Anhang/Appendix G

Radioactivity around the Sellafield Nuclear Complex, UK: a review of ITE studies, 1990-1994

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Introduction

The Sellafield Nuclear Complex, operated by the British Nuclear Fuels Ltd (BNFL) and located in West Cumbria, is the largest nuclear complex in the United Kingdom. Sellafield is the site of one of the world's largest nuclear fuel reprocessing plants, and is also the location of the Calder Hall nuclear power station and facilities related to the storage and treatment of radioactive wastes. Nuclear-related operations began at Sellafield in 1947 and have continued to expand since that time. The latest major facility, the Thermal Oxide Reprocessing Plant (THORP) for the reprocessing of uranium oxide fuels, underwent commissioning in 1994 and is now in full operation. A new manufacturing plant for the production of mixed oxide fuel using uranium (U) and plutonium (Pu) recovered from reprocessing began construction during 1994 and is expected to start operations in 1998.

There are thus several potential sources of radioactivity in the environment around Sellafield, and these have been studied in detail for several decades. The Institute of Terrestrial Ecology (ITE) has been carrying out radioecological investigations on Sellafield discharges since the 1980s. The aim of this report is to present an overview of the technical operations at the Sellafield site and how these have contributed to radioactive contamination in the area of west Cumbria. This report is not intended to be a comprehensive review; as a basis for assessing the impact of Sellafield discharges in the terrestrial and marine environments, the results of recent radioecological studies at ITE (1990 onwards) will be discussed.

Technical operations at Sellafield

The main activities at Sellafield are nuclear fuel reprocessing and management of the radioactive waste that is produced. Fuel reprocessing is the mechanical and chemical extraction of uranium and plutonium from nuclear fuel after it has been irradiated in a reactor. Used nuclear fuel rods are transported to Sellafield and stored in fuel storage ponds prior to reprocessing. Typically, a used uranium fuel rod contains around 96% U, 1% Pu and 3% waste fission products (8). Fuel reprocessing at Sellafield is designed to separate the U and Pu from the fission products for the manufacture of new fuel. Two facilities for reprocessing are now operating at Sellafield: the Magnox reprocessing facility and more recently, the Thermal Oxide Reprocessing Facility (THORP). As the name implies, the Magnox reprocessing facility is used for the reprocessing of spent magnox fuel which originates from UK nuclear power plants; since starting operation in 1964, some 35000 tonnes of magnox fuel have been reprocessed by 1992 (8). THORP was completed in 1992 and became fully

operational in 1995. This facility is used for the reprocessing of oxide fuels originating from more advanced reactors in the UK and from BNFL's overseas clients.

The radioactive wastes generated at Sellafield following storage and reprocessing operations are classified in terms of their radioactivity content into low, intermediate, and high level solid and liquid wastes. These wastes are treated at a series of waste retrieval and treatment plants on site. High-level waste (HLW), also sometimes referred to as heat-generating waste, is radioactive enough to generate heat. The acid solution used to dissolve the U rods during reprocessing is the main source of HLW at Sellafield. According to BNFL (8), reprocessing one tonne of used nuclear fuel produces about 0.1 cubic metres of high-level waste, which contains almost 99% of the waste radioactivity.

Solid intermediate-level wastes consist mainly of fuel cladding materials, sludge and ion exchange resins resulting from low-level effluent treatment processes, contaminated equipment, and materials contaminated with Pu. These wastes are incorporated into a cemented matrix inside 500 litre stainless steel drums in the Waste Packaging and Encapsulation Plant (WPEP) and are stored on site pending permanent disposal.

The low-level wastes generated at Sellafield occur in either solid or liquid form. Solid low-level waste originates from areas where radioactive materials are used, and includes items such as gloves, paper towels, scrap metal, and contaminated protective clothing and equipment. These wastes are compacted at the Waste Monitoring and Compaction Facility (WAMAC), encapsulated in steel containers, and disposed of at the Drigg waste disposal facility located 6 km south-east of Sellafield. Drigg is a land burial facility also operated by BNFL.

The bulk of the low-level wastes is in liquid form, consisting mainly of fuel storage pond water and of process liquors arising from magnox and THORP reprocessing operations. Over the years, several facilities have been built at Sellafield to reduce the radioactivity levels in these effluents prior to discharge into the Irish Sea. The Site Ion Exchange Effluent Plant (SIXEP), which began operation in 1985, was designed to remove Cs and Sr ions from storage pond water. SIXEP uses ion exchange on clinoptilolite, a naturally occurring zeolite, to decontaminate the pond water; once saturated, the clinoptilolite is removed and processed as intermediate-level waste. Following the operation of the SIXEP facility, significant reductions in radiocaesium levels in liquid discharges have been attained (Figure 1a). The Salt Evaporator Plant, which also began operation in 1985, is used to condition the effluents from the magnox reprocessing plant. The Enhanced Actinide Removal Plant (EARP), designed to remove alpha activity and further reduce beta activity from effluents using chemical treatment and filtration started operation in 1994. The acidic effluents arising from reprocessing contain significant amounts of iron; when neutralised with sodium hydroxide, ferric hydroxide floc is formed. Most of the alpha activity co-precipitates with this floc, which can be removed by filtration and then treated as intermediate-level waste. The amounts of actinides (mainly Pu isotopes and Am) discharged into the Irish Sea have decreased significantly following operation of EARP (Figure 1b).

Discharges to the environment

BNFL is allowed to discharge limited amounts of radioactivity to the environment from the Sellafield site, under authorisation by the relevant UK authorities. Up to 1995, the Authorising Departments were Her Majesty's Inspectorate of Pollution (HMIP) and the Ministry of Agriculture, Fisheries and Food (MAFF). Following the Environment Act of 1995, the Certificates of Authorisation are now issued by the Environment Agency, which was created in 1996, with the provision that MAFF is consulted before an authorisation is granted (11). The certification allows the discharge of low level waste gases, mists and dusts from the Sellafield premises into the atmosphere, and of low level liquid effluents (following decontamination at the various waste treatment facilities) via a 2.5 km pipeline to the Irish Sea. A list of radiologically important nuclides, their discharge limits and the amounts discharged during 1996, is provided in Table 1. The discharge limits are established by the authorising bodies in accordance with radioactive waste management principles as set out by the UK government, following international guidelines.

Aerial discharges

Ventilation air from the process plants and the Calder Hall reactors are discharged from several stacks with different release heights. Most of the stacks are equipped with particulate filters and are monitored continuously; however, there are a number of minor and unmonitored release points. The main components are noble gases (^{41}Ar and ^{85}Kr) and ^{3}H; gases and vapours containing ^{14}C, ^{129}I, ^{131}I and small amounts of ^{241}Am and Pu isotopes are also released (Table 1). There have been significant decreases in the amounts of ^{137}Cs, ^{131}I, ^{241}Am and Pu isotopes discharged to the atmosphere in recent years compared to releases in the late 1970s to the mid 1980s (Figure 2). On the other hand, following the recent operation of THORP, significant increases in ^{85}Kr release have been reported (10, 11).

