer of ²³⁰Th out of the estuary (P. J. Kershaw, unpublished). Relatively high concentrations of ²³⁷Np have been measured within the estuary (Assinder *et al.*, 1991), attributable to waste arising from Springfields owing to the use of reprocessed (i.e. via Sellafield) uranium.

Pu concentrations and isotope ratios have been used to illustrate the dominance of sediment re-working and transport in intertidal sediments (Aston *et al.*, 1985). Intertidal sediments along the southern Scottish coast have revealed interesting profiles of ¹³⁷Cs and ^{239,240}Pu which MacKenzie and Scott (1982) have used to test different possible interpretations of sedimentation and remobilisation, and demonstrate the limitations of using solid phase concentration profiles to examine the effects of diagenesis (cf. pore-water chemistry, subsection 5.4). Sediment diagenesis has been shown to be significant in the case of Cs mobility in Scottish sea lochs (McKay and Baxter, 1985), with sequential leaching techniques being employed. The cycling of Fe and Mn is thought to exert considerable influence on the remobilisation of Pu and Am in the Esk estuary, and in particular the seasonal occurrence of 'organoliths' associated with *Corophium* burrows (Hamilton, 1989).

Laboratory experiments and field studies in the Esk Estuary have demonstrated the non-conservative behaviour of Pu. The tidal circulation results in the transport of particle-associated Pu to the upper reaches of the river. The low salinity and pH changes result in the rapid desorption of a labile form of Pu (Assinder *et al.*, 1985; Hamilton-Taylor *et al.*, 1987; Kelly *et al.*, 1988; Mudge *et al.*, 1988). This mechanism could result in the net loss of Pu from the estuary, although the magnitude and persistence of such a process are difficult to evaluate (Burton, 1986). Kelly *et al.*, (1991) estimated that, over a full tidal cycle, there was a net gain to the inner Esk Estuary of ca. 18 t of sediment and ca. 85 MBq of particulate phase ^{239,240}Pu and a probable net loss of ca. 1 to 2 MBq of solution phase ^{234,240}Pu. They considered this net input to be typical of the Esk Estuary under the normal conditions of low river flows.

The presence of suspended sediment is thought to be the principal cause of the enrichment of sea spray by the actinides (Eakins and Lally, 1984). A considerable effort has been made to assess and model (section 9) the magnitude and importance of the sea-to-land transfer of radionuclides along the Cumbrian coast (e.g. Pattenden *et al.*, 1989). The ²⁴¹Am/^{239,240}Pu and ¹³⁷Cs/^{239,240}Pu ratios for airborne and deposited (on land) materials were in better agreement with the sea water particulate ratios rather than those of the filtrate. Concentrations in airborne material reflect both current and past discharges. An overall downward trend at Eskmeals, in the period 1978 to 1987, was less evident

than the decline in the Sellafield discharge. McKay and Pattenden (1990) have reviewed studies of this transfer process along the coasts of Cumbria and Caithness over the same time. A significant seasonal signal is imposed. The transfer mechanism has also been identified on the coasts of Northern Ireland (Garland *et al.*, 1990). There is limited evidence of airborne sea-to-land transfer along the northern Solway coastal zone but tidal inundation does result in elevated concentrations of ^{137}Cs , Pu (α) and ^{241}Am in some riverine systems (McDonald *et al.*, 1992). Deposition of seaborne sediment by tidal action has been shown to be the main route by which radionuclides of Sellafield origin reach grazing land bordering the Esk Estuary (Bradford *et al.*, 1984; Horrill, 1984; Curtis *et al.*, 1991). The mapping of such deposits has been assisted by the development of aerial radiometric techniques (D. Sanderson, pers. comm.).

6. ACCUMULATION OF RADIONUCLIDES BY BIOTA

6.1 Fission-product nuclides

From the earliest commissioning of the Sellafield plant, samples of marine biological materials in the area were analysed for their radionuclide content (Dunster, 1956, 1958). One of the first important observations was that benthic algae concentrated ^{106}Ru , which was of significance because one species, *Porphyra umbilicalis*, was harvested for human consumption (Fair and McLean, 1956; Williams and Davidge, 1962). Detailed radioecological surveys of the area were made in 1959/1960 (Mauchline, 1963; Mauchline and Templeton, 1963), which included measurements of ^{40}K , and of weapons fallout in areas away from Sellafield. Concentrations of the major gamma-emitting fission products were recorded in a large range of benthic algae, invertebrates and fish, and some concentration factors (CFs; concentration per unit mass biota (Bq kg^{-1})/concentration per unit volume sea water (Bq l^{-1})) were calculated for ^{106}Ru , ^{144}Ce , ^{137}Cs , $^{95}\text{Zr}/^{95}\text{Nb}$ and ^{90}Sr in benthic algae. The intertidal area in the immediate vicinity of Sellafield was also studied in detail (Mauchline *et al.*, 1964).

Most of the early data relating to Sellafield, and elsewhere, were summarised by Mauchline and Templeton (1964); monitoring was discussed by Dunster *et al.* (1964) and Longley and Templeton (1965); and, early estimates of CFs and biological half-times were summarised by Bryan *et al.* (1966). The results of detailed surveys of the concentrations at various distances from Sellafield, were published by Templeton and Preston (1966).

Large numbers of environmental samples have been collected and analysed as part of monitoring and surveillance programmes; summaries of these data since 1963 have been published routinely by MAFF (e.g. Tables 10 and 11; MAFF, 1990) (section 8). Many data have also been published elsewhere, both relating to environmental samples and to supporting laboratory studies. Some of the earliest of these were on ^{106}Ru . Jones (1960) analysed locally caught plaice for ^{106}Ru and also studied the accumulation of ^{106}Ru by plaice, mussels, and three benthic species of algae, in aquaria, in relation to chemical form. The results of continuing environmental studies were published by Preston and Jefferies (1969(a)), and in this paper further estimates of concentration factors were made for ^{106}Ru , $^{95}\text{Zr}/^{95}\text{Nb}$, ^{137}Cs and ^{90}Sr in a range of biological materials (Table 12). Data were also presented on the relationship between the concentration of ^{106}Ru in *Porphyra* and the rate of its discharge from Sellafield for the period 1959 to 1968. The difficulties of relating concentrations of radionuclides in fish to those of water were expressed, drawing attention to the disequilibrium between the two that was likely to exist close to the discharge point.

Apart from the accumulation of ^{106}Ru by *Porphyra*, the accumulation of Cs nuclides by fish was also recognised to be important because of the quantities of fish caught for human consumption in the area. Laboratory studies on the accumulation of ^{134}Cs from sea water by a large number of fish species, and some invertebrates, were made by Morgan (1964) and an environmental study of the thornback ray (*Raja clavata*) was made by Mauchline and Taylor (1964). It is the plaice (*Pleuronectes platessa*), however, which has received the greatest attention. Jefferies and Hewett (1971) made a detailed laboratory study on the accumulation of ^{134}Cs from sea water by the plaice, with comparative observations on the thornback ray. It was concluded that at least 50% of the environmental body burden of Cs in the plaice, and 80% of that in the ray, must be due to absorption from food rather than direct uptake from sea water. A detailed environmental study of I-group plaice off Sellafield was made by Pentreath and Jefferies (1971), in which the fraction of ^{137}Cs taken up from food was related to diet, temperature, body size and rate of growth. An assessment of the other gamma-emitting fission product nuclides in the diet of plaice was also made (Pentreath and Jefferies, 1971; Pentreath *et al.*, 1973) with the conclusion that apart from ^{137}Cs , and to some extent ^{106}Ru , other fission products are not accumulated by fish muscle. This is in contrast to, for example, crabs (*Cancer pagurus*) which have readily detectable concentrations of ^{144}Ce , $^{95}\text{Zr}/^{95}\text{Nb}$ as well as ^{106}Ru and ^{137}Cs in hepatopancreas and muscle tissue (Pentreath, 1980(a)). Laboratory studies of the accumulation of Cs by the plaice were continued, particularly with regard to the effects of changes in body size and water temperature (Pentreath, 1975), and on the relative importance of food and

Table 10(a). Beta/gamma radioactivity in fish from the Irish Sea vicinity and further afield, 1990 (from: MAFF 1992)

Sampling area/ landing point	Sample	No. of sampling observa- tions	Mean radioactivity concentration (wet), Bq kg ⁻¹		
			Total beta	¹³⁴ Cs	¹³⁷ Cs
Sellafield coastal area ¹	Cod	6	190	0.8	37
	Plaice	2	190	1.0	37
Sellafield offshore area ¹	Cod	3	190	0.5	27
	Plaice	4	130	0.2	14
	Flounder	1	150	0.9	69
	Dab	3	160	0.4	24
	Whiting	2	170	0.6	40
Ravenglass ²	Cod	16	160	0.5	29
	Plaice	8	150	0.3	24
	Whiting	2	120	0.3	23
	Flounder	1	180	0.8	45
	Hake	1	140	0.8	21
	Whitebait	1	99	ND	13
	Sea trout	1	160	0.7	31
	Salmon	1	100	ND	2.9
Whitehaven ²	Cod	4	160	0.4	21
	Plaice	4	130	0.1	14
	Rays	3	150	0.6	37
	Whiting	1	160	0.5	35
	Herring	3	110	ND	10
	Hake	1	140	0.5	20
Morecambe Bay ¹	Flounder	4	160	0.4	54
	Plaice	3	130	0.07	25
	Bass	2	170	0.6	50
	Whitebait	1	94	0.3	20
Cumbrian rivers ³	Brown trout	2	120	ND	ND
	Sea trout	5	130	"	19
Fleetwood ²	Cod	4	140	0.2	17
	Plaice	4	120	0.2	14
	Fish meal ⁴	2	240	0.2	3.7
Isle of Man ²	Cod	4	150	0.1	11
	Plaice	3	100	ND	6.0
	Herring	2	170	0.1	7.5
	Whiting	1	120	0.3	14
Inner Solway ¹	Salmon	1	160	ND	1.1
	Sea trout	2	140	"	15
	Flounder	4	180	0.6	65
Kirkcudbright ²	Plaice	3	100	0.2	9.7
North Anglesey ¹	Plaice	2	100	ND	2.5
	Spurdog	2	110	0.2	12
Northern Ireland ²	Cod	6	130	0.07	7.5
	Whiting	8	120	ND	8.8
	Herring	4	120	0.3	7.2
	Dogfish	5	90	0.1	7.4
	Saithe	1	100	ND	5.0
Ayr ²	Cod	4	140	0.2	8.9
	Plaice	3	100	0.06	5.8
	Whiting	1	130	ND	2.6
Loch Leven ¹	Salmon	1	130	"	1.1
Minch ¹	Cod	4	150	ND	2.0
	Plaice	4	100	0.03	1.3
	Haddock	4	120	ND	1.0
	Herring	2	120	"	1.5
	Mackerel	7	110	"	0.7
Shetland ¹	Fish meal ⁴	1	500	ND	2.1
Northern North Sea ¹	Plaice	4	100	"	1.4
	Cod	6	130	"	1.3
	Haddock	8	120	"	1.0
	Saithe	4	NA	"	1.4
	Herring	6	120	"	0.8
	Mackerel	2	NA	"	0.4
	Whiting	1	"	"	1.4
Mid North Sea ¹	Plaice	8	96	0.02	1.1
	Cod	8	130	0.04	1.9
	Haddock	5	NA	ND	0.9
	Herring	8	110	"	1.1
	Mackerel	1	NA	"	1.0
	Whiting	2	"	"	1.1
Southern North Sea ¹	Plaice	3	91	"	1.0
	Cod	3	130	"	1.0
	Herring	3	110	"	0.7
	Mackerel	1	NA	"	0.6
	Whiting	1	"	"	1.0
Norwegian Sea ¹	Cod	2	110	ND	1.1
Baltic Sea ¹	Cod	3	110	2.1	16
	Herring	2	110	1.1	7.9
English Channel ¹	Plaice	2	110	ND	0.3
	Cod	2	130	"	0.6
	Whiting	1	98	"	ND
	Mackerel	1	100	"	0.5
Iceland area ¹	Cod	2	110	"	0.3
Icelandic processed	Cod	2	110	"	0.8
	Haddock	1	110	"	0.1

ND = not detected; NA = not analysed; ¹Sampling area; ²Landing point; ³Samples collected from a number of rivers by the North West Water Authority; ⁴Concentrations refer to weight of sample as supplied

water in the maintenance of body burdens (Hewett and Jefferies, 1978). Laboratory studies were also made on the accumulation of ^{137}Cs , ^{90}Sr , ^{90}Y , ^{106}Ru , ^{144}Ce , ^{95}Zr and ^{95}Nb by plaice eggs (Woodhead, 1970). The concentrations of ^{137}Cs in samples of plaice taken off Sellafield, normalised to a discharge rate of 1 Ci day^{-1} (37 GBq), from 1963 to 1977, have been discussed by Pentreath (1980(a), 1982) and Preston and Portmann (1981).

The occurrence of ^{137}Cs in samples other than fish taken from the north-eastern Irish Sea has been variously reported on, and in some instances estimates of CFs have been made (Broom *et al.*, 1975; Pentreath, 1976(a); Baxter *et al.*, 1979). Sellafield-derived ^{137}Cs has been detected in *Mytilus edulis* collected from the east and north-east coasts of Ireland, with fallout levels being found on the south and west coasts (Crowley *et al.*, 1990). Concentrations of ^{137}Cs in *Fucus vesiculosus* collected from the east coast of Ireland decreased by approximately 20% per annum during the period 1983 to 1986 (McAulay and Pollard, 1988). Elevated levels of ^{103}Ru , ^{110m}Ag , ^{131}I and ^{129m}Te in winkles from the Irish

Sea were reported following the Chernobyl reactor accident, in April 1986, but concentrations decreased rapidly thereafter (Camplin *et al.*, 1986).

There have been relatively few studies of Tc in the marine environment. Recent reviews have been completed by Desmet and Myttenaere (1986) and Sparkes and Long (1988). ^{99}Tc concentrations in the Irish Sea have been reported for a variety of shellfish, *Fucus vesiculosus* and *Porphyra* (Dutton and Ibbett, 1973; Pentreath *et al.*, 1980), and are routinely included in MAFF's monitoring reports (see, for example, MAFF, 1992). Laboratory uptake experiments with the lobster (*Homarus gammarus*) using ^{99m}Tc have been carried out (Pentreath, 1981), with later studies examining the effects of moulting (Swift, 1985). Uptake is rapid with whole body CFs of >2000 and the highest percentage of the total activity is found in the digestive gland. Experiments have been conducted on the accumulation and retention of ^{99m}Tc by the edible winkle (*Littorina littorea*) (Swift, 1989), with starvation shown to be a significant factor in controlling elimination.