Accidents have also resulted in atmospheric releases of radioactive materials around Sellafield and beyond. The most well known of these is the fire at Windscale Pile No. 1 in October 1957, which resulted in a deposition of radionuclides over England and Wales and parts of northern Europe. Several early publications provide descriptions of the events leading to and following the accident and the measurements and surveys that were carried out (eg, 17, 18, 23, 24). The accident occurred during a deliberate release of stored Wigner energy, a routine maintenance operation. (Wigner energy is the energy stored in graphite due to neutron bombardment). The UK National Radiological Protection Board (NRPB) published an assessment of the impact of this accident in 1962 (24). A complex dispersion pattern was observed because the meteorological conditions varied at the time of the release (23). The major radionuclide deposited was ^{131}I, with major intake routes via inhalation (and subsequently, doses to the thyroid) and milk ingestion (via the pasture-cow-milk pathway). An immediate ban on milk supplies was imposed, and milk distribution was resumed 44 days after the accident. From collective dose calculations based on the radionuclide data, the NRPB concluded that the pathway contributing the most to the collective dose was the ingestion of contaminated milk, with ^{131}I contributing nearly all of the collective dose to the thyroid and the largest part of the collective

Table 1. Authorised discharge limits of selected radionuclides (marine and airborne releases). For airborne discharges, only the major sources of the 1996 discharges are indicated; also note the change in discharge units for some airborne releases (from Reference 11).

Radionuclide	1996 Discharge	Authorised Limit	
	(TBq)	*(TBq)*	
Marine discharges			
Tritium	3000	31000	
Carbon-14	11	20.8	
Strontium-90	16	48	
Technetium-99	150	200	
Caesium-137	10	75	
Plutonium alpha	0.21	0.7	
Plutonium-241	4.4	27	
Americium-241	0.07	0.3	
Airborne discharges			*Source*
Tritium	520	1400	high stacks
	4.3	43	THORP
Carbon-14	0.28	7.5	high stacks
	0.33	0.47	Calder Hall
Argon-41	2600	3700	Calder Hall
Krypton-85	55000	120000	high stacks
	39000	470000	THORP
	(GBq)	*(GBq)*	
Sulphur-35	140	210	Calder Hall
Iodine-131	2.3		high stacks
Caesium-137	0.62	5.1	intermediate stacks
	0.13	2.0	low stacks
Plutonium alpha	0.057	0.66	high stacks
Plutonium-241	0.53	4	high stacks
Americium-241	0.036	0.2	high stacks

effective dose. The radiologically significant nuclides that contributed to the inhalation dose were ^{131}I, ^{132}Te, ^{137}Cs, ^{103}Ru, ^{144}Ce, ^{91}Y and ^{129m}Te; those that also contributed to the ingestion and external irradiation dose were ^{131}I, ^{132}Te and ^{137}Cs. It was further concluded that in the longer term following the release, ^{137}Cs was the most important nuclide in terms of external dose and the ingestion of contaminated foodstuffs. The most exposed group in the population were young children drinking milk produced in the northern counties; the maximum individual thyroid dose was 160 mSv to a child in the Sellafield area (24). Other incidents at Sellafield involving unplanned releases of radioactivity to the environment have been noted (31, 84, 85). These incidents have resulted in radionuclide deposition at a local level and not as extensive as the October 1957 accident.

Marine discharges

Liquid discharges via a 2.5-km pipeline to the Irish Sea started in 1952. Although the discharges are generally classified as 'low-level' liquid wastes deemed suitable for marine disposal, the amounts of radioactivity released to the Irish Sea during the 40+ years of operations at Sellafield are significant. The radionuclide composition of liquid effluents has varied considerably during this period (84). Low level liquid effluents are held in sea tanks and monitored prior to discharge. The discharges are primarily in liquid form, but the occurrence of radioactive particulate matter (so-called 'hot particles' containing Pu and Am) in the effluents has been documented (eg, 32, 74). It is believed that some very fine solid debris may be released with the liquid effluents, and some particle formation may also occur upon contact of the effluents with seawater.

The principal radionuclides occurring in liquid discharges are ^{3}H, ^{137}Cs, ^{241}Am and isotopes of Pu; in terms of total activity, ^{3}H is the dominant radionuclide (Table 1). As mentioned previously, the discharges of ^{137}Cs, ^{241}Am and Pu isotopes have decreased significantly compared to releases during the period between 1970 to 1980, as a result of the operation of several effluent treatment plants (Fig 1). In recent BNFL reports (10, 11,12), it has been shown that the amount of radioactivity in liquid discharges to the Irish Sea is now less than 1% of the discharges in the mid-1970s. However, there have been significant increases in ^{99}Tc discharges following the treatment at the EARP facility of a backlog of stored wastes and the operation of THORP (11, 12, 50, 53). The annual authorised limit of ^{99}Tc discharge in liquid effluents has been increased from 10 to 200 TBq during 1994 (10); in 1995, the amount discharged (190 TBq) corresponded to 95% of this limit (11). There is growing public concern over the increasing levels of this radionuclide in seafoods such as lobster; significant increases in ^{99}Tc content of the seaweed *Fucus vesiculosus* have also been reported in 1995 compared to previous years (3).

Monitoring activities

As part of its authorisation to discharge radioactive wastes, BNFL is required to monitor the wastes prior to disposal in order to demonstrate compliance with the discharge limits, and to perform regular environmental monitoring. Reports on radioactive discharges and environmental monitoring around BNFL sites, including Sellafield, are issued annually (e.g., 10, 11, 12). The monitoring activities focus on those routes of exposure considered to make significant dose contributions to the critical group (i.e., those individuals in the population expected to receive the highest dose) and to the public. For Sellafield discharges, the following exposure pathways have been identified as relevant contributors to human doses: (i) internal exposure from high rate consumption of seafood (fish and shellfish) and of local agricultural produce (mainly milk); (ii) external irradiation from exposed sediments (especially silts and muds in estuaries and harbours); and (iii) inhalation of, and exposure to, airborne radioactivity (12). Regular sampling and analysis programmes are performed to determine radionuclide activity concentrations or radiation levels relevant to these

pathways. The habits and consumption rates relating to each pathway are also kept under review.

Independent monitoring surveys around the Sellafield site and in the Irish Sea are also carried out regularly by MAFF. The annual results are published in various reports, such as the Terrestrial Radioactivity Monitoring Programme (TRAMP) Reports (e.g., 52), and the Aquatic Environment Monitoring Reports (e.g., 15, 16, 46). More recently, the terrestrial and aquatic monitoring results of MAFF have been combined into a single report called Radioactivity in Food and the Environment (53, 54). Reviews of various aspects of the monitoring programmes have also been published over the years of Sellafield operations (e.g., 27, 39, 40, 43, 46).

Radioecological surveys have recently been carried out by ITE in support of the terrestrial and marine monitoring programmes of MAFF. The results of these surveys have been reported to MAFF and have been published in the literature (35, 36, 37, 82, 83). A brief summary of the results of these studies is presented below.

Terrestrial radioactivity

A survey of radionuclide contamination in grass and soil samples from around 18 nuclear establishments in England and Wales, including Sellafield, was carried out during 1993. Two sets of samples (paired soil and grass from 500 m outside the boundary fence of Sellafield on 3 sites along the three most dominant downwind directions) were collected in February and October 1993, and analysed for several radionuclides. Grass and soil are used as indicator materials of environmental contamination, particularly for artificial radionuclides released in atmospheric discharges. Thus, in addition to the regular monitoring of agricultural produce (milk, meat, fruits and vegetables) around nuclear sites, data on the radionuclide contents of soil and grass are also reported annually by MAFF (e.g., 52).

The activity concentrations of ^{137}Cs, ^{90}Sr, $^{239,240}Pu$ and ^{241}Am in samples collected from Sellafield during 1993 are shown in Table 2; these activity concentrations were among the highest found in the survey of 18 nuclear sites in England and Wales (36, 82). Some of these radionuclides have been deposited globally as a result of nuclear weapons testing; the Chernobyl accident in 1986 also resulted in additional deposition of radiocaesium to west Cumbria. Based on the 1993 data, an assessment was made to determine if Sellafield discharges have increased the soil contents of ^{137}Cs and $^{239+240}Pu$ in the surrounding environment (82). This was done by calculating the ratios between the ^{137}Cs and $^{239+240}Pu$ levels found in 1993 to their deposition as measured in previous surveys of Cumbrian soils. The calculated ratio for ^{137}Cs was 1.2, indicating only a slightly enhancement from Sellafield discharges for this radionuclide. The ratio for $^{239+240}Pu$ was 5, suggesting that the increase in Pu levels to the surrounding soils was more significant. It has to be noted that the 1993 measurements were only for 3 samples collected from 500 m outside the site perimeter fence; to what extent Sellafield discharges have increased the radionuclide levels in soils further afield can not be discerned from this limited data set.

Table 2. Activity concentrations of selected radionuclides in soil and vegetation samples collected near Sellafield, 1993 (from Reference 35).

Radionuclide	Sample #	Vegetation			Soil		
		site 1	*site 2*	*site 3*	*site 1*	*site 2*	*site 3*
Sr-90	1*	43.4	64.1	20.8	6.9	15.7	17.4
	2	35.8	51.7	25	9.9	12.6	17
Cs-137	1	37.2	81.1	34.1	10	104.5	235.5
	2	10.7	3.6	8.0	283	105	253
Pu-238	1	0.65	1.04	0.46	0.09	0.52	1.01
	2	0.09	0.26	0.1	1.16	0.53	1.07
Pu-239+240	1	5.53	14.04	8.33	2.02	14.45	21.07
	2	0.57	3.08	0.55	28.38	13.89	18.85
Am-241	1	3.19	3.51	2.52	0.63	3.26	9.20
	2	0.32	< 0.01	0.48	6.47	2.33	0.04

*Sample #: 1- samples collected 11 February 1993; 2- samples collected 13 October 1993.

In February 1993, BNFL reported an unusual release of Pu, Am and Cm to the atmosphere; the radionuclides originated from a redundant plant being prepared for decommissioning (9). In Figure 1, a small increase in Pu discharge during 1993 can be noted, compared to the releases between 1988 and 1992. Coincidentally, the ITE grass and soil samples were collected at the time of this unplanned release. Table 2 that the activity concentrations of Pu isotopes and ^{241}Am in vegetation samples were relatively higher in February compared to October 1993; a possible explanation would be interception of this unusual release by the vegetation. The higher Pu and Am levels measured in the soil at site 1 collected in October may also have resulted from the intercepted radionuclides being washed down onto the surface soil.

Marine radioactivity

There have been numerous studies on the radioactive discharges to the Irish Sea over the 40^{+} years of Sellafield operations (e.g., 27, 34, 41, 46, 47, 55, 56, 60, 70, 72). As a simplified approach to put the radioactive discharges in perspective, Walker & Rose (86) estimated the increase in radioactivity that would be contributed by Sellafield discharges in seawater along the Cumbrian coast (assuming that all the radioactivity is confined within its boundaries). Based on its salinity and the concentrations of naturally occurring radionuclides, 'average' seawater contains about 12.5 Bq (of total radioactivity) per kg, the major component being ^{40}K. The less saline Cumbrian coastal waters (with boundaries defined at 10 km north and south of the discharge point and extending 2 km out to sea) are estimated to contain 11.5 Bq kg^{-1} natural radioactivity. The contribution from Sellafield, based on discharge data up to 1987, was estimated to have increased the total radioactivity content by 59.4 Bq kg^{-1}. When

these discharges are diluted within the larger volume of the Irish Sea (between 52° N and 54° 50' N latitude), the contribution from Sellafield would be only an additional 0.7 Bq kg^{-1} of seawater. These are, of course, only theoretical estimates to show the relative contribution of Sellafield discharges to the surrounding marine water; the actual situation is much more complex because the individual radionuclides undergo various interactions which influence their eventual distribution in the environment.

In general, the distribution of radionuclides in the marine environment is controlled by their geochemical behaviour. Following release from the pipeline, the effluents would be diluted and dispersed, and individual radionuclides would interact with seawater, suspended particles, and the sediments. The so-called 'conservative' radionuclides such as ^{3}H, ^{137}Cs, and ^{90}Sr remain primarily in the dissolved form and are dispersed rapidly and transported by water currents within, and out of, the Irish Sea. For example, ^{137}Cs has been used as an oceanographic tracer to track the movement of water masses (e.g., 38, 55, 63, 64, 76). Other radionuclides, particularly the actinides, adsorb strongly onto suspended particulate matter, and a major fraction is deposited in sediments within the Irish Sea (e.g., 6, 7, 74). The sediment-associated radionuclides would undergo redistribution within the Irish Sea as part of the suspended load, and move around with the tidal currents (e.g., 49, 57). Some remobilisation of the scavenged radionuclides may also occur; for example, a small proportion of the discharged Pu has been found to be transported in soluble form out of the Irish Sea (e.g., 20, 42, 66, 67). Sellafield-derived radionuclides have been identified as far afield as the North Sea and the Arctic (e.g., 28, 33, 48, 66, 67, 68, 78, 81).

The marine discharges from Sellafield have been a major concern because the Irish Sea is an important fishing area, and several commercial fish species are known to take up some of the discharged radionuclides (e.g., 73, 75). The consumption of locally collected fish and shellfish is a major contributor to the dose received by a critical group of consumers in west Cumbria. The levels of selected radionuclides in seafood are thus monitored regularly. Figure 3 shows the activity concentrations of ^{106}Ru, ^{137}Cs, ^{241}Am and Pu alpha emitters in seafood collected near the Sellafield area. The activity concentrations of these nuclides show a decreasing trend reflecting the decrease in discharge levels.