Table 10(b). Other beta/gamma radioactivity in fish from the Irish Sea vicinity, 1990 (from: MAFF 1992)

Sampling area/ landing point	Sample	No. of sampling observa- tions	Mean radioactivity concentration(wet), Bq kg ⁻¹					
			¹⁴ C	⁶⁰ Co	⁶⁵ Zn	⁹⁰ Sr	⁹⁹ Tc	¹⁴⁷ Pm
Sellafield offshore area ¹	Plaice	1	82	ND	ND	0.065	0.68	0.0074
	Cod	1	95	"	"	0.12	0.39	0.013
Ravenglass ²	Whitebait	1	NA	0.5	"	0.38	NA	NA
Whitehaven ²	Plaice	1	43	ND	"	0.070	"	"
	Cod	1	43	"	"	0.051	"	"
Morecambe Bay ¹	Plaice	1	35	"	"	NA	"	"
	Flounder	1	46	"	"	"	"	"
Fleetwood ²	Plaice	1	34	"	"	"	"	"
	Cod	1	23	"	"	"	"	"
	Fish meal ³	1	NA	"	"	0.50	"	"
Isle of Man ²	Plaice	1	30	"	"	NA	"	"
Inner Solway ¹	Flounder	1	22	"	"	"	"	"
North Anglesey ²	Dogfish	1	20	"	"	"	"	"
Minch ¹	Mackerel	1	64	"	"	"	"	"
Shetland ²	Fish meal ³	1	NA	"	"	0.042	"	"
Northern North Sea ¹	Cod	2	"	0.1	"	NA	"	"
	Haddock	1	12	ND	"	"	"	"
	Herring	2	NA	0.3	"	"	"	"
	Mackerel	2	"	0.9	"	"	"	"
Mid North Sea ¹	Cod	1	7.8	ND	"	"	"	"
Iceland area ¹	Cod	1	19	"	"	"	"	"
Icelandic processed	Cod	1	15	"	"	"	"	"
English Channel ¹	Plaice	2	NA	0.06	0.24	"	"	"

NA = not analysed

ND = not detected

¹ Sampling area

² Landing point

³ Concentrations refer to weight of sample as supplied

Table 10c. Beta/gamma radioactivity in shellfish from the Irish Sea vicinity and further afield, 1990 (from: MAFF 1992)

Sampling area/ landing point	Sample	No. of sampling observa- tions	Mean radioactivity concentration (wet), Bq kg ⁻¹								
			Total beta	¹⁴ C	⁵⁴ Mn	⁶⁰ Co	⁶⁵ Zn	⁹⁰ Sr	⁹⁵ Zr	⁹⁵ Nb	⁹⁹ Tc
Sellafield coastal area ¹	Crab	5	150	100	ND	3.4	ND	1.2	ND	ND	5.8
	Lobster	5	420	160	"	2.0	"	0.65	"	"	630
	Winkles ³	12	380	72	"	5.6	"	17	0.8	0.4	76
	Winkles ⁴	4	360	NA	"	6.4	"	NA	2.5	3.7	NA
	Winkles ⁵	3	240	"	"	3.4	"	"	ND	ND	"
	Mussels ³	5	220	"	"	3.3	"	"	0.6	0.5	"
Limpets ³	3	410	"	"	6.0	"	"	0.5	0.8	"	
Sellafield offshore area ¹	Whelks	2	230	"	"	5.3	"	"	ND	ND	"
St Bees ¹	Winkles	4	290	32	"	4.8	"	9.4	1.1	0.7	45
	Mussels	4	260	NA	"	4.0	"	NA	4.1	1.1	NA
	Limpets	4	430	"	"	5.0	"	"	1.8	2.9	"
Nethertown ¹	Winkles	12	350	65	"	5.6	"	13	2.7	3.2	72
Drigg ¹	Winkles	4	430	59	"	8.5	"	7.5	1.9	1.6	150
Ravenglass ¹	Cockles	4	230	NA	"	9.6	"	NA	1.6	2.6	NA
	Mussels	4	230	"	"	4.6	"	"	1.3	ND	"
Ravenglass ²	Crabs	3	110	"	"	1.6	"	"	ND	"	"
	Lobsters	3	240	"	"	0.5	"	"	"	"	"
	Whelks	3	160	"	"	3.4	0.1	"	"	"	"
Tam Bay ¹	Winkles	4	350	"	"	5.1	ND	"	3.1	4.7	"
Whitehaven ²	<i>Nephrops</i>	4	120	"	"	1.3	"	"	ND	ND	"
	Whelks	4	120	"	"	0.6	"	"	"	"	"
Parton ¹	Winkles	12	240	"	"	2.7	"	"	0.7	0.4	"
Roosebeck ¹	Oysters	4	75	"	"	0.7	"	"	ND	ND	"
Haverigg ¹	Cockles	2	140	"	"	3.5	"	"	0.7	0.7	"
Millom ¹	Mussels	2	170	"	"	1.1	"	"	ND	ND	"
Whitrigg scar ¹	Shrimps	1	120	"	"	0.9	"	"	"	"	"
Morecambe Bay ¹	Shrimps	4	96	"	"	ND	"	0.15	"	"	"
	Cockles	4	100	"	"	1.6	"	1.0	"	"	"
Heysham ¹	Cockles	4	85	"	"	1.4	"	NA	"	"	"
	Mussels	4	71	"	"	0.3	"	"	"	"	"
Fleetwood ²	Squid	1	86	"	"	ND	"	"	"	"	"
	Whelks	3	110	"	"	0.5	"	"	"	"	"
Isle of Man ²	Scallops	4	86	"	"	ND	"	"	"	"	
Inner Solway ¹	Shrimps	4	100	"	"	"	"	"	"	"	
Southerness ¹	Winkles	4	230	"	"	1.0	"	"	0.1	"	"
Kirkcudbright ²	Scallops	4	49	"	"	ND	"	"	ND	"	"
	Queens	4	64	"	"	0.09	"	"	"	"	"
North Solway coast ¹	Cockles	3	110	"	"	2.0	"	"	0.3	0.2	"
	Winkles	4	110	"	"	1.0	"	"	ND	ND	"
Wirral ¹	Shrimps	2	72	"	"	0.3	"	"	"	"	0.72
	Cockles	2	69	"	"	0.4	"	"	"	"	1.1
Conwy ²	Mussels	2	38	"	"	ND	"	"	"	"	NA
North Anglesey ¹	Crabs	2	89	"	"	"	"	"	"	"	"
	Winkles	2	88	"	"	0.4	"	"	"	"	"
Northern Ireland ²	<i>Nephrops</i>	8	110	"	"	0.2	"	"	"	"	"
	Winkles	4	85	"	"	0.1	"	"	"	"	"
Minch ¹	<i>Nephrops</i>	4	89	"	"	ND	"	"	"	"	
Northern North Sea ¹	<i>Nephrops</i>	5	93	"	"	"	"	"	"	"	
Mid North Sea ¹	Queens	1	NA	"	"	"	"	"	"	"	"
	Mussels	1	54	"	"	"	"	"	"	"	"
	Mussels ⁶	2	29	"	"	"	"	"	"	"	"
Southern North Sea ¹	Cockles	2	21	"	"	1.7	"	"	"	"	"
	Cockles ⁷	2	45	"	"	0.4	"	"	"	"	"
	Mussels	5	36	"	"	ND	"	"	"	"	"

Table 10(c). Continued

Sampling area/ landing point	Sample	No. of sampling observa- tions	Mean radioactivity concentration (wet), Bq kg ⁻¹									
			¹⁰³ Ru	¹⁰⁶ Ru	^{110m} Ag	¹²⁵ Sb	¹³⁴ Cs	¹³⁷ Cs	¹⁴⁴ Ce	¹⁴⁷ Pm	¹⁵⁴ Eu	¹⁵⁵ Eu
Sellafield coastal area ¹	Crab	5	ND	11	4.4	0.7	ND	10	ND	2.7	ND	ND
	Lobster	5	"	8.9	6.7	0.2	0.2	14	0.6	4.8	0.08	0.06
	Winkles ³	12	"	6.5	6.5	3.3	0.06	25	1.4	12	ND	0.3
	Winkles ⁴	4	0.2	90	9.0	4.3	0.1	23	3.1	NA	1.2	0.5
	Winkles ⁵	3	ND	33	5.5	3.1	ND	15	ND	"	ND	ND
	Mussels ³	5	0.09	44	ND	2.1	0.1	8.5	0.5	"	1.0	0.6
	Limpets ³	3	ND	37	2.9	3.7	ND	13	0.6	"	0.7	0.6
Sellafield offshore area ¹	Whelks	2	"	57	9.2	2.2	"	6.4	1.5	"	0.6	0.4
St Bees ¹	Winkles	4	"	60	6.6	4.5	0.2	24	1.7	8.0	0.6	0.3
	Mussels	4	"	86	3.0	2.5	ND	11	2.4	NA	0.2	0.3
	Limpets	4	"	47	3.3	6.0	0.2	26	2.7	"	1.5	0.9
Nethertown ¹	Winkles	12	0.2	87	8.0	5.5	0.4	31	4.0	14	1.3	0.7
Drigg ¹	Winkles	4	0.4	130	8.7	5.9	ND	23	4.3	16	0.5	0.5
Ravenglass ¹	Cockles	4	ND	61	1.1	0.9	"	15	3.4	NA	2.0	1.4
	Mussels	4	"	71	ND	3.7	"	7.6	ND	"	ND	0.6
Ravenglass ²	Crabs	3	"	5.5	2.2	0.4	"	6.6	"	"	"	ND
	Lobsters	3	"	ND	3.3	ND	"	7.8	"	"	"	0.2
	Whelks	3	"	30	6.2	1.3	"	5.1	"	"	"	ND
Tam Bay ¹	Winkles	4	0.5	92	5.8	3.9	0.2	26	3.4	"	0.9	0.6
Whitehaven ²	<i>Nephrops</i>	4	ND	ND	ND	ND	0.09	12	ND	"	ND	ND
	Whelks	4	"	4.1	1.3	"	ND	3.5	"	"	"	"
Parton ¹	Winkles	12	0.1	37	2.2	3.4	0.2	26	0.09	"	0.03	0.1
Roosebeck ¹	Oysters	4	ND	3.5	2.1	ND	ND	5.0	ND	"	ND	ND
Haverigg ¹	Cockles	2	"	20	ND	1.0	0.2	18	1.6	"	1.1	0.5
Millom ¹	Mussels	2	0.2	21	"	0.7	0.09	8.4	0.9	"	ND	0.1
Whitrigg scar ¹	Shrimps	1	ND	ND	"	ND	0.7	20	ND	"	"	ND
Morecambe Bay ¹	Shrimps	4	"	"	"	"	0.2	20	"	"	"	"
	Cockles	4	"	4.4	"	0.4	0.2	15	"	"	"	"
Heysham ¹	Cockles	4	"	3.7	"	0.5	0.06	8.7	"	"	0.07	0.1
	Mussels	4	"	2.1	"	0.2	0.1	4.5	"	"	ND	ND
Fleetwood ²	Squid	1	"	ND	"	ND	ND	4.7	"	"	"	"
	Whelks	3	"	2.3	0.5	"	"	3.1	"	"	"	"
Isle of Man ²	Scallops	4	"	ND	ND	"	"	1.1	"	"	"	"
Inner Solway ¹	Shrimps	4	"	"	"	"	0.2	21	"	"	"	"
Southerness ¹	Winkles	4	"	8.6	1.6	0.9	0.1	24	"	"	0.1	"
Kirkcudbright ²	Scallops	4	"	ND	ND	ND	ND	0.4	"	"	ND	"
	Queens	4	"	"	0.3	"	"	1.2	"	"	"	"
North Solway coast ¹	Cockles	3	"	3.0	ND	"	"	13	"	"	"	"
	Winkles	4	"	3.8	2.0	0.5	"	4.8	"	"	"	"
Wirral ¹	Shrimps	2	"	ND	ND	ND	"	7.1	"	"	"	"
	Cockles	2	"	"	"	"	"	7.0	"	"	"	"
Conwy ²	Mussels	2	"	"	"	"	"	1.1	"	"	"	"
North Anglesey ¹	Crabs	2	"	"	0.07	"	"	1.7	"	"	"	"
	Winkles	2	"	0.7	0.2	"	"	3.7	"	"	"	"
Northern Ireland ²	<i>Nephrops</i>	8	"	ND	ND	"	"	3.7	"	"	"	"
	Winkles	4	"	"	0.08	"	"	1.1	"	"	"	"
Minch ¹	<i>Nephrops</i>	4	"	"	0.08	"	"	0.7	"	"	"	"
Northern North Sea ¹	<i>Nephrops</i>	5	"	"	ND	"	"	0.7	"	"	"	"
Mid North Sea ¹	Queens	1	"	"	"	"	"	ND	"	"	"	"
	Mussels	1	"	"	"	"	"	0.5	"	"	"	"
	Mussels ⁶	2	"	"	"	"	"	0.08	"	"	"	"
Southern North Sea ¹	Cockles	2	"	"	"	"	"	0.1	"	"	"	"
	Cockles ⁷	2	"	"	"	"	"	0.2	"	"	"	"
	Mussels	5	"	"	"	"	"	0.4	0.1	"	"	"

NA = not analysed; ND = not detected; ¹Sampling area; ²Landing point; ³Samples collected by Consumer 116; ⁴Samples collected by Consumer 460; ⁵Samples collected by Consumer 311; ⁶Landed in Denmark; ⁷Landed in The Netherlands

Table 11. Transuranic radioactivity in fish and shellfish from the Irish Sea vicinity and further afield, 1990 (from: MAFF 1992)

Sampling area/ landing point	Sample	No. of sampling observa- tions	Mean radioactivity concentration (wet), Bq kg ⁻¹						
			²³⁷ Np	²³⁸ Pu	²³⁹ Pu+ ²⁴⁰ Pu	²⁴¹ Pu	²⁴¹ Am	²⁴² Cm	²⁴³ Cm+ ²⁴⁴ Cm
Sellafield coastal area ¹	Plaice	1	NA	0.012	0.049	NA	0.068	ND	0.00019
	Cod	2	"	0.0088	0.036	"	0.049	0.00029	0.00016
	Crab	3	0.060	0.25	1.0	29	3.6	0.0045	0.011
	Lobster	3	0.12	0.16	0.67	21	7.4	ND	0.029
	Winkles ³	4	0.16	4.9	22	370	37	0.038	0.098
	Winkles ⁴	2	NA	4.7	21	360	38	0.13	0.080
	Winkles ⁵	3	"	2.0	8.8	150	15	ND	0.043
	Mussels ³	1	"	3.5	16	NA	28	0.083	0.082
Limpets ³	1	"	4.1	19	"	33	0.090	0.14	
Sellafield offshore area ¹	Plaice	1	0.0007	0.0022	0.0098	0.18	0.018	ND	0.00004
	Cod	1	0.0004	0.0028	0.012	NA	0.022	0.00012	0.00008
	Whelks	1	NA	1.0	4.4	"	15	0.056	0.043
St Bees ¹	Winkles	4	0.12	3.8	17	280	29	0.043	0.064
	Mussels	2	NA	3.7	16	280	26	0.061	0.077
	Limpets	1	"	5.3	24	NA	39	ND	0.11
Nethertown ¹	Winkles	4	0.32	5.6	25	430	41	0.11	0.10
Drigg ¹	Winkles	4	0.31	6.3	27	470	54	0.11	0.16
Ravenglass ¹	Whitebait	1	NA	0.11	0.49	7.8	0.64	ND	0.0020
	Cockles	1	"	4.5	19	330	49	0.12	0.21
	Mussels	1	"	4.1	17	310	29	0.078	0.12
Ravenglass ²	Plaice ⁶	1	"	0.0027	0.012	NA	0.024	ND	0.00009
	Cod ⁶	1	"	0.0029	0.013	"	0.019	"	0.00005
	Crab ⁷	1	"	0.14	0.63	"	2.7	"	0.010
	Lobster ⁷	1	"	0.11	0.48	"	9.4	"	0.026
	Whelks ⁷	1	"	0.48	2.1	36	4.6	0.018	0.019
Tam Bay ¹	Winkles	1	"	5.1	22	390	37	0.084	0.11
Whitehaven ²	Plaice	1	"	0.0015	0.0063	NA	0.011	0.00004	0.00005
	Cod	1	"	0.00060	0.0027	"	0.0060	ND	0.00002
	Herring	1	"	0.0045	0.022	"	0.031	"	0.00007
	Rays	1	"	0.00093	0.0045	"	0.0068	"	0.00002
	<i>Nephrops</i>	1	"	0.052	0.26	"	0.67	"	0.0020
	Whelks	1	"	0.20	0.99	15	1.4	"	0.0036
Parton ¹	Winkles	1	"	2.7	13	210	20	"	0.058
Roosebeck ¹	Oysters	1	"	0.29	1.4	NA	1.2	0.0031	0.0028
Haverigg ¹	Cockles	1	"	2.7	12	"	28	0.061	0.076
Millom ¹	Mussels	1	"	0.98	4.6	"	8.0	0.024	0.020
Morecambe Bay ¹	Shrimps	1	"	0.011	0.052	0.79	0.077	ND	0.00029
	Cockles	1	"	0.77	3.4	54	8.5	"	0.024
Heysham ¹	Mussels	1	"	0.23	1.1	NA	1.8	"	0.0054
	Cockles	1	"	0.68	3.4	"	8.4	0.033	0.011
Fleetwood ²	Cod	1	"	0.00029	0.0013	"	0.0025	ND	ND
	Plaice	1	"	0.00060	0.0030	"	0.0062	"	0.00002
	Fish meal ⁸	1	"	0.0034	0.019	"	0.023	"	ND
	Whelks	1	"	0.10	0.55	7.9	0.83	"	0.0023

Table 11. Continued

Sampling area/ landing point	Sample	No. of sampling observa- tions	Mean radioactivity concentration (wet), Bq kg ⁻¹						
			²³⁷ Np	²³⁸ Pu	²³⁹ Pu+ ²⁴⁰ Pu	²⁴¹ Pu	²⁴¹ Am	²⁴² Cm	²⁴³ Cm+ ²⁴⁴ Cm
Isle of Man ²	Cod	1	NA	0.00016	0.00079	NA	0.0014	ND	ND
	Plaice	1	"	0.00047	0.0023	"	0.0030	"	0.00001
	Herring	1	"	0.00028	0.0013	"	0.0016	"	ND
	Scallops	1	"	0.029	0.14	"	0.045	"	"
Inner Solway ¹	Sea trout	1	"	0.00060	0.0030	"	0.0042	"	0.00001
Southerness ¹	Winkles	1	"	1.1	5.3	"	8.7	"	0.034
Kirkcudbright ²	Plaice	1	"	0.00060	0.0031	"	0.0059	"	ND
	Scallops	1	"	0.018	0.088	"	0.034	"	"
	Queens	1	"	0.019	0.097	"	0.11	"	0.00024
North Solway coast ¹	Cockles	1	"	1.2	5.6	"	13	"	0.046
	Winkles	1	"	0.43	2.0	"	3.3	"	0.0080
Ayr ²	Cod	1	"	0.00012	0.00059	"	0.00087	"	ND
	Plaice	1	"	0.00020	0.00096	"	0.0017	"	0.00001
Wirral ¹	Cockles	1	"	0.33	1.7	"	3.0	"	0.011
Conwy ²	Mussels	1	"	0.046	0.22	"	0.35	"	0.00086
North Anglesey ¹	Spurdog	1	"	0.00009	0.00042	"	0.00074	"	ND
	Winkles	1	"	0.20	0.98	"	1.3	"	0.0030
Northern Ireland ²	Whiting	1	"	0.0011	0.0058	"	0.0079	"	0.00002
	<i>Nephrops</i>	1	"	0.014	0.069	"	0.17	"	0.00034
	Winkles	1	"	0.052	0.25	"	0.15	"	ND
Minch ¹	Cod	1	"	0.00006	0.00035	"	0.00061	"	"
	Haddock	1	"	0.00011	0.00067	"	0.00057	"	"
	Mackerel	1	"	0.00013	0.00072	"	0.00016	"	"
	<i>Nephrops</i>	1	"	0.00072	0.0046	"	0.0048	"	0.00002
Shetland ¹	Fish meal ⁸	1	"	0.0012	0.0085	"	0.0015	"	ND
Northern North Sea ¹	Cod	1	"	0.00007	0.00030	"	0.00036	"	0.00001
	Haddock	1	"	0.00011	0.00052	"	0.00054	"	ND
	<i>Nephrops</i>	1	"	0.00080	0.0038	"	0.0060	0.00015	0.00010
Mid North Sea ¹	Mussels	1	"	0.0045	0.027	"	0.0067	ND	ND
	Mussels ⁹	1	"	0.00025	0.0044	"	0.0021	"	"
Southern North Sea ¹	Mussels	1	"	0.0026	0.015	"	0.0047	"	"
	Cockles	1	"	0.0020	0.0083	"	0.0094	"	0.00091
	Cockles ¹⁰	1	"	0.0018	0.0062	"	0.0065	"	0.00051
Icelandic processed	Cod	1	"	0.00005	0.00024	"	0.00022	"	ND

ND = not detected

NA = not analysed

¹ Sampling area

² Landing point

³ Samples collected by Consumer 116

⁴ Samples collected by Consumer 460

⁵ Samples collected by Consumer 311

⁶ Samples provided by Fisherman A

⁷ Samples provided by Fisherman B

⁸ Concentrations refer to weight as supplied

⁹ Landed in Denmark

¹⁰ Landed in The Netherlands

Table 12. Estimates of concentration factors for several fission product nuclides (from: Preston and Jefferies, 1969(a))

Sample	¹⁰⁶ Ru	⁹⁵ Zr/ ⁹⁵ Nb	¹³⁷ Cs	⁹⁰ Sr
<i>Fucus vesiculosus</i>	330	1700	100	6
<i>Porphyra</i>	1800	420	-	1
Mussels	2000	950	15	8
Winkles	2000	1000	100	17
Limpets	1200	750	70	15
Lobster	25	10	25	-
Shrimps (whole)	600	200	100	-
Plaice	10	-	45	0.3
Skate	1	-	25	0.3

6.2 Neutron-activation products

Neutron activation products of transition element nuclides have not received detailed study, although ⁶⁰Co is detectable in the livers of plaice taken off Sellafield, and ^{110m}Ag is detectable in the livers of thornback rays (Pentreath, 1977) and shellfish (MAFF, 1990). Laboratory studies on the accumulation of nuclides of Zn, Mn, Co, Fe and Ag by plaice and rays have been made (Pentreath, 1973(a),(b),(c), 1976(b), 1977).

6.3 Transuranium nuclides

It is the transuranium nuclides which have received the most detailed research attention in recent years. Apart from the routine monitoring data, an early assessment of the distributions of ^{239,240}Pu in various samples was made by Hetherington *et al.* (1975, 1976), the latter paper including some detailed tissue analyses of Pu and Am in crabs (*Cancer pagurus*) and plaice. A detailed study of ^{239,240}Pu and ²⁴¹Am in plaice was made from February 1975 to February 1977, and some ²⁴²Cm and ^{243,244}Cm data have been reported (Pentreath and Lovett, 1976, 1977, 1978). The study concluded that ²⁴¹Am appeared to be more available to the plaice than ^{239,240}Pu, and the data were discussed both in terms of concentrations relative to sea water and in relation to the rates of discharge. Laboratory studies were also made on the accumulation of ^{239,240}Pu by the plaice (Pentreath, 1978(a); Leonard and Pentreath, 1981) and on the thornback ray (Pentreath, 1978(b)). The accumulation of ^{239,240}Pu by plaice eggs was also studied (Hetherington *et al.*, 1976). It was generally concluded that neither Pu nor Am was highly accumulated by benthic or pelagic fish, and that the concentrations observed in the muscle of Irish Sea fish were partly dependent upon the origin of the samples and the method of preparation for analysis (Pentreath *et al.*, 1979).

It had long been clear that the primary pathway of transuranium nuclides to man would be through the consumption of invertebrates (or algae) rather than via

the consumption of fish. Detailed analyses have been made of Pu and Am in edible shellfish, and in other invertebrates in the Sellafield area, by Pentreath *et al.* (1980(a), 1982), Pentreath (1981(a),(b)), Hamilton and Clifton (1980), Clifton *et al.* (1983), Thompson *et al.* (1982) and, as reported in DFR's annual Aquatic Environment Monitoring Reports (Hunt, 1979-1985(a), 1986-1989; MAFF, 1990, 1992, in press). Concentrations of ²³⁸Pu and ^{239,240}Pu in *Mytilus edulis* have been reported for the east coast of Ireland (Crowley *et al.*, 1990). Laboratory experiments have been carried out on ²³⁷Pu accumulation by the edible winkle (*Littorina littorea*) from labelled sea water, food (seaweed) and silt (Swift and Pentreath, 1988) and the results compared with environmental data from the Irish Sea. CFs, biological half-times, transfer factors and depuration rates were estimated. The food pathway was the main route for Pu accumulation with the ingestion of silt playing a critical role in determining the final flesh burden. The accumulation of ²³⁷Pu by the European lobster (*Homarus gammarus* L.), from labelled sea water and food, has been investigated experimentally (Swift, 1992). Organ/tissue concentrations and CF values for individual lobsters reflect both the time since their last moult and their exposure time to ²³⁷Pu. This precludes the calculation of mean whole-body, steady state CF values. Most of the Pu is retained in the hepatopancreas. The presence of ²³⁷Np in biological samples from the Sellafield area has also been described (Pentreath and Harvey, 1981; Pentreath, 1981(a),(b); Pentreath *et al.*, 1982), with some experimental studies on uptake of ²³⁵Np by the plaice (*Pleuronectes platessa*) being reported (Swift, in press (b)).

Studies have been made on the accumulation of U, Pu and Am in *Mytilus edulis* from the Esk Estuary, both in relation to the particulate matter in the area - including 'hot' particles - and to their location within the estuary (Hamilton, 1980; Hamilton and Clifton, 1980; Clifton *et al.*, 1983). Assessments of biological half-times were made by transplant experiments. Values of >200 days were recorded for Pu, Am and ¹⁴⁴Ce, 40 days for ¹³⁷Cs and 260 days for the 'long' component of ¹⁰⁶Ru excretion. These data highlight the difficulties of deriving CF data from few observations. Without a knowledge of turnover times, changes in discharge rates, and other detailed environmental information, it is not possible to model adequately the relationship between the fauna/flora and the ambient water in order to assess steady state CFs. The detailed chemical analysis required limits the number of data which can reasonably be obtained, however, and the data which have been published on concentration factors from the Irish Sea are usually accompanied by comments on the need for caution in their interpretation. Some of these data are presented in Tables 12 and 13 and this need for caution in their interpretation needs to be stressed yet again, particularly as the concept of steady state, when applied to biological systems, may be difficult to justify.

Table 13. Preliminary concentration factor data for transuranium nuclides in the Irish Sea, calculated relative to filtrate (<0.22 µm) sea water. These data were derived from a single sampling of biological materials and sea water in June 1980. It is not known if they represent equilibrium conditions. (Errors are based on ± 2 σ propagated counting errors) (from: Pentreath et al., 1982)

	^{239,240} Pu	²⁴¹ Am	^{243/244} Cm	²³⁷ Np
St. Bees Head				
<i>Ascophyllum nodosum</i>	7.5 x 10 ³ ± 0.5 x 10 ³	9.4 x 10 ³ ± 0.7 x 10 ³	1.7 x 10 ⁴ ± 1.3 x 10 ⁴	2.5 x 10 ± 0.8 x 10
<i>Fucus spiralis</i>	5.7 x 10 ³ ± 0.3 x 10 ³	9.2 x 10 ³ ± 0.6 x 10 ³	1.6 x 10 ⁴ ± 0.9 x 10 ⁴	6.0 x 10 ± 2.7 x 10
<i>Fucus vesiculosus</i>	4.0 x 10 ³ ± 0.3 x 10 ³	8.2 x 10 ³ ± 0.7 x 10 ³	8.4 x 10 ³ ± 8.0 x 10 ³	2.5 x 10 ± 0.8 x 10
<i>Fucus serratus</i>	6.7 x 10 ³ ± 0.4 x 10 ³	9.7 x 10 ³ ± 0.7 x 10 ³	9.3 x 10 ³ ± 7.1 x 10 ³	2.0 x 10 ± 0.9 x 10
<i>Patella vulgata</i> (soft parts)	7.0 x 10 ³ ± 0.5 x 10 ³	4.1 x 10 ⁴ ± 0.3 x 10 ⁴	5.7 x 10 ⁴ ± 3.6 x 10 ⁴	8.9 x 10 ² ± 2.9 x 10 ²
<i>Patella vulgata</i> (shell)	2.2 x 10 ³ ± 0.2 x 10 ³	2.5 x 10 ⁴ ± 0.2 x 10 ⁴	3.4 x 10 ⁴ ± 2.2 x 10 ⁴	5.5 x 10 ² ± 2.0 x 10 ²
<i>Nucella lapillus</i> (soft parts)	3.3 x 10 ³ ± 0.2 x 10 ³	1.4 x 10 ⁴ ± 0.9 x 10 ³	3.1 x 10 ⁴ ± 1.8 x 10 ⁴	5.8 x 10 ² ± 1.9 x 10 ²
<i>Nucella lapillus</i> (shell)	1.8 x 10 ³ ± 0.1 x 10 ³	3.3 x 10 ⁴ ± 0.2 x 10 ⁴	3.6 x 10 ⁴ ± 2.3 x 10 ⁴	2.4 x 10 ² ± 0.8 x 10 ²
Balcary Point				
<i>Ascophyllum nodosum</i>	2.8 x 10 ³ ± 0.2 x 10 ³	5.8 x 10 ³ ± 0.6 x 10 ³	7.3 x 10 ³ ± 7.6 x 10 ³	1.5 x 10 ± 0.7 x 10
<i>Fucus serratus</i>	4.5 x 10 ³ ± 0.3 x 10 ³	9.7 x 10 ³ ± 0.9 x 10 ³	1.1 x 10 ⁴ ± 1.0 x 10 ⁴	2.7 x 10 ± 1.0 x 10
<i>Patella vulgata</i> (soft parts)	6.5 x 10 ² ± 0.6 x 10 ²	5.9 x 10 ³ ± 0.6 x 10 ³	- 2.6 x 10 - ± 1.0 x 10	
<i>Patella edulis</i> (soft parts)	1.1 x 10 ³ ± 0.1 x 10 ³	1.3 x 10 ⁴ ± 0.1 x 10 ⁴	1.5 x 10 ⁴ ± 1.4 x 10 ⁴	1.8 x 10 ² ± 0.8 x 10 ²
<i>Mytilus edulis</i> (shell)	1.0 x 10 ³ ± 0.8 x 10 ²	9.8 x 10 ³ ± 0.9 x 10 ³	1.7 x 10 ⁴ ± 1.6 x 10 ⁴	3.1 x 10 ² ± 1.3 x 10 ²

6.4 Relating concentrations to rates of discharge

In view of the fluctuating nature of the Sellafield discharges, and the local hydrographic conditions, it has always been difficult to describe the accumulation of radionuclides in the local fauna, with any degree of certainty, in terms of concentration factors. This is particularly difficult for the longer-lived radionuclides in the mobile fauna because there is insufficient knowledge of the biological half-times required to relate these to ambient concentrations. From the radiological aspect this is not important because samples of marine foodstuffs are monitored directly. Nevertheless, the Irish Sea is generally regarded as a useful source of data on a wide range of radionuclides,

and concentration factors are sometimes required when comparing sites. The long term relationship between ¹⁰⁶Ru in algae, particularly *Porphyra*, and the rate of discharge has been studied for many years (Preston and Jefferies, 1969(a); Pentreath, 1980(a), 1982; Preston and Portmann, 1981). The consistency of the relationship is quite remarkable, even close to the discharge point (Figure 14(a)). The concentrations of radionuclides in fish, however, would be expected to vary considerably from month to month, and from year to year, if only because of their mobility. But, relating concentrations in locally caught plaice to discharge rates does give useful data (Figure 14(b)) and average values remain consistent over long periods (Pentreath, 1980(a), 1982; Pentreath and Lovett, 1978; Preston and Portmann, 1981).