Marine discharges are also known to contribute to radioactivity in coastal areas particularly in west Cumbria, via sea-to land transfer processes. Two major mechanisms for the transfer of radioactivity back to land have been identified: transfer in seaspray enriched with particle-associated nuclides (e.g., 59, 62) and onshore migration of contaminated sediments and their subsequent deposition in the intertidal zone and in tidally inundated areas (e.g., 4, 5, 25, 34, 37, 44, 56, 66, 71, 78, 83).

ITE carried out an extensive survey of tide-washed pastures in 17 estuaries spanning the eastern seaboard of the Irish Sea from the Solway (north of the Sellafield pipeline) to St. David's Head in south Wales between November 1991 and June 1992 (35, 83). Figure 4 shows the location of the estuaries surveyed. A uniform sampling methodology was used throughout this survey, thus making it possible to compare the data sets for each pasture. The activity concentrations of ^{137}Cs, ^{238}Pu, $^{239,240}Pu$ and ^{241}Am were measured in root mat and vegetation samples collected from each pasture.

The ranges of activity concentrations obtained in the survey are summarised in Tables 3 and 4. In general, the spatial distributions of the radionuclides were found to be consistent with their transport within the Irish Sea environment, with the highest activities occurring in the Esk estuary (closest to Sellafield) and lowest at the Welsh sites. There were exceptions to this trend in estuaries around Morecambe Bay (Leven and Kent); although these estuaries are geographically closer to Sellafield, water circulation within Liverpool Bay is known to be more efficient at transporting suspended materials to the estuaries further south than to the sheltered estuaries of Morecambe Bay (15, 16, 35). The data also show that Sellafield-derived radionuclides are near detection limits at tide-washed pastures located south of the Gwrfai estuary in Wales. The measured activity concentrations for vegetation samples were typically an order of magnitude less than for the root mat samples. These measurements were broadly in agreement with those reported by other workers (e.g., 7, 14, 15, 16, 44, 65).

Radiological significance

The principal method that has been used to assess the radiological impact of Sellafield discharges is to estimate doses to Man arising from several exposure pathways. The various pathways considered in dose assessments have been mentioned previously. There have been some investigations on the effect of Sellafield discharges on terrestrial and marine biota, where the doses and dose rates to various species have been evaluated (e.g., 45, 90); however, the majority of assessments have focused on doses to Man. Doses to the critical group, or to average individuals (adults or children), or a specified population (collective doses) have been estimated (e.g., 39, 43). This review will not attempt to compare the doses that have been derived in the many publications, as the dose assessment approaches can be different; rather, the results of some studies will be highlighted to provide a brief summary of the radiological significance of the Sellafield discharges.

Most dose assessment studies found in the published literature have concentrated around the Cumbrian coastal areas, primarily due to the relatively higher radionuclide concentrations compared to other sites and the free access of the public to these areas. Annual dose estimates resulting from exposure through the various pathways are reported in the BNFL and MAFF monitoring reports (e.g., 9-12, 52-54). The calculated doses would vary depending on the amounts of radioactivity discharged during any given year. Over the years of BNFL surveys, milk consumption has been identified as the major contributor to critical group dose from terrestrial foodstuffs, and an extensive milk sampling programme has been implemented (12). From the 1996 monitoring data, the estimated dose to 1 year old infants from milk consumption was 12 - 35 μSv, mainly due to ^{106}Ru, ^{90}Sr, ^{14}C and ^{137}Cs. However, this assessed dose is considered to be an overestimate because of the high detection limit for ^{106}Ru. The highest dose (to infants) from the consumption of other local produce (animal products, wild and cultivated fruits and vegetables) was 11 μSv. The estimated maximum dose to adults from ^{41}Ar (based on dispersion modelling of the discharge) was 36 μSv. Taking all pathways (food consumption, inhalation, beach occupancy, and exposure to ^{41}Ar and ^{85}Kr), the maximum critical group dose to 1-year-old infants for 1996 was estimated as 93 μSv. These assessed doses are broadly comparable to

Table 3. Range of activity concentrations of selected radionuclides in the root mat of tide-washed pastures in England and Wales bordering the Irish Sea, sampled in 1991/1992.

Estuary	Activity concentration (Bq kg^{-1} dw)			
	^{137}Cs	^{238}Pu	^{239,240}Pu	^{241}Am
Solway	720 - 2130	30 - 50	130 - 270	210 - 790
Esk	6170 - 9330	460 – 1150	1130 - 4750	3570 - 10230
Duddon	300-4940	50 - 440	210 - 2130	330 - 4250
Leven				
Ashes Point	345 - 2595	12 - 100	50 - 460	85 - 630
Creephaw & Wylock	370 - 3650	50 - 140	240 - 800	390 - 1450
Kent	430 - 895	4 - 14	35 - 70	80 - 105
Lune	1010 - 3500	40 - 70	160 - 330	240 - 590
Wyre	720 - 4130	25 - 50	120 - 280	250 - 850
Ribble	920 - 2610	30 - 50	120 - 270	280 - 610
Mersey	900 - 1920	22 - 28	110 - 150	180 - 330
Dee	2010 - 3130	20 - 32	100 - 160	260 - 430
Conway	360 - 1230	1.2 - 9.9	6 - 55	50 - 160
Anglesey	650 - 1080	2.2 - 8.5	11 - 48	14 - 110
Gwyrfai	850 - 1270	8.2 - 12	37 - 59	60 - 130
Dwyrdd	330 - 1270	<0.8	1.1 - 3	< 1
Mawddach	20 - 100	<0.5	1.1 - 2.2	< 1
Dovey/Dysynni				
Traeth Maelgwn	8 - 50	<0.3	0.6 - 1.6	< 1
Broad Water	2 - 110	<0.6 - 1.6	1.4 - 2	< 1
Teifi	5 - 120	<0.5	1.2 - 2.2	< 1

Table 4. Range of activity concentrations of selected radionuclides in vegetation from tide-washed pastures in England and Wales bordering the Irish Sea, sampled in 1991/1992.

Estuary	Activity concentration (Bq kg^{-1} dw)			
	^{137}Cs	^{238}Pu	^{239,240}Pu	^{241}Am
Solway	70 - 640	1.5 - 16	6 - 70	10 - 180
Esk	20 - 970	1.6 - 120	7 - 540	10 - 1260
Duddon	290 - 820	27 - 75	125 - 355	260 - 700
Leven				
Ashes Point	150 - 450	3.3 - 10	20 - 50	50 - 130
Creephaw & Wylock	230 - 340	13 - 36	60 - 170	150 - 350
Kent	8 - 305	0.3 - 7.2	1 - 36	20 - 70
Lune	300 - 560	8 - 18	40 - 80	90 - 190
Wyre	290 - 720	6 - 22	30 - 110	100 - 210
Ribble	140 - 350	3.6 - 7	19 - 40	50 - 110
Mersey	40 - 180	1.8 - 5.9	6 - 31	23 - 43
Dee	60 - 140	1 - 2.4	6 - 12	13 - 25
Conway	3 - 55	0.5 - 1.6	0.2 - 6.3	< 1 - 12
Anglesey	8 - 15	<0.1 - 0.2	0.3 - 1.3	< 1
Gwyrfai	5 - 105	< 0.07	0.2 - 8.8	< 1 - 14
Dwyrdd	45 - 220	< 0.07	0.1 - 0.4	< 1
Mawddach	2 - 9	< 0.1	0.1 - 0.3	< 1
Dovey/Dysynni				
Traeth Maelgwn	8 - 15	< 0.1	0.2 - 0.6	< 1
Broad Water	5 - 8	< 0.1	0.3 - 0.7	< 1
Teifi	3 - 12	< 0.1	0.3 - 0.8	< 1

those estimated by MAFF using their 1996 monitoring data, where the maximum dose to 1 year olds from the consumption of terrestrial foodstuffs was 55 μSv (54).