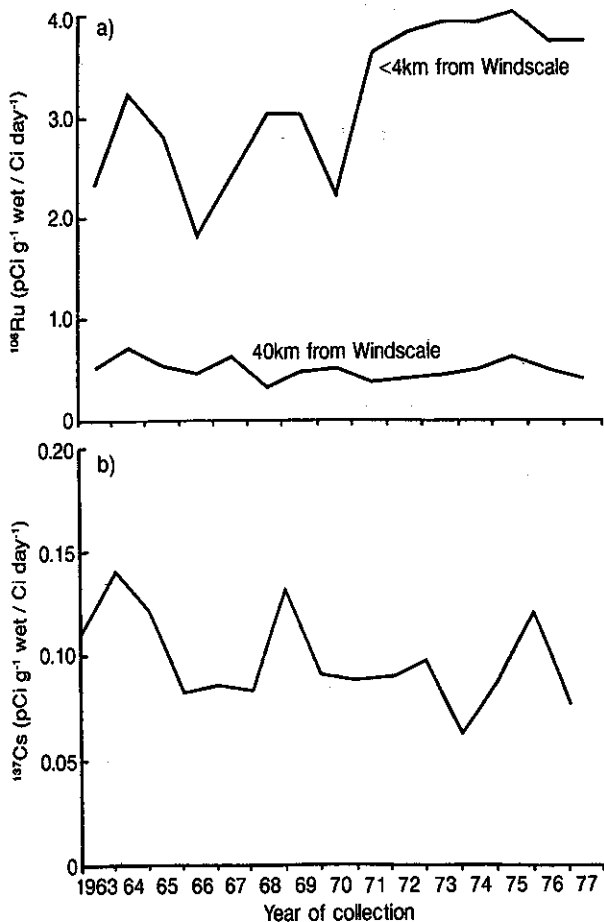


Figure 14. Concentration of (a) ^{106}Ru in *Porphyra*, and (b) ^{137}Cs in the flesh of plaice (*Pleuronectes platessa*), caught within the vicinity of Sellafield and normalised to a discharge rate of 1 Ci day^{-1} respectively (from: Pentreath, 1980(a))

7. NATURALLY-OCCURRING RADIONUCLIDES

7.1 Background information

Naturally-occurring radionuclides may be divided into two groups: primordial, having half-lives within an order of magnitude of the age of the earth, and their daughter products (e.g. ^{40}K , ^{87}Rb , ^{238}U -, ^{235}U - and ^{232}Th -decay series); and cosmogenic, produced continuously in the atmosphere by the action of cosmic rays (e.g. ^3H , ^7Be , ^{10}Be , ^{14}C , ^{26}Al , ^{32}Si , ^{53}Mn). Their differing chemical properties, half-lives and sources result in a non-uniform distribution in the marine environment and provide excellent 'chronometers' of key biogeochemical processes.

7.2 Sediments and sea water

The usual content of naturally-occurring radionuclides in sea water is about 12 Bq l^{-1} . This is mainly due to ^{40}K but there are appreciable quantities of other radionuclides, including ^{87}Rb ($\sim 0.1 \text{ Bq l}^{-1}$) and members of the uranium and thorium decay-series ($\sim 0.1 \text{ Bq l}^{-1}$). [In simplistic terms this can be compared to a mean concentration of Sellafield radionuclides in the Irish Sea of about 2 Bq l^{-1} , in 1988. However, the artificial radionuclides are not uniformly distributed spatially or temporally.] Concentrations of U and Th in oceanic sands and clays are typically within the ranges 1 to 4 and 1 to 30 ppm respectively (Ivanovich and Harmon, 1982). Concentrations of the principal members of the U- and Th-decay series, associated with Irish Sea sediments and sea water, have been reported by Kershaw *et al.* (1988(c)) and McCartney *et al.*, (1990) (Table 14). Both studies pointed out the influence of grain size and water depth on radionuclide concentrations. Mineralogy will also influence the concentrations of lattice-bound radionuclides. The differing half-lives and chemical properties of the ^{238}U decay-series radionuclides, specifically $^{210}\text{Pb}/^{226}\text{Ra}$ and $^{234}\text{Th}/^{238}\text{U}$, have been utilised to identify and quantify key sediment processes in the Irish Sea, such as scavenging bioturbation and accumulation rates (subsection 5.3).

There is evidence of additional, anthropogenic sources of naturally-occurring radionuclides in the region. Discharges of U and Th are made by pipeline to the Ribble Estuary from the BNFL Springfields' fuel fabrication plant. Public radiation exposure is low, (Doddington *et al.*, 1990) but very high concentrations occur in muddy sediments near the pipeline outlet (e.g. total β , 33 kBq ; ^{230}Th , 290 Bq ; $^{234\text{m}}\text{Pa}$, 75 kBq kg^{-1} (dry); Hunt, 1989). At present, the extent to which these inputs are exported from the estuary has not been quantified but work is in progress to address this question and to study transport within the estuary. It is evident, from measurements of the $^{230}\text{Th}/^{232}\text{Th}$ ratio, that some export does occur (P. J. Kershaw, unpublished).

An average of 4 tonnes year^{-1} of uranium have been discharged from Sellafield, and a slight enhancement in the ^{238}U concentration in sea water, over that expected from the salinity determination, has been detected near the pipeline (Kershaw and Young, 1988). Hamilton (1980) found no evidence of excessive enhancement of uranium in sediments, sea water or mussels as a result of the Sellafield discharges. How-

Table 14. A comparison of natural radionuclide activities ($Bq\ kg^{-1}$ dry) in the surface sediments of Whitehaven Harbour and the Irish Sea (from: McCartney *et al.*, 1990)

Location	^{238}U	^{235}U	^{234}U	^{232}Th	^{230}Th	^{228}Th
Whitehaven harbour	300-500	13-20	300-500	10-30	500-700	10-30
Irish Sea	1-20	<1	1-20	5-30	1-100	5-30

ever, Hamilton and Stevens (1985) were able to detect Sellafield-derived uranium in the sediments of the Esk Estuary, on the basis of ^{235}U depletion and the presence of ^{236}U , but not in subtidal sediments collected near Whitehaven or off the Sellafield plant. They concluded that the contribution from Sellafield would not be detected in sea water, assuming the normal conservative transport of uranium based on the dispersion of ^{137}Cs in the Irish Sea (Jefferies *et al.*, 1982).

The third significant source of naturally-occurring radionuclides has been from a chemical plant near Whitehaven (Marchon), which formerly processed phosphate ore to produce phosphoric acid. The plant ceased the importation of phosphate ore in July 1992 and introduced a new waste treatment process. These measures have resulted in very substantial decreases in the quantities of both radionuclides and heavy metals discharged. Phosphate ore contains relatively high concentrations of uranium. Discharges of uranium have been estimated to be about 35 tonnes year⁻¹ (McCartney *et al.*, 1990). Unlike Sellafield, the Marchon discharges are thought to contain substantial quantities of daughter nuclides, although the precise amounts are unknown. Significant enhancements (~ factor of 3) of dissolved ^{226}Ra have been observed in sea water in the near-shore zone, with the highest concentration (34m Bq l⁻¹) being measured in Whitehaven harbour where the ore is off-loaded (McCartney *et al.*, 1990). The sediments in the harbour have significantly higher concentrations of ^{238}U and ^{235}U decay-series radionuclides, but not of ^{232}Th when compared with equivalent offshore sediments (Table 14). This enhancement is observed in nearshore sediments adjacent to the pipeline, with relatively high sea-bed

inventories of ^{210}Pb (e.g. 14 kBq m⁻²) and $^{230}Th/^{232}Th$ ratios greater than unity. The temporal variability in ^{210}Pb and ^{226}Ra , as deduced from a dated sediment core, correlate well with the discharge data provided by the operators. Similar localised inputs, from phosphogypsum plants, have been observed in the Rhine and Scheldt estuaries (Pennders *et al.*, in press).

The combined inputs of Marchon and Springfields appear to have resulted in a significant enhancement of ^{230}Th in an approximately 25 km wide coastal zone running along the coast of Cumbria and Lancashire, from Maryport to Liverpool Bay (A. J. Poole, pers. comm.).

7.3 Biota

Naturally-occurring radionuclides are accumulated by marine biota to differing extents. ^{210}Po is the most important nuclide from a radiological viewpoint (section 8). Concentrations of ^{210}Po in fish are low (generally <10 Bq kg⁻¹) whereas those in crustaceans and molluscs are somewhat higher (10-50 Bq kg⁻¹) (Camplin and Aarkrog, 1989). The concentrations of naturally-occurring radionuclides in fish and shellfish from the Irish Sea, and elsewhere in UK coastal waters, have been reported by Pentreath and co-workers (Pentreath *et al.*, 1979, 1980; 1989(a); Pentreath and Allington, 1988) (Table 15). Concentrations of ^{210}Po in winkles collected from Saltom Bay and Parton, in the Whitehaven vicinity, are significantly higher than those in winkles from the Sellafield area (Tables 15 and 16), an increase which can be attributed to the Marchon phosphogypsum discharge. Similar enhancements have been observed in Dutch waters (van der Heijde *et al.* 1988).

Table 15. Concentrations of ^{210}Po (Bq kg^{-1} wet) in shellfish taken around the UK (from: Pentreath et al., 1989(a))

Material	Location	Date	No. of samples	^{210}Po
Molluscs				
Mussel flesh (<i>Mytilus edulis</i>)	Newlyn	Oct '87	2	18.5
	Lowestoft	June '86	3	4.9
	Lowestoft	Sept '87	3	23.2
	Carbis Bay	Aug '88	1	14.2
	Portling Bay	Aug '88	1	24.2
Winkle flesh (<i>Littorina littorea</i>)	Newlyn	Nov '87	2	7.6
	Cromer	July '87	3	7.1
	Saltom Bay	May '88	1	251.3
Crustaceans				
Brown crabmeat (<i>Cancer pagurus</i>)	Newlyn	Oct '87	2	47.5
	Cromer	July '86	4	4.1
White crabmeat (<i>Cancer pagurus</i>)	Newlyn	Oct '87	2	1.4
	Cromer	July '86	4	0.2
<i>Nephrops</i> tail* (<i>Nephrops norvegicus</i>)	North Shields	Feb '87	3	2.1
Shrimp tail* (<i>Crangon crangon</i>)	Lowestoft	Oct '88	1	1.2

* The digestive glands of the *Nephrops* and shrimp samples contained 453 and 927 Bq Kg^{-1} (wet) of ^{210}Po in their respective digestive gland tissue; this is not usually deliberately eaten but may affect the concentration in the fraction of the animal actually consumed, particularly for shrimps

Table 16. Concentration of ^{210}Po in shellfish taken from the Sellafield area (from: Pentreath et al., 1989(a))

Material	Location	Date	^{210}Po (Bq kg^{-1} wet)
Winkle flesh (<i>Littorina littorea</i>)	Sellafield	Apr '88	16.7
	Parton	Apr '88	54.0
Crab meat white + brown (<i>Cancer pagurus</i>)	Sellafield	May '88	14.0
	Parton	May '88	34.5

8. ENVIRONMENTAL MONITORING, SURVEILLANCE, AND ASSESSMENT OF RADIATION EXPOSURE OF THE PUBLIC

8.1 Exposure pathways and doses to individuals

The discharges from nuclear sites, including those from Sellafield, are subject to strict authorisation and environmental surveillance by UK Government departments. These measures are implemented in order to safeguard the public, based on the recommendations of the ICRP and the advice of national bodies on

radiological protection, a role played in the UK since 1970 by the National Radiological Protection Board (NRPB). The changes in the nature and relative importance of particular exposure pathways during the past four decades are summarised below. Dose limits for exposure of individuals and critical groups, and the assumptions made in calculating these limits, have been subject to review (ICRP, 1977, 1984, 1985, 1986, 1987, 1991; NRPB, 1987). MAFF (1992) provides a summary of the changes which have been made, or which are contemplated. The ICRP dose limitation system includes the requirement that 'all exposures shall be kept as low as reasonable achievable ...' (ALARA). This applies both to individual and collective doses in radiological control procedures. The remainder of this section covers, in some detail, the identification and monitoring of critical groups, and the estimation of doses, and how these have changed with time.

Annual reports of monitoring and surveillance programmes in the Sellafield area, and elsewhere in the UK coastal environment, have been published by MAFF since 1967 (Mitchell, 1967-1971(a)(b), 1973-1977(a)(b); Hunt, 1979-1985(a), 1986-1989; MAFF, 1990, 1992, in press). An annual account of discharges and environmental surveillance is also published by the operators (Howells, 1975(a),(b), 1976, 1977; Atherton, 1978; BNFL, 1979-1989). The first MAFF report (Mitchell, 1967) covered the period 1963 to 1966. Data were presented on the relationship of ^{106}Ru in *Porphyra* with distance from Sellafield and on ^{90}Sr and ^{144}Ce in *Porphyra*. Samples of various fish species, invertebrates, benthic algae, sea water and sediments were analysed by gamma-spectrometry and total β methods. At that time, the critical pathway was the consumption of the foodstuff laverbread, which is manufactured particularly in the Swansea area of South Wales. Laverbread is produced from the alga *Porphyra*, some of which, up to 1972, was harvested from the coast near Sellafield. The critical group was identified as being laverbread consumers, and typically were from the Swansea area. The results of a detailed habits survey of this pathway were published by Preston and Jefferies (1967), and covered the period 1963 to 1965. With the exception of 1959, an adult consumption rate of 75 g day⁻¹ of laverbread was estimated as delivering less than 5 mGy year⁻¹ to the large intestine, and between 5 and 10 mGy during 1959. From the results of a more detailed study (Preston and Jefferies, 1969(b)), it was estimated that a re-defined critical group had a median consumption rate of 160 g day⁻¹, resulting in doses of 4 to 7 mSv year⁻¹ to the lower large intestine of adults during the period 1962 to 1967. In the report for 1967 (Mitchell, 1968), data were presented of the results of monitoring *Porphyra* and of laverbread samples from the principal manufacturers. External dose-rates to a sub-critical group in the Ravenglass estuary were estimated by taking measurements of gamma-dose rates at selected sites. These data had been related to the radionuclide concentrations of the sediments (Jefferies, 1968). At that time, fish consumption was recognised as a relatively unimportant pathway of human radiation exposure, although samples were analysed, as were a large number of 'indicator' samples, particularly the *Fucus* algae.

In 1968 (Mitchell, 1969), *Porphyra* consumption remained the dominant pathway, and the alpha-radionuclide content of this seaweed was beginning to be studied. The situation remained much the same in 1969, with *Porphyra* consumption in South Wales being the critical pathway and the external dose rate to the public from contaminated sediment in the Esk Estuary providing a secondary pathway of importance (Mitchell, 1971(a)). But, in 1970, a third exposure pathway became evident as the concentrations of ^{134}Cs

and ^{137}Cs in fish responded to the increased discharges (Mitchell, 1971(b)). All three pathways were examined during 1971, and *Porphyra* consumption was found to dominate. The highest external dose-rate estimate for the Esk Estuary, for a salmon fisherman working nets in the area, was 11% of the ICRP limit at that time for the public (Mitchell, 1973).