The doses derived from the radionuclide contamination in tide-washed pastures have been addressed in a number of studies. Dose assessment calculations were performed for a tide-washed pasture at the Esk estuary, which showed the highest contamination levels of the 17 pastures surveyed during 1991-92 (83). The dose due to external exposure would be a maximum of 530 μSv, assuming occupancy of 2000 h y^{-1} at the most contaminated spot on this pasture. Relatively lower doses (maximum of 15 μSv in adults from the consumption of beef liver) were calculated from the ingestion of animal products (along the soil-vegetation-grazing animal pathway). It was concluded that although relatively high activity concentrations could be found in a number of sites along the eastern seaboard of the Irish Sea, the doses from ingestion of various animal products are low because of the low bioavailability of sediment-associated radionuclides (13, 21, 89).

The present and future doses to people who make use of the Cumbrian coastal area have been assessed recently by the NRPB (87, 88). The study considered various terrestrial pathways that are affected by atmospheric as well as liquid discharges from Sellafield (excluding the consumption of marine foodstuffs). Annual doses to 'average' adults, children and infants in the Cumbrian area were estimated for 1989, 1993, 1996, 2006, 2050 and 2200, using the radionuclide discharge data up to 1989 and assuming certain scenarios regarding future operations at Sellafield for the subsequent years. The study found that the highest average individual doses (~ 25 μSv y^{-1} in 1989) were received by people living closest to Sellafield, with doses to adults slightly higher than for children and infants. Liquid discharges were the main contributor to the dose, via external irradiation from beach sand and from seaspray deposited on land, and inhalation following resuspension. The most important radionuclides were ^{60}Co, ^{137}Cs, $^{239,240}Pu$, ^{241}Pu and ^{241}Am. With the assumed scenarios in future years, it was predicted that about 60% of the total average dose in the year 2200 would be due to discharges prior to 1989, with the actual dose about two orders of magnitude less than the 1989 value. All predicted doses were below the ICRP limit of 1 mSv y^{-1} for a member of the public. Mayall *et al.* (58) extended this work to include doses from the ingestion of contaminated seafoods to individuals living in the vicinity of Sellafield. The highest predicted dose to average adults was 36 μSv y^{-1} in 1989, 90% of which was due to marine discharges. About 27% of the marine contribution was due to the ingestion of molluscs. The small contribution of seafoods is due to the fact that seafood makes up a small component of the diet for the average individual, with the further assumption that only 25% of the shellfish intake was locally-derived.

On a European-wide perspective, the European Commission set up Project MARINA in 1985 to evaluate the impact of natural and anthropogenic radionuclides in northern European marine waters (22). Mathematical models were used to estimate collective doses to the European population, as a basis for comparing the relative importance of the various sources of radioactivity. One of the working groups investigated the impact of discharges from 72 civil nuclear sites, including Sellafield. The significance of Sellafield liquid discharges as a source of artificial radioactivity in European waters

was highlighted in the report. In terms of total discharges up to 1984, Sellafield contributed 95.2% and 86.9% of the total alpha and beta emitters, respectively, from all nuclear sites. Thus, in terms of annual collective dose, the UK population would have received in 1979 (the peak year) about 60% (210 man Sv) of the collective dose rate delivered to the entire EC population. In terms of total collective dose up to the year 2500, the UK population would also receive 60% (3100 man Sv) of the total collective dose to the EC population. The major exposure pathway is fish consumption, accounting for 50% of the collective dose rate in 1979 and 40% of the total collective dose to the year 2500. Other important exposure pathways were consumption of molluscs and external exposure from beach occupancy. The major contributor to the dose was ^{137}Cs, but significant contributions also came from ^{239}Pu, ^{241}Pu, ^{106}Ru and ^{134}Cs. It was also pointed out that discharges from Sellafield gave rise to the highest critical group doses (up to 3.5 mSv y^{-1} during 1977 to 1986) from any nuclear site; these doses were expected to decline following the reduction in total discharges as a result of the operation of effluent treatment plants. Naturally-occurring radionuclides, however, dominate the total collective dose; it was estimated that the collective dose rate is 3400 man Sv y^{-1} to the EC population, the main contributor (80%) is the ingestion of ^{210}Po in seafoods. A similar conclusion was reached in a world-wide study comparing the doses arising from ^{137}Cs and ^{210}Po via the ingestion of marine foods (1).

The above studies illustrate the complexity of assessing the radiological significance of the Sellafield discharges. There are several sources of uncertainty in model predictions and these are further compounded by uncertainties in the historical records as well as in model parameters (such as food intake and habit data obtained from various surveys). It would appear from these assessments that the ICRP dose rate limit of 1 mSv y^{-1} is unlikely to have been exceeded even using pessimistic assumptions for the critical groups.

Health implications

Much controversy exists regarding the impact of Sellafield on the health of the population living in its vicinity. It is not possible to provide, in this report, a comprehensive review of the health issues that have been discussed over the many years since Sellafield started operations. There is a considerable literature on the epidemiology of radiation exposures, which is outside the scope of this review.

One issue that has remained very much a public concern is the allegation of an increased incidence of leukemia and other cancers in children and young adults living in the village of Seascale, near Sellafield. In November 1983, the UK government set up an independent inquiry (the Black Advisory Group) to investigate this allegation, and the NRPB undertook studies on behalf of this group (84, 85). The Group found that due to uncertainties in the epidemiological data and the radiation dose estimate data, no firm conclusion could be reached on any possible relationship between the incidence of leukaemia in Seascale and its proximity to Sellafield (80). The advisory group recommended that more detailed epidemiological studies be carried out to reduce the uncertainty in the data.

Following this allegation, it became important to establish an accurate historical record of the discharges from Sellafield; a comprehensive review of discharges from Sellafield from 1952 onwards was thus carried out in order to assess radiation doses to children in Seascale (84, 85). Another recent study addressing some of the sources of uncertainty pointed out in these earlier reports has now been published (31). Earlier records of atmospheric discharges only provided estimates, as radiochemical analysis of stack samples did not start until 1964. Annual discharges of fission products such as ^{90}Sr, ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{137}Cs and ^{144}Ce during 1958 to 1963 were estimated based on fuel irradiation and throughput rates. The unmonitored sources of atmospheric discharge from the site also contribute to the uncertainty in the historical data. Using total alpha and beta measurements and soil deposition data, Gray *et al.* (31) suggested that estimates of discharges of alpha activity prior to 1964 would have been greater than assumed in the 1986 report, but the annual discharges of total beta activity were comparable. However, it was concluded that the discharge data given in the 1986 report in general have been overestimated and hence conservative, and that the conclusion of that report (that the assessed exposure to environmental radioactivity could not account for the excess leukaemia) remains unchanged.