In 1972, the collection of *Porphyra* along that part of the coastline was discontinued, because of transport difficulties, and the importance of this pathway diminished forthwith (Pentreath, 1980(b)). The experience gained from this pathway, particularly with regard to the identification and characterisation of critical groups at that time, has been discussed by Preston (1971), Preston and Mitchell (1973) and Preston *et al.*, (1974). Due to the widespread distribution of radionuclides throughout the Irish Sea, *Porphyra* collected elsewhere contains some ^{106}Ru , and other radionuclides, and laverbread has continued to be monitored and the dose to the public assessed. However, because there is no reason to assume that *Porphyra* growing close to Sellafield may not be harvested at some time in the future for laverbread production it, too, is still routinely monitored and its potential as a pathway, is kept under review. Discharges of ^{106}Ru have decreased markedly since the mid-1970s, reducing the potential importance of this pathway.

In 1972/73 the external exposure to individuals working in the Esk Estuary was the most important pathway (Mitchell, 1975) but, by 1974 (Hetherington, 1976(b)), the consumption of fish and shellfish had become the dominant pathway due to the increasing discharges of ^{134}Cs and ^{137}Cs . The maximum consumers of a critical group of fish and shellfish eaters (265 g day⁻¹) were estimated to be receiving just under 14% of the ICRP dose limit for members of the public at that time (5 mSv year⁻¹; this limit has since been revised, downwards; see below). This figure was predominantly a result of Cs nuclides with ^{106}Ru , $^{239,240}\text{Pu}$ and ^{241}Am collectively accounting for less than 2.5% of the limit. In 1975 and 1976 (Mitchell, 1977(a),(b)) the situation was similar to that of previous years with regard to the critical pathways but, due to the increasing discharges, the maximum consumers of a critical group consuming fish and shellfish were estimated to receive 34 and 44% respectively of the ICRP limit, again largely the result of the Cs nuclides. More detailed analysis of the fish/shellfish consumption pathways were reported, comparing the average and maximum consumers of locally caught sea food with the average consumers of commercial fish landed at the nearest fishing ports of Whitehaven and Fleetwood, and with average consumers amongst the public in general.

The estimated percentages of the ICRP dose limit (5 mSv year⁻¹, at that time) for members of the public during the period 1970 to 1976 are given in Table 17, taken from the publications cited above. From 1977 onwards, the dose estimates have been calculated on the basis of ICRP-26 (i.e. as effective dose equivalents). The data obtained are not, therefore, strictly comparable with those derived earlier although, for the years 1977 and 1978, estimates based on both ICRP-9 and ICRP-26 were made. In 1977, the maximum consumers of plaice and crab in the local fishing community (at 224 g and 41 g day⁻¹ respectively) were estimated to be receiving 31% of the then ICRP effective dose equivalent limit for the public, at these consumption rates, the major fraction arising from ¹³⁷Cs (Hunt, 1979). A separate estimate was made for consumers of winkles (*Littorina littorea*), who were estimated as receiving 11% of the ICRP effective dose equivalent at a consumption rate of 5 g day⁻¹ - primarily from the ²⁴¹Am, ¹⁰⁶Ru and Pu isotope content. A third group, those associated with commercial fisheries along the coast, primarily around Whitehaven, Fleetwood and in Morecambe Bay, consumed greater quantities of sea food (290 g, 70 g and 45 g day⁻¹ respectively of fish, crustaceans and molluscs) but received a percentage of the ICRP-recommended limit similar to that of the local fishing community, because the food contained lower concentrations of radionuclides, having been caught at greater distances from the discharge point.

Table 17. Maximum exposures, as a percentage of ICRP-9 limits for members of the public (5 mSv year⁻¹) resulting from Sellafield discharges from 1970 to 1976. (For references, see text)

Year	Porphyral laverbread (Critical group)	External exposure (Ravenglass)	Fish consumption (Critical group)
1970	5	12	1
1971	33	11	3
1972/3	2	7	3
1974	< 0.2	7	14
1975	< 0.2	9	34
1976	0.2	8	44

In 1978, further habit survey data indicated that, using ICRP-recommended techniques (Hunt and Shepherd, 1980), the consumption rates in the local fishing community were somewhat lower than those in 1977, but those of consumers associated with commercial fisheries were somewhat higher. At the new consumption rates, the two groups received 25% and 15% respectively of the appropriate effective dose equivalent limit, with ¹³⁷Cs being the dominant radionuclide (Hunt, 1980). In 1979, at the same consumption rates, the results were similar to those of the previous year (Hunt, 1981), but for the 1980 estimates the situation had been re-evaluated, with changes having been made

in the techniques of habit-survey data collection (Hunt, 1982; Leonard *et al.*, 1982; Hunt *et al.*, 1982). The revised data indicated that the consumption rates appropriate to the local fishing community were 100 g of fish and 18 g each of crustaceans and molluscs per day. On the basis of the then current ICRP recommendations, the results indicated that this group of people were receiving 24% of the appropriate effective dose equivalent limit, with 8% arising from ¹³⁷Cs; but allowing for advice from the NRPB, to the effect that Pu in low concentrations in food could be absorbed by a factor of 5 greater than that assumed in ICRP-30, the predominant nuclides were ^{239,240}Pu, contributing 9.5% of a revised total of 39% of the limit.

By the time of the 1981 report (Hunt, 1983), the situation had been reviewed yet again. A further study had been made which, although indicating that the range of consumption rates was similar to that observed previously, identified an effectively new critical group with an average mollusc consumption rate of 45 g day⁻¹. The consumption rates for fish and crustaceans were considered to be essentially unaltered. The revised molluscan consumption rate was thought to be partly due to the general effects of the economic recession, in which some people had more time to exploit a food supply that was freely available, and thus could not be applied retrospectively prior to 1981 with any degree of confidence. Even so, the substantially increased molluscan contribution indicated that this group received 46% of the annual 5 mSv ICRP-recommended effective dose equivalent, of which the major contributor (20%) was ²⁴¹Am. If allowance is made for possible increased Pu absorption, as advised by the NRPB, the total dose estimate increases to 69% of the appropriate limit (Table 18).

The relative contribution of nuclides with long body-retention times needs to be carefully considered, however, because the percentile of the effective dose equivalent limit, which is calculated in any one year, would only be received in the 50th year following continued consumption at the rate calculated for that year, assuming similar concentrations of the nuclide in the foodstuff.

The relative importance of transuranium nuclides in the critical pathways increased in the 1980s for a number of reasons. Their concentrations in fish flesh are low (Pentreath *et al.*, 1979), but they are accumulated by molluscan shellfish (Pentreath, 1981(a)). The importance of ¹³⁷Cs has also decreased, the quantities discharged having been substantially reduced by the use of zeolite, placed in skips in the cooling ponds, as an interim measure, before the new ion exchange effluent treatment plant (SIXEP) became fully commissioned in 1985. The implementation of this procedure was described by Handyside *et al.*, (1982) as an example of ICRP-26 optimisation techniques in practice.

Table 18. Individual radiation exposures due to consumption of Irish Sea fish and shellfish, 1981 (from: Hunt, 1983)

Exposed population	Consumption rate used in assessment	Nuclide	Effective dose equivalent (as % of ICRP-recommended dose limit of 5 mSv year ⁻¹ for members of the public)	
			On basis of current ICRP recommendations	Effect of Pu enhanced by a factor of 5
Consumers in local fishing community	100 g d ⁻¹ fish } 18 g d ⁻¹ crustaceans } 45 g d ⁻¹ molluscs }	⁹⁰ Sr	0.3	0.3
		¹⁰⁶ Ru	9.6	9.6
		¹³⁴ Cs	0.7	0.7
		¹³⁷ Cs	8.6	8.6
		²³⁸ Pu	0.7	3.4
		^{239,240} Pu	2.9	14.5
		²⁴¹ Pu	2.3	11.5
		²⁴¹ Am	20.2	20.2
		Total	46	69
Consumers in local fishing community	100 g d ⁻¹ fish } 18 g d ⁻¹ crustaceans } 18 g d ⁻¹ molluscs }	⁹⁰ Sr	0.3	0.3
		¹⁰⁶ Ru	3.9	3.9
		¹³⁴ Cs	0.7	0.7
		¹³⁷ Cs	7.7	7.7
		²³⁸ Pu	0.3	1.4
		^{239,240} Pu	1.2	6.0
		²⁴¹ Pu	1.0	4.8
		²⁴¹ Am	8.7	8.7
		Total	24	34
Consumers associated with commercial fisheries (Whitehaven, Fleetwood, Morecambe Bay)	360 g d ⁻¹ fish } 70 g d ⁻¹ crustaceans } 50 g d ⁻¹ molluscs }	¹³⁴ Cs	0.3	0.3
		¹⁰⁶ Ru	0.1	0.1
		¹³⁴ Cs	1.0	1.0
		¹³⁷ Cs	11.7	11.7
		²³⁸ Pu	0.1	0.3
		^{239,240} Pu	0.4	1.8
		²⁴¹ Pu	0.3	1.3
		²⁴¹ Am	2.0	2.0
		Total	16	19

Lower concentrations of Cs in fish and ¹⁰⁶Ru and the transuranic nuclides in winkles in 1982 were responsible for a decrease in the critical group dose to 54% of the ICRP limit. Doses, in general, have continued to decline (e.g. Figure 19) following the sustained reduction in the quantities discharged by Sellafield (Figure 2; section 2). Concentrations of ¹³⁴Cs increased significantly in May 1986, in nearshore waters and marine biota, as a result of fallout from the Chernobyl reactor accident. However, concentrations declined rapidly and the resulting doses were very low (Camplin *et al.*, 1986; Mitchell and Steele, 1988).

Consumption rates of the Cumbrian coastal community have been kept under review (Leonard *et al.*, 1982; Leonard, 1984; Hunt *et al.*, 1982; Hunt, 1983) and note has been taken of the changes in recommendations regarding dose limits by the ICRP and the NRPB. The ICRP has recommended the adoption of a principal dose limit for a member of the public of 1 mSv year⁻¹ (ICRP, 1985). However, it considered that it was

permissible to use a subsidiary dose limit of 5 mSv year⁻¹, for a limited number of years, provided that the average annual committed effective dose equivalent over a lifetime did not exceed the principal dose limit. Exposures have been within the 1 mSv year⁻¹ limit in recent years and it is considered that they are likely to remain so, given the continuing decline in discharges, unless there is a substantial increase in shellfish consumption. Past exposures have been above this level but not for long enough for lifetime exposure to have exceeded, on average, 1 mSv year⁻¹. It follows that non-stochastic effects will also be avoided (Hunt, 1989).

A significant factor in the calculation of doses has been the choice of gut transfer factors for Pu and Am. A cautious, generic value of 1×10^{-3} is recommended by the ICRP (ICRP, 1986), although it is acknowledged that this may not be appropriate when a best estimate of absorption is required. Experimental work, using adult human volunteers consuming winkles collected

from the Sellafield area, suggests that a lower gut transfer, of 2×10^{-4} , is more realistic (Hunt *et al.*, 1986, 1990). Both this value and the more conservative value of 5×10^{-4} , recommended by the NRPB (NRPB, 1990), are used in current assessments of exposure (Table 19).

By the late 1980s the external exposure pathway was perceived to be the critical pathway resulting in the highest radiation dose. External radiation exposure, due to man-made radionuclides, can result from a wide range of recreational and occupational activities. Recreational activities include bait digging for angling, wildfowling, living on houseboats, amateur small boat maintenance, coal picking and beach walking. Occupational activities include fishing, farming, commercial bait digging, turf cutting, boat and river maintenance work, and policing of nature reserves and fisheries. The methodology used, critical groups identified and doses estimated have been described in detail (Jefferies, 1970; Hunt and Jefferies, 1981; Hunt, 1979-1985(a), 1986-1989; MAFF, 1990, 1992 in press; Doddington *et al.*, 1989, 1990; Emptage and Kelly, 1990). The most significant doses result from boat occupancy with critical groups in the Wyre and Ribble estuaries receiving a mean dose of 0.14 mSv and 0.17 mSv year⁻¹ respectively in 1989, corresponding to occupancy rates of 2300 and 3000 h year⁻¹ (Doddington

et al., 1990) with the largest contribution being from Sellafield-derived radionuclides. Other important groups include bait diggers in Whitehaven inner harbour (0.07 mSv, 530 h year⁻¹) and a nature warden in the Esk Estuary (0.07 mSv, 320 h year⁻¹). The additional exposure of these people from consumption of fish and shellfish has also been assessed.

An additional pathway to those discussed above is that of inhalation. Inhalation has been allowed for in the calculations made by Hunt and Jefferies (1981), based on information from Knight (1980). More recently, detailed studies have been made of the transfer of radionuclides discharged to sea to the Cumbrian terrestrial environment via aerial pathways (Pattenden, 1981; Pattenden *et al.*, 1980, 1989; Cambray and Eakins, 1982; Peirson *et al.*, 1982). There is some evidence for small quantities of Pu and Am being transferred in this manner. The highest annual average concentration of ^{239,240}Pu observed was 93×10^{-18} Ci kg⁻¹ (3.4 mBq kg⁻¹) in air at Eskmeals in the Ravenglass Estuary, which represents 0.2% of the derived air concentration as modified for the public (Peirson *et al.*, 1982). Recent model calculations indicate that annual doses peaked at 24 µSv year⁻¹ in 1973, at Seascale, reduced to 5 µSv year⁻¹ in the mid-1980s, and are expected to reduce further to 3 µSv year⁻¹ by the year 2000 (Howorth, 1989).

Table 19. Individual radiation exposures due to consumption of Irish Sea fish and shellfish, 1990 (from: MAFF, 1992)

Exposed population	Consumption rate used in assessment (see text), kg year ⁻¹	Nuclide	Exposure, mSv		
			ICRP-26*	ICRP-60#	
Consumers in local fishing community	Fish (plaice and cod)	37	⁹⁰ Sr	0.004	0.003
	Crustaceans (crabs and lobsters)	6.0	¹⁰⁶ Ru	0.005	0.008
	Molluscs (winkles)	8.3	¹³⁷ Cs	0.015	0.015
			²³⁸ Pu	0.007	0.004
			²³⁹⁺²⁴⁰ Pu	0.034	0.021
			²⁴¹ Pu	0.012	0.007
			²⁴¹ Am	0.075	0.045
			Total	0.16	0.11
Consumers associated with commercial fisheries (Whitehaven)	Fish (plaice and cod)	49	⁹⁹ Tc	0.002	0.004
	Crustaceans (<i>Nephrops</i>)	11	¹³⁷ Cs	0.013	0.013
	Molluscs (whelks)	6	²³⁹⁺²⁴⁰ Pu	0.004	0.003
			²⁴¹ Am	0.008	0.005
			Total	0.03	0.03
Consumers in Morecambe Bay area	Fish (flounders and plaice)	54	¹³⁷ Cs	0.035	0.035
	Crustaceans (shrimps)	21	²³⁹⁺²⁴⁰ Pu	0.025	0.014
	Molluscs (cockles and mussels)	23	²⁴¹ Am	0.056	0.033
			Total	0.14	0.1
Consumers associated with commercial fisheries (Fleetwood)	Fish (plaice and cod)	82	¹³⁷ Cs	0.023	0.023
	Crustaceans (shrimps)	17	²³⁹⁺²⁴⁰ Pu	0.022	0.013
	Molluscs (cockles and whelks)	23	²⁴¹ Am	0.053	0.031
			Total	0.11	0.08
Typical member of the fish-eating public consuming fish landed at Whitehaven/Fleetwood	Fish (plaice and cod)	15	¹³⁷ Cs	0.003	0.003
			Total	0.004	0.004

* Committed effective dose equivalent for comparison with current dose limits and criteria
Committed effective dose calculated using methodology of ICRP-60

8.2 Collective dose

The collective committed effective dose equivalent rates arising from the Sellafield discharges have also been evaluated in detail, and the estimates quoted in MAFF's annual Aquatic Environment Monitoring Report Series since 1970 (Hunt, 1970-1985(a), 1986-1989; MAFF, 1990, 1992, in press). The collective dose for any given population may be computed by using fish landing data, compiled by the International Council for the Exploration of the Sea (ICES), together with measurements of radioactivity in fish samples (e.g. Jefferies *et al.*, 1977; Preston *et al.*, 1978). This approach is supplemented by estimating the concentrations in fish in different areas from a knowledge of the water concentrations and the concentration factors for the particular species of fish. The nuclides primarily responsible for collective effective dose equivalents are ^{134}Cs and ^{137}Cs and estimates of these for the period 1974 to 1978 are discussed by Hunt and Jefferies (1981). These data have been corrected, as appropriate, for the collective dose equivalent for the years prior to ICRP-26, and after subtracting the contribution from other radionuclides and from fallout.

Hunt and Jefferies (1981) have, for the purposes of optimisation, used modelling techniques to estimate the collective effective dose equivalent commitment normalised to a unit rate of discharge for ^3H , ^{90}Sr , ^{95}Zr , ^{95}Nb , ^{99}Tc , ^{106}Ru , ^{129}I , ^{134}Cs , ^{137}Cs , ^{144}Ce , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu and ^{241}Am , truncated at 10^4 years. This gives the dose due to discharges in a given year (Table 20). For the 1979 discharges, a collective total detriment commitment of some 200 man-Sv was estimated, of which >85% was due to ^{137}Cs . A compartmental model to calculate collective dose has been described by Camplin *et al.* (1982). The collective dose arising from ^3H has been discussed in a separate paper (Hetherington and Robson, 1979), and the difficulties in assessing the dose commitment arising from discharges of transuranium nuclides are discussed by Pentreath *et al.* (1984). The collective dose to the UK population decreased from 70 to 40 man-Sv in the period 1983 to 1988. The collective dose to other European countries also fell in this period, from 110 to 50 man-Sv, with an increase in 1986 resulting from the contamination of mainly the North Sea, Baltic Sea and Scottish waters following the Chernobyl accident

Table 20. Collective effective dose equivalents from ^{134}Cs and ^{137}Cs in edible fish: Irish Sea, Scottish Waters, North Sea (from: Hunt and Jefferies, 1981)

Year	Collective effective dose equivalent (man-Sv)		Windscale discharge (TBq)				Normalised collective effective dose equivalent on the basis of (c), (man-Sv TBq ⁻¹)	
	United Kingdom	Western Europe	(a) In year of discharge	(b) Average of year and preceding year	(c) Average of year and preceding two years	(d) Average of year and preceding three years	United Kingdom	Western Europe
1974	6.8	12.2	997	581	459	403	0.0148	0.0266
1975	11.7	19.8	1081	1039	748	615	0.0156	0.0265
1976	21.7	38.4	738	910	939	745	0.0231	0.0409
1977	9.9	18.5	594	666	805	853	0.0123	0.0230
1978	11.0	19.5	404	499	579	704	0.0190	0.0337
Coefficient of correlation (r) with collective effective dose equivalent (12%)			0.13	0.54	0.81	0.45	Mean 0.0170	0.0301
			(U.K.)					
			(W.E.)	-0.17	0.51	0.82	Std. error 0.0021 (12%)	0.0036
^{137}Cs								
1974	41	75	4061	2415	2040	1862	0.0201	0.0368
1975	72	122	5231	4646	3353	2837	0.0215	0.0364
1976	111	211	4289	4760	4527	3587	0.0245	0.0466
1977	76	146	4478	4383	4666	4515	0.0163	0.0313
1978	102	191	4088	4236	4285	4522	0.0238	0.0446
Coefficient of correlation (r) with collective effective dose equivalent (8.1%)			0.08	0.79	0.84	0.72	Mean 0.0212	0.0391
			(U.K.)					
			(W.E.)	-0.17	0.75	0.86	Std. error 0.0016 (7.7%)	0.0032

(Hunt, 1985-1989). The contribution due to fallout, from Chernobyl in these waters, excluding the Baltic Sea, was estimated to be about 15 man-Sv to the UK population and 20 man-Sv for other countries in 1986. By 1988, this had fallen to 4 man-Sv and 7 man-Sv respectively. It has been estimated that the collective dose to other European countries from the consumption of Chernobyl-contaminated Baltic Sea fish could have been up to 150 man-Sv in both 1989 and 1990 (MAFF, 1992).

In 1990, the UK collective dose, expressed on a *per caput* basis, through the fish and shellfish pathway, as a result of liquid radioactive waste discharges, amounted to less than 0.01% of the annual dose equivalent, averaged over the population, of 2.2 mSv (Hughes *et al.*, 1988) due to natural background radiation. It should be noted that in neither case is the dose evenly distributed.

9. MODELLING

In addition to the development of several 2-D and 3-D hydrodynamic models of water movement in the Irish Sea (section 3.1) there have been a number of models designed specifically to predict the distribution of Sellafield-derived radionuclides within the Irish Sea and the resulting dose to man.

9.1 Caesium model

A box model has been used to describe the behaviour of Cs in the Irish Sea (Jefferies and Steele, 1989), adapted from earlier versions described elsewhere (Camplin *et al.*, 1982; Grimwood, 1982; Jefferies *et al.*, 1982; Evans, 1985). The model was used to predict sea-bed inventories of ^{137}Cs and to estimate the net flow of sea water through the Irish Sea. It was found necessary to increase the flow from $2.2 \text{ km}^3 \text{ day}^{-1}$ up to September 1976 to $4.4 \text{ km}^3 \text{ day}^{-1}$ thereafter, to maintain a reasonable agreement between predicted and observed concentrations. This change has a degree of support from similar studies in west coast Scottish waters (McKinley *et al.*, 1981; McKay and Baxter, 1985), although no convincing explanations of why such a change occurred have been put forward. We lack the long time-series measurements of physical and chemical properties required to back up such explanations.

9.2 Harwell model

A finite-difference model has been developed to predict marine dispersion as part of the long-running sea-to-land transfer study (sub-section 5.1). The detailed structure has been described in a series of reports (Howorth and Eggleton, 1988(a),(b); Howorth

and Kirby, 1988; Peirson, 1988). Residual flows are treated as advection, whilst tidal mixing is treated as diffusion, with the water column assumed to be well-mixed. The original model was based on a 6 km grid of the eastern Irish Sea, reducing to 1.5 km near the pipeline. It is assumed that a reversible equilibrium exists, described by a K_d , between dissolved and particle-adsorbed radionuclides. Recent developments have included the extension of the area covered by the grid, to include virtually the whole of the Irish Sea, and the incorporation of transport rates derived from a hydrodynamic continental shelf model (Howorth, 1989). The effect of these developments has been: to reduce earlier discrepancies between predicted and observed dissolved $^{239,240}\text{Pu}$ concentrations; to permit a more comprehensive validation of predictions, and, hence, increase confidence in the predicted doses; and, to provide an improved basis for the selection of the transport rates, to which the model is sensitive. The model, generally, gives good agreement between predicted and observed dissolved ^{137}Cs concentrations (Figure 15), although the seabed distributions of Pu and Am are less successfully reproduced (Figure 16).

9.3 MIRMAID model

The MIRMAID model (MAFF Irish Sea Modelling Aid) was developed to predict doses to man from seafood consumption, incorporating current knowledge on the distribution and behaviour of radionuclides in the Irish Sea (sections 4 and 5), with particular emphasis on sediment interactions. A series of papers has described the model development (Gurbutt *et al.*, 1988; Kershaw *et al.*, 1988(a),(d); Gurbutt and Kershaw, 1989; Gurbutt *et al.*, in press) and the dose calculations which have been made (Pentreath *et al.*, 1989(a),(b)).

The model consists of a number of regions (Figure 17), for each of which has a stack of boxes in the vertical (Figure 18). Vertical exchanges between the boxes were derived from a knowledge of the biogeochemistry of the system (section 5). Spatially variable suspended loads and variable sediment 'resuspension' were incorporated, with the aid of $^{234}\text{Th}/^{238}\text{U}$ disequilibria data (Kershaw *et al.*, 1988(a)). This provided an independent means of tuning the model, resulted in an improved match between predicted and observed artificial radionuclide concentrations and, hence, gave greater confidence in the model's dose predictions. Dose predictions, using appropriate concentration factors, consumption rates and gut transfer factors, for a critical group of seafood consumers, consisting of the local fishing community at Sellafield, are shown in Figure 19 for the period 1952 to 2050, together with estimates based on monitoring and habits-survey data for the period 1980 to 1987 (Pentreath *et al.*, 1989(b)).

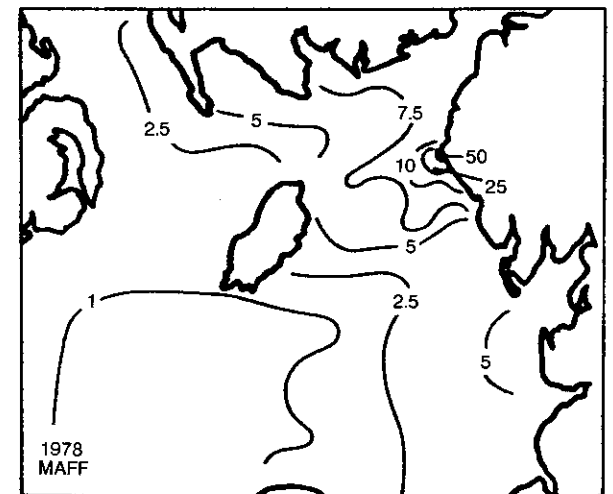
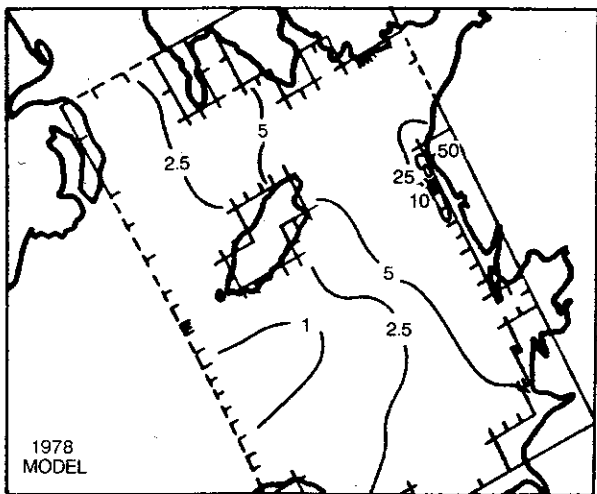
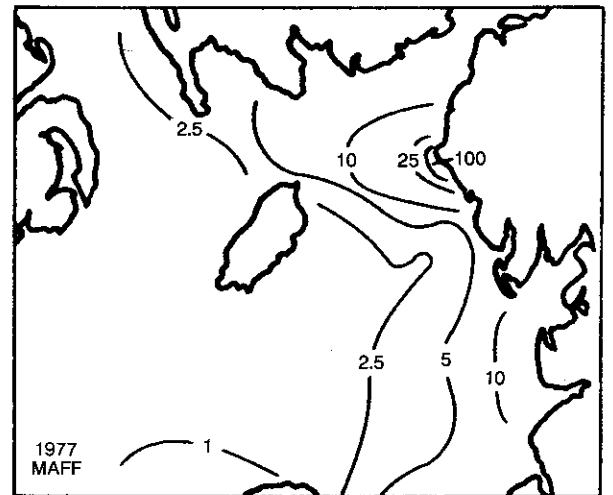
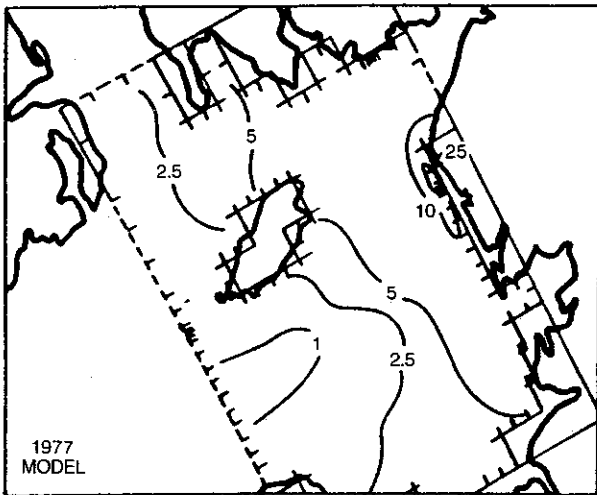
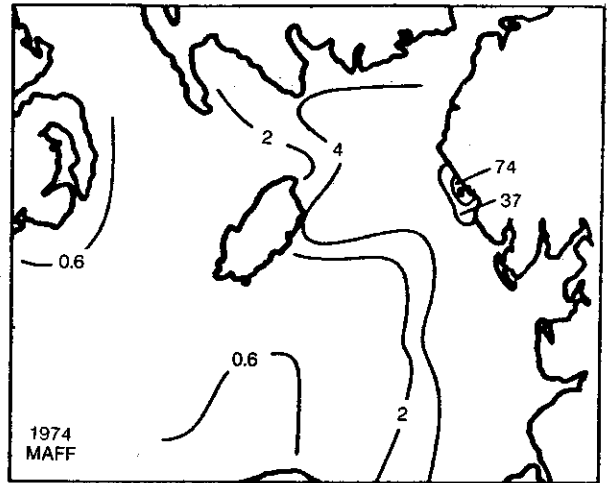
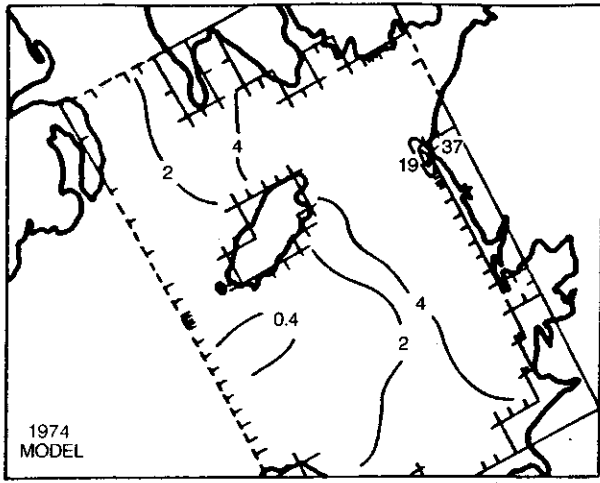


Figure 15. Comparison between predicted and measured concentrations of ^{137}Cs (Bq l^{-1}) in filtered sea water, using the Harwell model (from: Howorth, 1989)