In 1990, Gardner and colleagues reported that the excess of childhood leukaemia in Seascale was linked to high radiation doses received by their fathers (working at Sellafield) before their children's conception (e.g., 2, 29). This has become known as the Gardner hypothesis. A recent survey has confirmed that there is indeed a higher incidence of leukaemia in children of 120,000 workers in Britain occupationally exposed to radiation; however, a dose-response relationship could not be established from the data, suggesting that there may be a factor (or factors) other than radiation (30). This latest study apparently provides overwhelming evidence against the Gardner hypothesis, but a satisfactory explanation to the excess leukaemia cases still needs to be found. Further research is currently being carried out.

There is also public concern over the continuing production of plutonium from reprocessing operations at Sellafield. Plutonium is a major concern because of its known toxicity, and the long half-life of its most widely used isotope, ^{239}Pu ($t_{1/2}$ = 24,100 y). Many studies have shown that when Pu taken into the body (via inhalation or food ingestion) enters the bloodstream, it can be retained in the lungs, liver, and skeleton (e.g., 19). A recent study investigated the $^{239+240}Pu$ contents of children's teeth across Britain and Ireland (69). These workers found a decreasing trend in Pu levels in teeth with increasing distance from Sellafield; the activity concentrations measured were 7.1 (± 4.1) mBq/kg at 0 to 50 miles from Sellafield, decreasing to 3.0 (± 2.7) mBq/kg at distances >150 miles. These results implicated Sellafield as the source of the Pu; however, it has to be noted that there is overlap in the data. Furthermore, the Pu contents of the samples were very low and the possible contribution of Pu derived from atmospheric weapons testing has not been discussed.

Summary

This report reviewed the technical operations at the Sellafield nuclear complex and provided an assessment of how these have contributed to radioactivity in the terrestrial and marine environment around west Cumbria and beyond. During more than 40 years of operation, there is considerable published material on Sellafield and its environmental, radiological and health impacts. This report was not intended as a comprehensive review; rather, the aim was to highlight the key issues that have been investigated over the years, particularly dealing with the radioecological aspects of Sellafield discharges to the environment.

Apart from the Windscale accident of October 1957, much of the terrestrial impact of atmospheric discharges from Sellafield has been confined to the area of west Cumbria. Thus, monitoring and dose assessment studies in the terrestrial environment tended to be of interest mainly to UK scientists. Although there is a large amount of literature on this subject, it is not as voluminous as the publications related to the marine discharges. The marine releases have been dispersed over a wide geographic area, and have generated interest in their applications as oceanographic tracers; marine studies thus involved a large group of investigators, not only within the UK but also internationally.

Although there has been a steady increase in fuel reprocessing operations at Sellafield, a general decline in both atmospheric and marine discharges for most radionuclides compared to the releases in the 1970s and early 1980s is now observed. This has resulted from additional waste treatment facilities that were introduced in the mid-1980s and early 1990s. Thus, the current (1997) discharges of ^{137}Cs and actinides (^{241}Am and isotopes of Pu) now constitute less than 1% of their earlier peak levels. There are exceptions to this trend; for example, marine discharges of ^{99}Tc have increased following the treatment of a backlog of stored wastes. Significant increases in ^{85}Kr release to the atmosphere have also occurred following the operation of THORP.

The Institute of Terrestrial Ecology has carried out investigations related to Sellafield discharges since the 1980s. A recent (1993) survey found higher activity concentrations of several radionuclides in grass and soil samples close to the Sellafield site compared to similar samples from other nuclear sites in England and Wales. When compared to background concentrations of some of the radionuclides derived from atmospheric weapons testing and the Chernobyl accident, it appears that Sellafield discharges may have contributed a significant proportion of Pu in soils close to its perimeter fence. In a 1991/92 survey covering tide-washed pastures in 17 major estuaries along the eastern seaboard of the Irish Sea, the highest contamination levels in sediments and vegetation samples were found at the Esk estuary (closest to the discharge pipeline). The contamination levels decreased with distance from Sellafield (with some exceptions); some of the sites further south in Wales did not show any measurable concentrations of Sellafield-derived radionuclides.

The radiological impact of the discharges has been assessed using estimates of human dose arising from different exposure pathways. The doses to critical groups and to

'average' members of the public (adults, children and infants), as well as collective doses, have been evaluated in numerous studies, including the regular monitoring activities performed by BNFL and MAFF. Several different approaches to dose modelling have been used. Even with overly conservative assumptions for the critical groups, it has been shown in general that the recommended annual dose limit of 1 mSv to a member of the public has not been exceeded. Nevertheless, Sellafield discharges have contributed the most significant component to the collective dose of the UK population from artificial radioactivity compared to other sources. The controversy regarding excess leukaemia in children of workers living near Sellafield remains a public concern, and has spurred many investigations. However, the epidemiological publications were outside the scope of this review.

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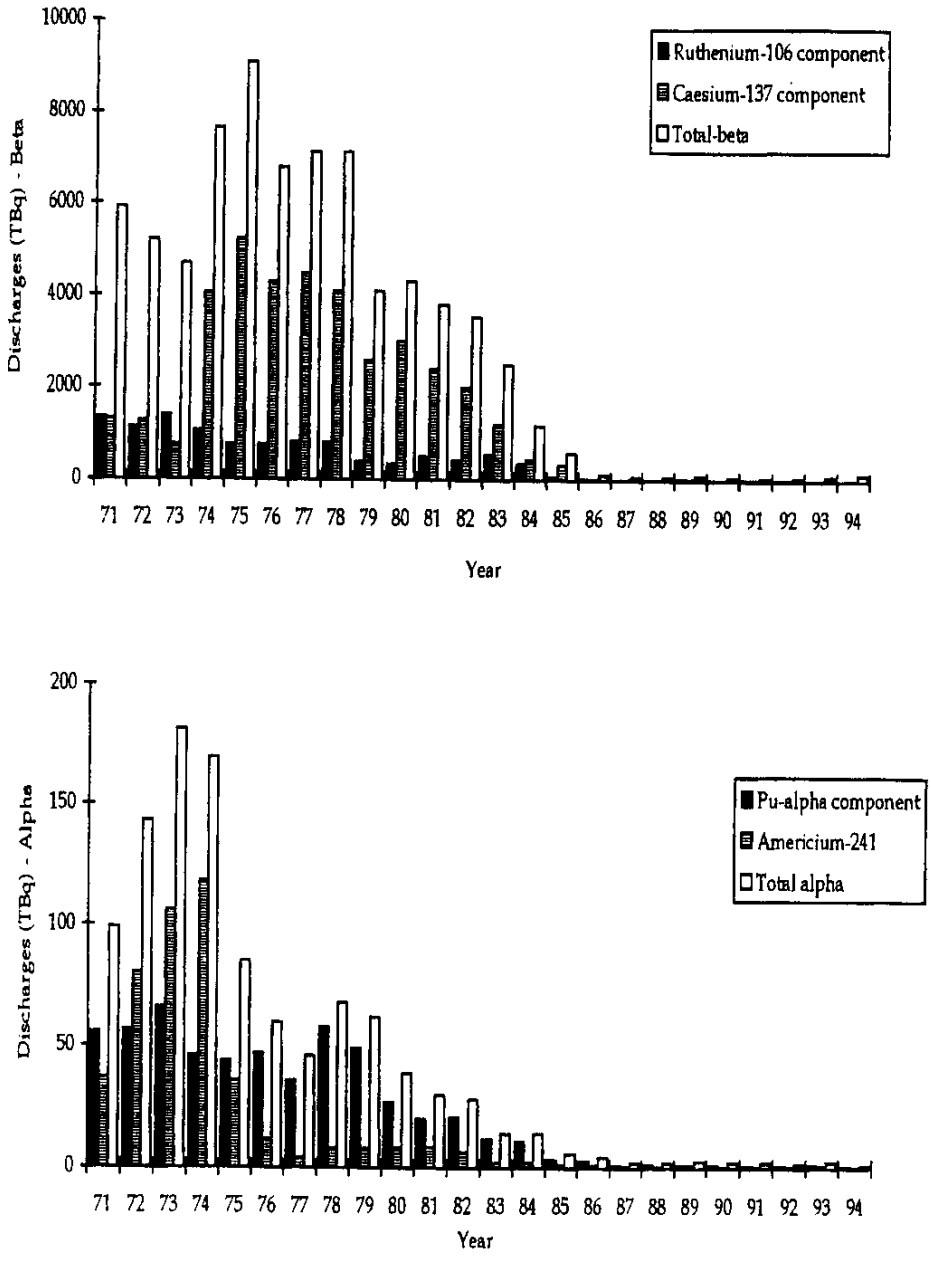

Figure 1 Marine pipeline discharges from Sellafield (from BNFL 1995)

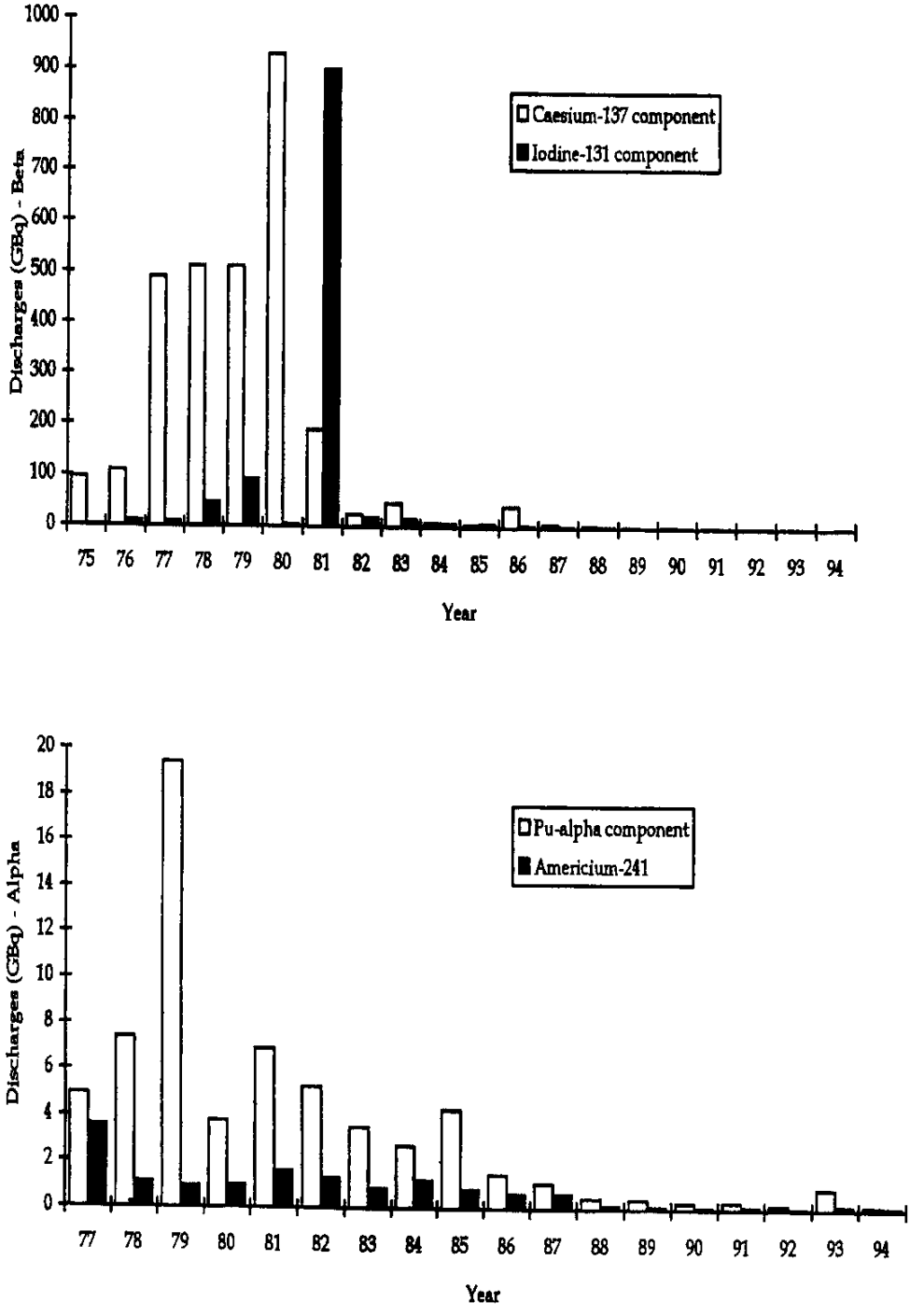

Figure 2 Discharges to atmosphere from Sellafield (from BNFL 1995)