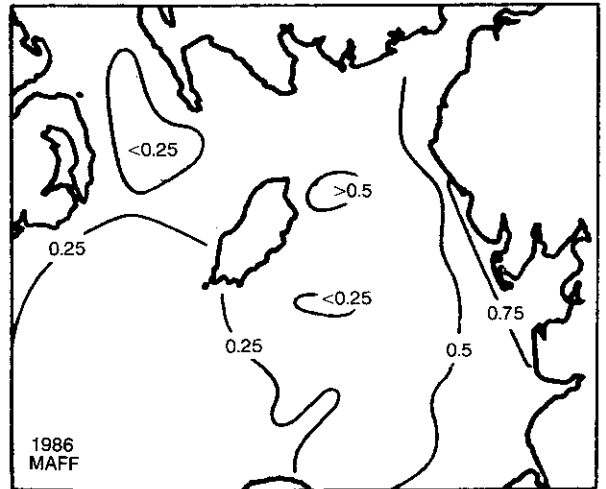
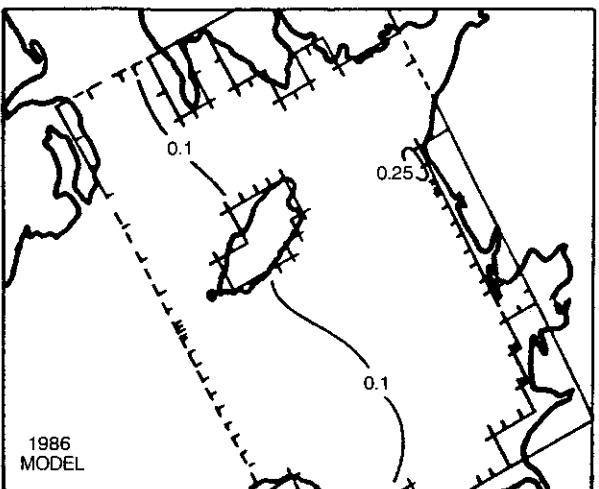
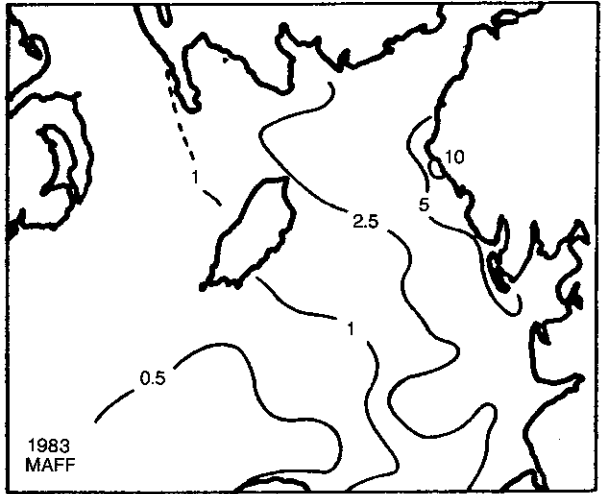
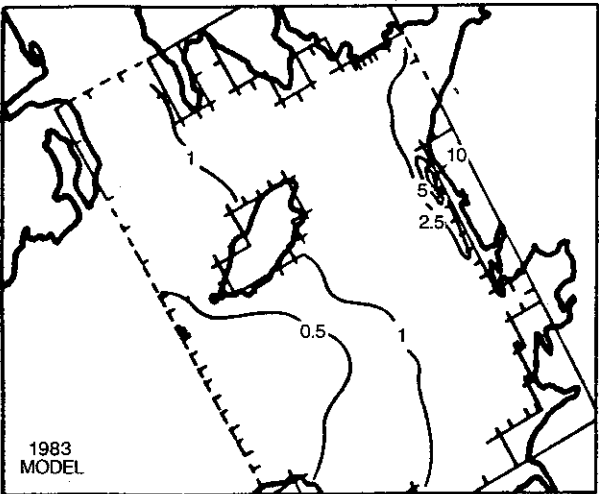
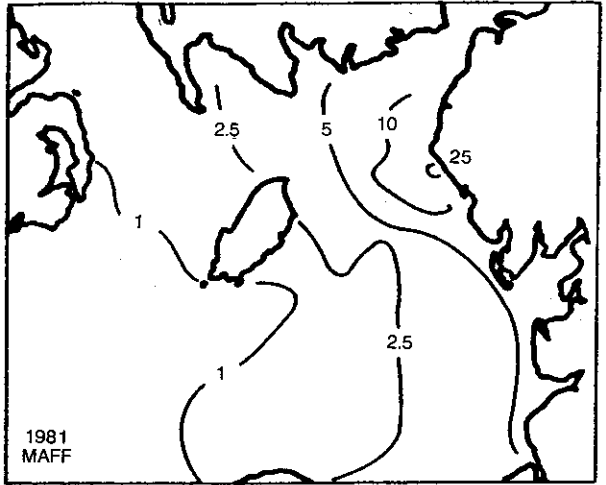
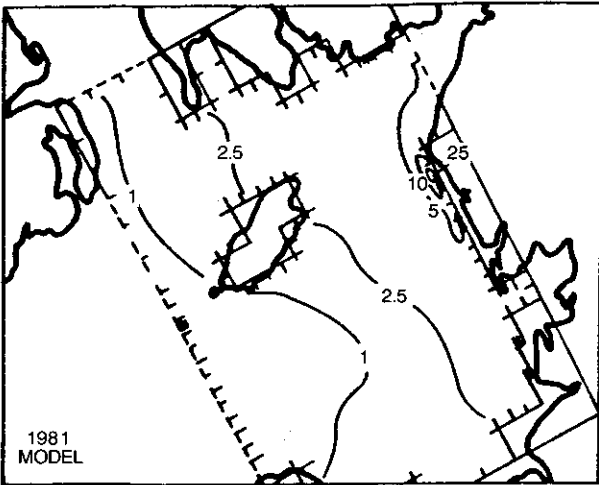


Figure 15. Continued

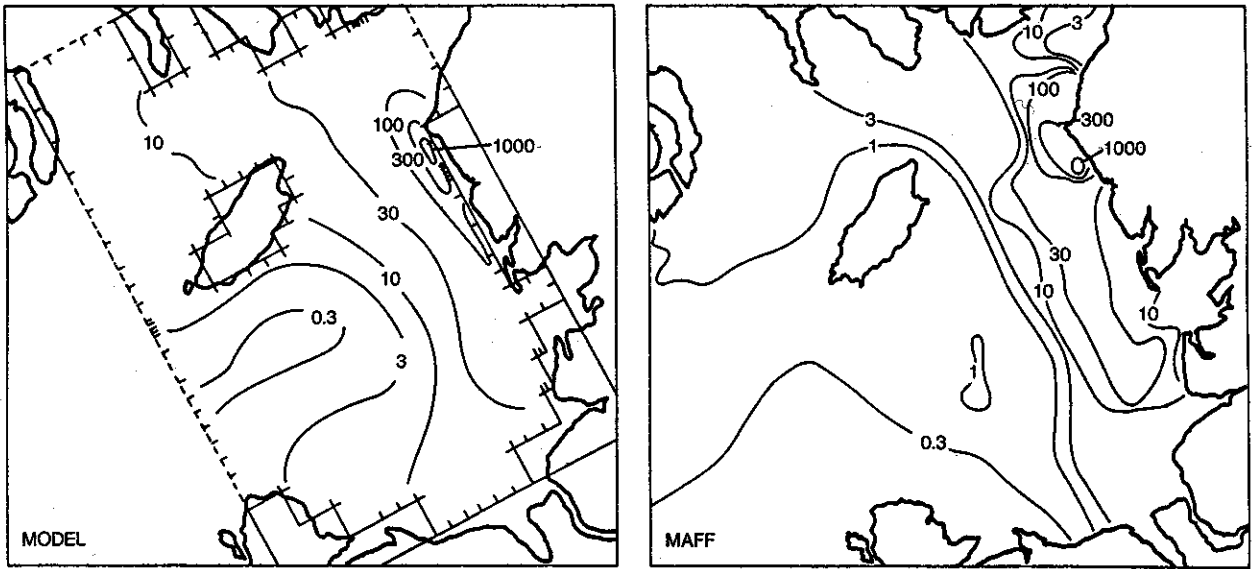


Figure 16. Comparison between predicted and measured concentrations of $^{239,240}\text{Pu}$ (kBq m^{-2}) in deposited sediments in 1978 (from: Howorth, 1989)

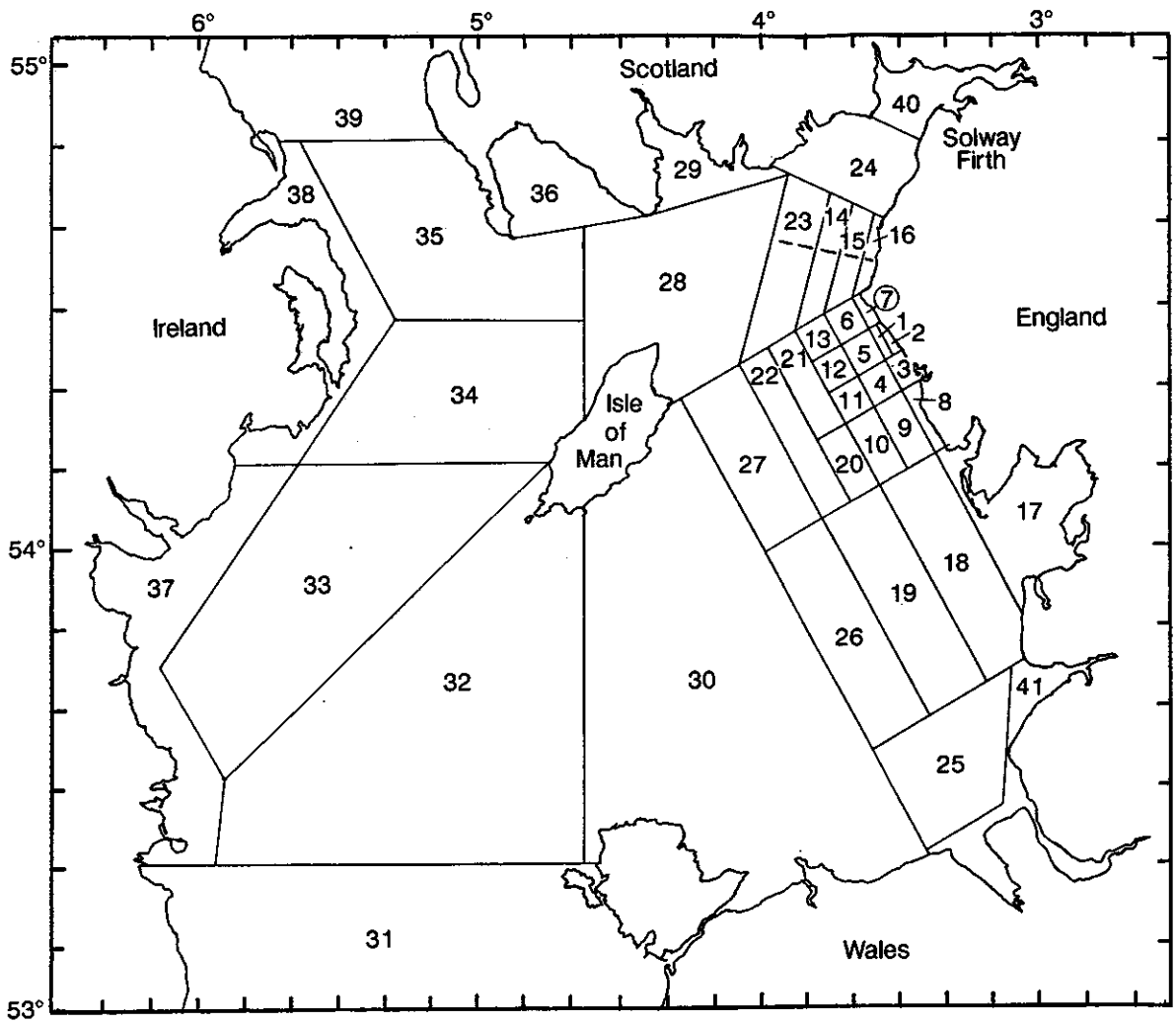


Figure 17. Regional structure of the MAFF MIRMAID compartmental model

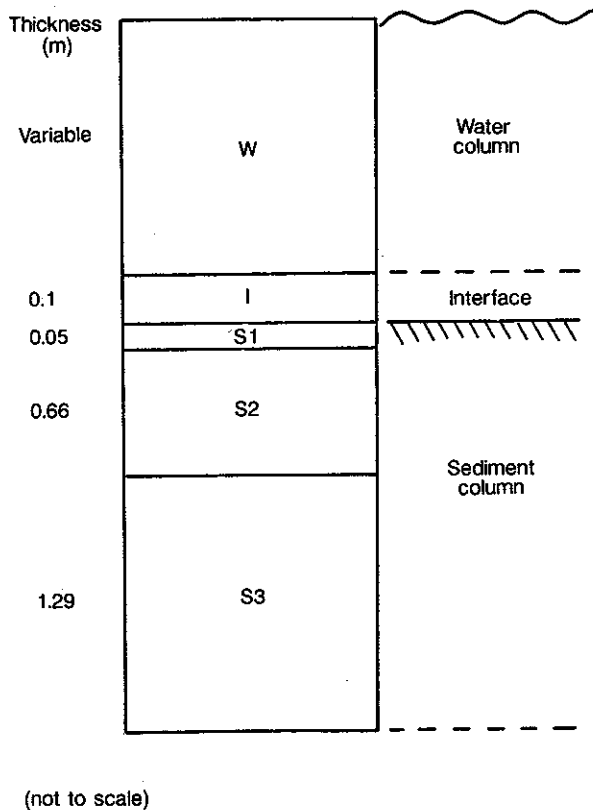


Figure 18. Vertical structure of the MAFF MIRMAID compartmental model

10. IMPACT ON THE ENVIRONMENT

The assessments of the impact of discharges from Sellafield have always taken the irradiation of the local marine fauna and flora into consideration. Initial calculations had estimated that, at the maximum permissible discharge rate, the maximum possible dose rate would be about $0.45 \text{ mSv hour}^{-1}$ over a relatively small area. It was concluded that such a dose rate would be unlikely to have any significant effect on the marine ecosystem (Dunster, 1964).

The most radiosensitive components of the marine environment were generally considered to be the eggs of marine fish and, in view of the proximity of a plaice (*Pleuronectes platessa*) spawning area to Sellafield, an evaluation of the dose rates to developing plaice embryos was made (Woodhead, 1970). Plaice eggs were reared in sea water containing ^{144}Ce , ^{137}Cs , ^{106}Ru , ^{90}Sr and $^{95}\text{Zr}/^{95}\text{Nb}$. Concentration factors were determined and, by relating these to ambient sea-water concentrations off Sellafield, the dose rates to developing embryos were calculated using modelling techniques. It was concluded that, as the estimated β and γ

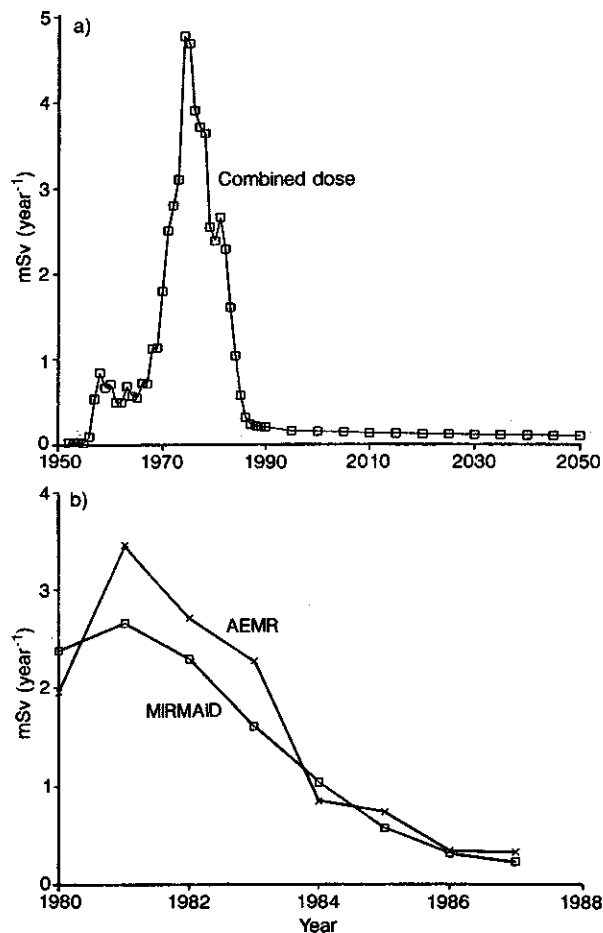


Figure 19. Calculated dose rates, using the MIRMAID model, received by a critical group from seafood consumption: (a) for the period 1952 to 2050; (b) compared with estimated dose rates from monitoring data, reported in MAFF's Aquatic Environment Monitoring Reports (AEMR), for the period 1980 to 1987 (from: Pentreath *et al.*, 1989)

irradiation dose rates were less than those from natural background (^{40}K), no adverse effects on the embryos would be expected. Subsequent experiments were made with ^{239}Pu (Hetherington *et al.*, 1976), from which it was concluded that irradiation from α -particles would be negligible, due to the short hatching time of the eggs (~ 2 weeks) relative to the decay rate of the accumulated radionuclide burden. The direct effect of irradiation of plaice eggs has also been studied (Templeton, 1966; Ward *et al.*, 1971): the LD_{50} at metamorphosis has been estimated as being 0.9 Gy , whereas the dose rates to the eggs off Sellafield were estimated as being $\sim 1.0 \text{ nGy hour}^{-1}$ (Woodhead, 1970).