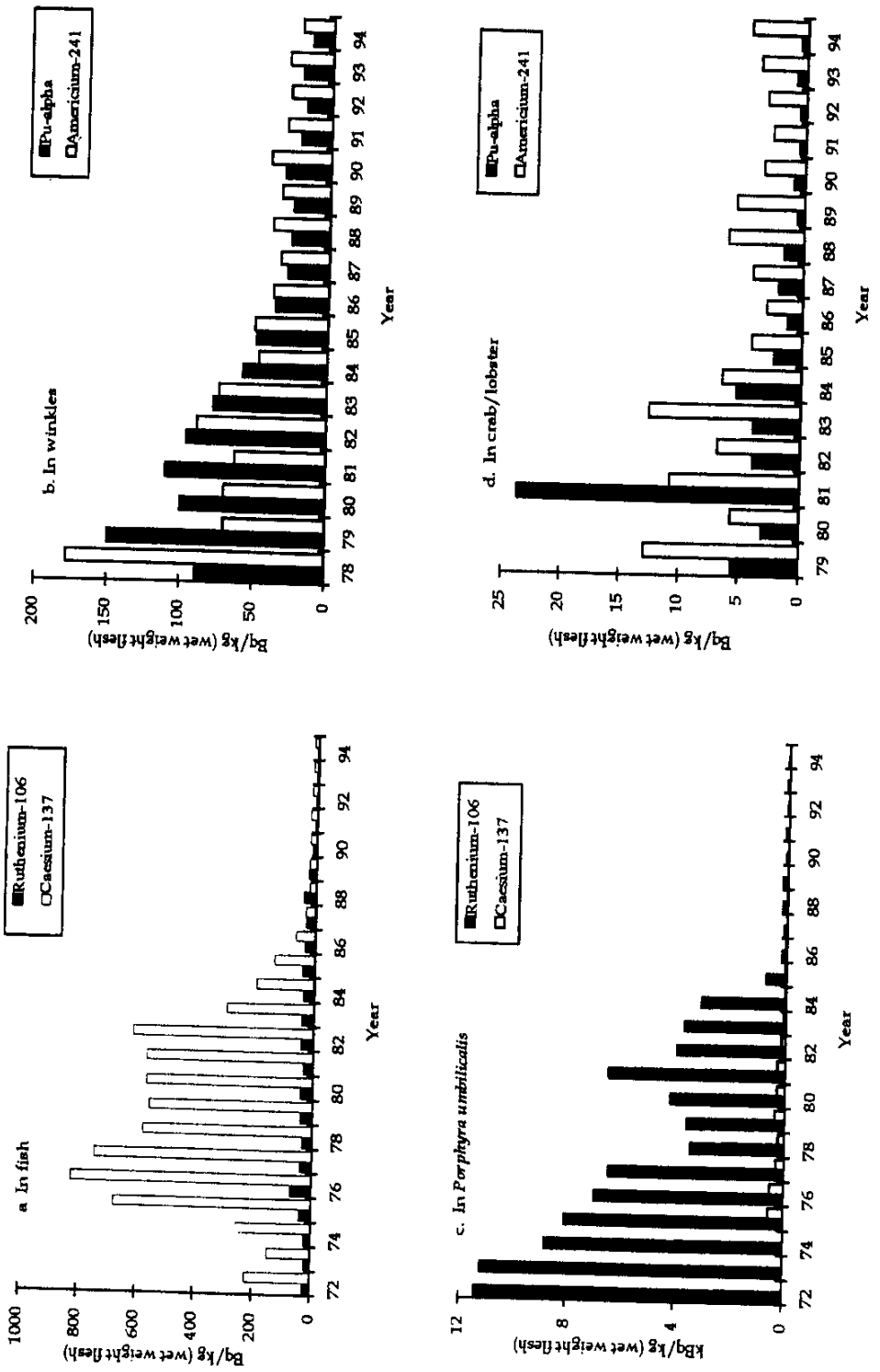

Figure 3 Radioactivity levels (from BNFL 1995)

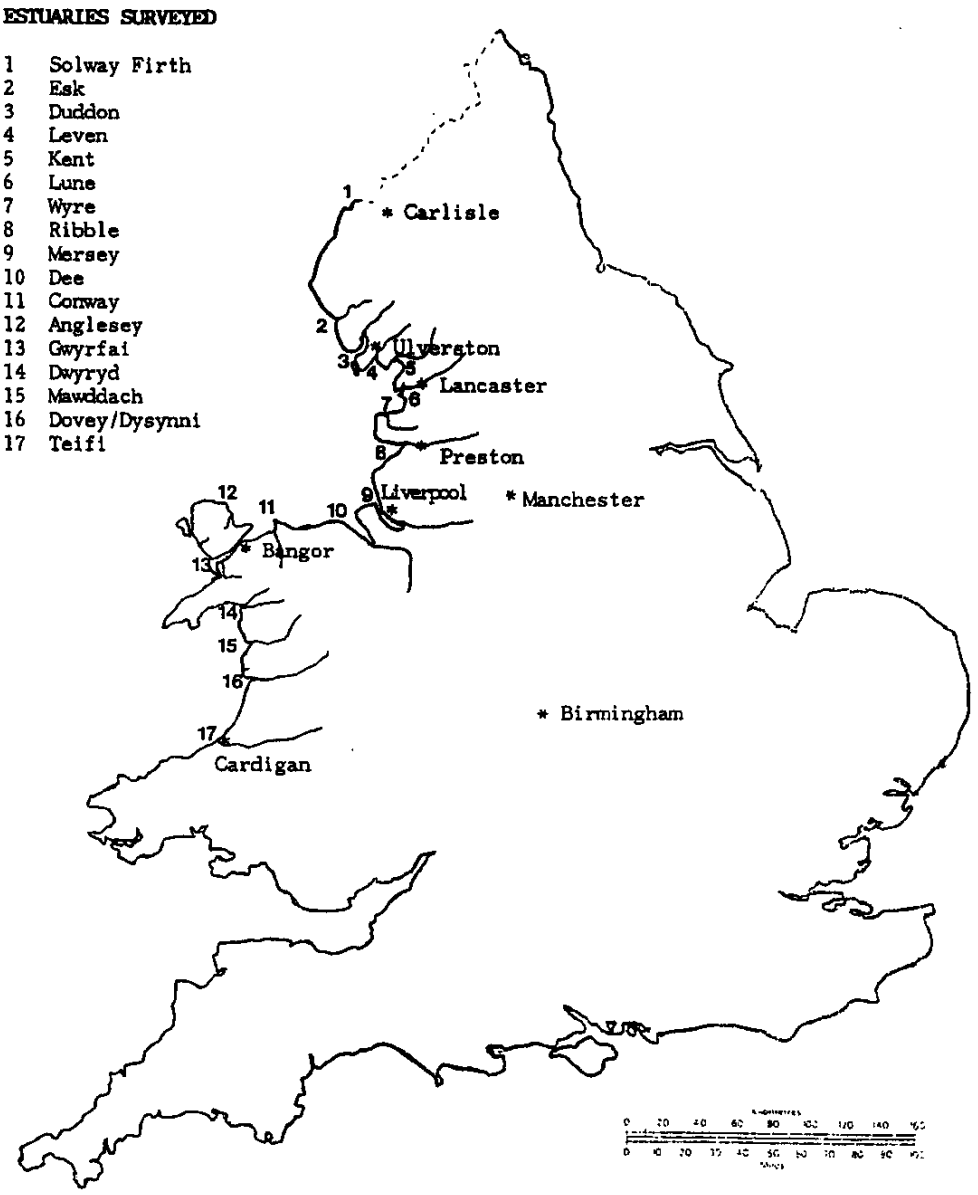

Figure 4 Location of tidally inundated pastures surveyed during 1991 - 1992

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