The dose rates to the gonads of adult plaice have also been studied (Woodhead, 1971, 1973 (a), (b), 1974; Pentreath *et al.*, 1973). Calculations based on sea-bed radioactivity measurements indicated that the dose rate could range up to 50 $\mu\text{Gy hour}^{-1}$ from β and γ radiation, and direct estimates were obtained by using LiF-sachet dosimeters attached to the fish together with Petersen disc tags. A total of 3580 marked plaice were released 0.4 km from the discharge point, of which 1053 were subsequently recaptured. A mean dose rate to the gonad of some 2 $\mu\text{Gy hour}^{-1}$ was calculated from these data (Woodhead, 1973(a)), allowing for the habits of the fish throughout a 24-hour period. It was also estimated that plaice living in the vicinity of Sellafield for a period of 5 years (by which age most have been caught) would receive a life-time dose in the region of 90 mGy (Pentreath *et al.*, 1973). The dose rates to other components of the fauna were also calculated, from a knowledge of water and sediment concentrations and using simple dosimetry models (Woodhead, 1973(b)), the results of which are given in Table 21.

It must be assumed that any exposure to radiation carries some degree of risk of harm. For marine organisms, however, if the damage to individuals is not manifest at the population level, and does not damage the overall reproductive capacity of the population, then the effect may be regarded as being of little significance. Evaluation of the dose rates received by the marine fauna off Sellafield have been compared with the known effects of irradiation on aquatic organisms, and a programme of irradiation effects studies has been in progress at MAFF, Lowestoft, for some 15 years (Woodhead, 1976, 1977; Knowles, in press). In a comprehensive review of radiation effects, particularly on teleosts, it was concluded that the lowest dose rate at which minor radiation-induced disturbances of physiology or metabolism might be detectable was about 400 $\mu\text{Sv hour}^{-1}$ (IAEA, 1976). The dose rates around Sellafield are at least an order of magnitude below those which would be expected to elicit any effect under controlled laboratory conditions, and about two orders of magnitude below those which

Table 21. Dose rates ($n\text{Gy h}^{-1}$) to marine organisms from waste disposal radionuclides (from: Woodhead, 1973(b))

Isotope and source of radiation		Phytoplankton	Zooplankton	Mollusca
Windscale				
^{134}Cs	External	-	-	-
	Internal	-	-	-
^{137}Cs	External	(0.2-2.0)	(0.2-1.6)	(0.2-1.6)
	Internal	-	-	(0.9-8.1) $\times 10^2$
$^{144}\text{Ce}/^{144}\text{Pr}$	External	(0.4-1.6)	(0.3-1.2)	(0.1-0.6)
	Internal	(51-160) $\times 10^2$	(11-40) $\times 10^2$	(15-55) $\times 10^2$
$^{106}\text{Ru}/^{106}\text{Rh}$	External	(0.8-7.5)	(0.6-6.4)	(0.1-1.1)
	Internal	(14-140) $\times 10^2$	(41-410) $\times 10^2$	2-20
$^{95}\text{Zr}/^{95}\text{Nb}$	External	(0.7-22.0)	(0.6-21.0)	(0.6-21.0)
	Internal	(2-51) $\times 10^2$	(11-280) $\times 10^2$	0.3-11.1
Isotope and source of radiation		Crustacea	Fish	Sediment
Windscale				
^{134}Cs	External	-	-	-
	Internal	-	1.4-4.9	-
^{137}Cs	External	0.2-1.6	0.2-1.6	α -dose (0.1-1.0) $\times 10^2$ β -dose (0.1-0.6) $\times 10^2$
	Internal	0.9-7.3	3.6-10	-
$^{144}\text{Ce}/^{144}\text{Pr}$	External	0.01-0.06	0.01-0.06	α -dose (0.1-2.5) $\times 10^2$ β -dose (8 - 170) $\times 10^2$
	Internal	0.8-3.1	-	-
$^{106}\text{Ru}/^{106}\text{Rh}$	External	0.1-1.1	0.1-1.1	α -dose (0.9-20) $\times 10^2$ β -dose (12-290) $\times 10^2$
	Internal	67-660	-	-
$^{95}\text{Zr}/^{95}\text{Nb}$	External	0.6-210	0.6-21.0	α -dose (25-310) $\times 10^2$ β -dose (0.6-7.7) $\times 10^2$
	Internal	0.3-8.4	-	-

might be expected to have an effect at the population level (Woodhead, 1980) during the period of maximum discharges. Dose rates are likely to decline significantly as a result of the dispersion and mixing of water and sediments and the actual and predicted decreases in Sellafield discharges.

The possible influence of contaminant input on the prevalence of fish diseases has been assessed (Dickson and Boelens, 1988). Studies of fish disease in the Irish Sea have been reported from 1904 onwards (Woodcock, 1904). A number of investigations of demersal species took place in the 1970s and 1980s, with a dedicated cruise in 1986 being the most relevant. This compared the incidence of disease in dab and plaice near Point of Ayr (Liverpool Bay), St Bees (Cumbria) and Dundrum Bay (Ireland) with a 'control' site in

northern Cardigan Bay. Dab showed the highest disease prevalence off Point of Ayr and Dundrum Bay whereas for plaice this occurred off St Bees. The lowest disease prevalence occurred in Cardigan Bay. However, there is undoubtedly considerable uncertainty in the degree of natural spatial and temporal variability, to the extent that Dickson and Boelens (1988) maintain that it remains to be demonstrated that anthropogenic inputs (which would include radioactive wastes) are having a significant effect on the prevalence of fish disease in the Irish Sea. The effects of radiation on the humoral immune response of fish are being investigated (Knowles, 1992) together with work on the combined effects of heavy metals and radiation on marine biota and the possible genotoxic effect of contaminants (including radiation) on fish chromosomes (J. K. Knowles, pers. comm.).

Table 22. External radiation exposure of black-headed gulls in the Ravenglass Estuary (from: Woodhead, 1986)

Substrate	Measured dose rate at 1 m (Hunt, 1983) (Gy h ⁻¹)
Silt/mud	(4.6-9.2) x 10 ⁻⁷
Sand	(1.6-2.0) x 10 ⁻⁷
Saltmarsh	9.0 x 10 ⁻⁷
Average bird behaviour over 24 h	Estimated mean dose rate (Gy h ⁻¹)
2h flying	0
8h feeding over silt/mud	8.7 x 10 ⁻⁷
8h feeding or roosting over Saltmarsh	1.4 x 10 ⁻⁶
5h roosting over sand	2.8 x 10 ⁻⁷
1h incubating eggs on sand	3.7 x 10 ⁻⁷
Overall 24h mean dose rate	8.3 x 10 ⁻⁷

Table 23. Radionuclide concentrations in black-headed gull faeces and regurgitated pellets (Allen et al., 1983): estimated total radionuclide content of the alimentary tract, and equivalent whole body and gut content concentrations (from: Woodhead, 1986)

Nuclide	Radionuclide concentration (Bq g ⁻¹ wet weight)		Total activity in the gut (Bq)	Equivalent whole body concentration, (C _{WB}), for γ-ray dosimetry (Bq g ⁻¹)	Equivalent concentration, (C _G), in gut contents for α- and β-dosimetry (Bq g ⁻¹)
	Faeces	Pellets			
⁶⁰ Cr	7.0 x 10 ⁻³	1.4 x 10 ⁻²	7.1 x 10 ⁻²	2.8 x 10 ⁻⁴	8.2 x 10 ⁻³
⁹⁵ Zr	5.1 x 10 ⁻²		3.7 x 10 ⁻¹	1.5 x 10 ⁻³	4.3 x 10 ⁻²
⁹⁵ Nb	7.6 x 10 ⁻¹	1.7	7.9	3.2 x 10 ⁻²	9.2 x 10 ⁻¹
¹⁰⁶ Ru	6.0 x 10 ⁻¹	7.7 x 10 ⁻¹	5.4	2.2 x 10 ⁻²	6.2 x 10 ⁻¹
¹³⁴ Cs	1.1 x 10 ⁻²	1.2 x 10 ⁻²	9.7 x 10 ⁻²	3.9 x 10 ⁻⁴	1.1 x 10 ⁻²
¹³⁷ Cs	2.4 x 10 ⁻¹	2.9 x 10 ⁻¹	2.1	8.6 x 10 ⁻³	2.5 x 10 ⁻¹
¹⁴⁴ Ce	2.1 x 10 ⁻¹	2.4 x 10 ⁻¹	1.8	7.3 x 10 ⁻³	2.1 x 10 ⁻¹
¹⁵⁴ Eu	2.7 x 10 ⁻²		1.9 x 10 ⁻¹	7.8 x 10 ⁻⁴	2.3 x 10 ⁻²
¹⁵⁵ Eu	2.4 x 10 ⁻²		1.7 x 10 ⁻¹	6.8 x 10 ⁻⁴	2.0 x 10 ⁻²
²³⁸ Pu	8.7 x 10 ⁻²		6.3 x 10 ⁻¹	2.5 x 10 ⁻³	7.3 x 10 ⁻²
^{239/240} Pu	3.3 x 10 ⁻¹		2.4	9.5 x 10 ⁻³	2.8 x 10 ⁻¹
²⁴¹ Am	5.8 x 10 ⁻²	6.7 x 10 ⁻²	5.1 x 10 ⁻¹	2.0 x 10 ⁻³	5.9 x 10 ⁻²

Table 24. Estimates of the radiation exposure of the black-headed gulls using the Ravenglass Estuary (from: Woodhead, 1986)

Target	Source	Dose rate (Gy h ⁻¹)
Whole body	β/γ -emitters in the whole body	1.3×10^{-9}
	β/γ -emitters in the gut	2.0×10^{-9}
	γ -emitters in the whole body	7.2×10^{-9}
	External γ -emitters	8.3×10^{-7}
	Total	8.4×10^{-7}
Cells lining the alimentary tract	β/γ -emitters in the whole body	1.3×10^{-9}
	β/γ -emitters in the gut	3.7×10^{-7}
	α -emitters in the gut	6.4×10^{-7}
	External γ -emitters	8.3×10^{-7}
	Total	1.8×10^{-6}
Eggs	Internal β/γ -emitters	4.1×10^{-9}
	External γ -emitters	3.7×10^{-7}
	Total	3.7×10^{-7}

The Sellafield discharges have been cited as the cause for the disappearance of a breeding colony of black-headed gulls from the Ravenglass Estuary. This colony was once the largest in England, with about 10,000 pairs, but declined rapidly in the 1970s and was abandoned by 1985 (Phillips and Coggle, 1988). The radiation exposure of the gulls was examined by Woodhead (1986) who made use of existing data on external γ -ray exposure (Table 22) (Hunt, 1983) and the concentrations of Sellafield radionuclides in gulls from the area (Table 23) (Allen *et al.*, 1983). The whole body dose rate to the gulls from contaminant radionuclides (Table 24) was estimated to be ten times that from natural background ($\sim 10^{-7}$ Sv h⁻¹), the dose to the eggs approximately four times background, but the dose to the gut was two orders of magnitude higher, principally due to α -radiation. Comparing these doses to the available data on radiation effects in birds led Woodhead (1986) to conclude that the increased radiation exposure could have no observable effects on either individual birds or the total population. In a further study, Phillips and Coggle (1988) concluded that the radiation dose required to significantly reduce the hatching rate of gull eggs was about 20,000 times greater than the maximum dose which could have been received by the eggs at Ravenglass, and that the dose to produce abnormalities was about double this. From these two studies, it can be concluded that an agent, or agents, other than the Sellafield discharges was responsible for the disappearance of the black-headed gull from the Ravenglass Estuary. A recent study (Anderson *et al.*, 1988) has confirmed this conclusion and identified disturbance and predation by forces gaining access to the breeding area (formerly a fenced nature reserve) as the cause of the decline.

11. CONCLUDING REMARKS

It should be apparent that a considerable body of knowledge exists on the behaviour of radionuclides in the Irish Sea and the transfer processes responsible for their redistribution. This is represented by the large number of references cited in the text (~ 340). Many of these have arisen because of a need to assess the radiological consequences of the principal discharge source, BNF Sellafield (formerly referred to as Windscale). Other studies have used the opportunity provided by the elevated concentrations of artificial radionuclides to investigate their fundamental chemical properties and post-depositional behaviour. This is information which has a much wider applicability in other actual or potential waste disposal or accidental release events. A third category has made use of the time- and space-dependent distribution of artificial radionuclides to elucidate the nature of transfer processes within the Irish Sea and beyond over much of the NW European continental shelf.

The subtidal sediments of the eastern Irish Sea represent a significant potential source of long-lived radionuclides. There is evidence that Cs has been remobilised from the seabed since the early 1980s. The estuaries of the coasts of NW England and SW Scotland appear to be acting as continuing sinks for Sellafield-derived nuclides. It is not possible to predict with any confidence what the longer term consequences of sediment redistribution will be, particularly in response to infrequent storm events, on radionuclide distributions. However, it is unlikely that the dose consequences will approach the levels experienced in the late 1970s to early 1980s in view of the sustained

decrease in Sellafield discharges and the effect of dispersion. The 'ultimate sink(s)' is unknown, although it is conceivable that the area of muddy sediments between Ireland and the Isle of Man, in deep, relatively quiescent conditions, will play an increasingly important role, albeit over a very long time-scale.

Studies are in progress, or are planned, to attempt to address some of the unknowns cited above. For example, the response of sediments to waves and tidal currents will be linked to a fine resolution hydrodynamic model in a study conducted by a number of University departments and research institutions. A second example is the proposal to utilise the probable increase in Tc discharges in 1993-94 to improve estimates of water transport times, exchanges through the North Channel and to validate existing transport models.

The success of such proposals will depend on many factors, including being able to attract an appropriate level of funding. The field is of interest beyond the UK and increasingly the large-scale, multi-disciplinary approaches required to carry out this type of environmental research is attracting collaboration with scientists in the EC and the wider international community.

It is hoped that the present review will provide a useful summary of past efforts and a stimulus for further work.

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