The environmental impact of the Sellafield discharges

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1. Introduction

Low-level liquid radioactive wastes from the British Nuclear Fuels pcl (BNFL) Sellafield establishment (formerly known as the Windscale and Calder Works) have been discharged into the north-eastern Irish Sea since operations began in the early 1950s. Although, at first, the major source of these discharges was the processing of nuclear fuel for the production of nuclear weapons, operations since the late 1950s have been dominated by the reprocessing of fuel from commercial nuclear power programs. The most important low-level radioactive wastes from Sellafield arise both in water used to purge the cooling ponds in which spent fuel elements (Magnox) are kept, and from the reprocessing plant whose low-level liquid waste are collected and neutralised in 'sea tanks' before being discharged around high water (Kershaw *et al.*, 1992). Discharges take place, under authorisation, through a series of pipelines extending 2.5 km seaward of the high water mark, with a very much smaller level of activity being discharged via the site's sewer (Jackson *et al.*, 2000). Discharges from Sellafield have always been subject to controls by regulatory governmental departments or agencies in the UK. Currently, authorisation to discharge is granted to BNFL by the Environment Agency under the Radioactive Substances Act, 1993 (United Kingdom – Parliament, 1993).

Over the years, the Sellafield discharges have acted as a large point source of a complex amalgam of α–, β– and γ–emitting radionuclides originating from both fission and neutron activation processes, and resulted in significant increases of artificial radionuclide concentrations in a variety of environmental compartments throughout the Irish Sea and beyond, with minute traces even detectable as far away as the Central Arctic Ocean. The aim of this overview is to present the current state of knowledge on the environmental impact of the Sellafield discharges. This, of course, mainly depends on the way in which discharged radionuclides are dispersed and accumulated on different key environmental compartments upon their introduction into the Irish Sea. The spatial and temporal distributions of Sellafield-derived radionuclides is governed by a number of complex mechanisms. Some important factors which influence their behaviour include the rate of the input from authorised discharges, their chemical speciation on the effluent and upon contact with sea water, prevailing hydrographic conditions, and interactions with suspended particles, sediments and biota. In this paper, the effects of these factors on the environmental impact of selected radiologically important radionuclides is emphasised, and the manner in which this knowledge can be used to predict future trends highlighted. Discussion is mainly limited to ^{137}Cs and transuranium nuclides, not only because they have been the subject of the most intensive study over the years, but also as representative of radionuclides exhibiting conservative and non-conservative behaviour. When appropriate, however, reference is made to other radionuclides.

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2. Liquid waste discharges from Sellafield

2.1. Discharge chronology

Discharges of liquid radioactive effluents from the site commenced in 1952, when a total of some 370 TBq of radioactivity was discharged to the north-eastern Irish Sea (Howells, 1966). A breakdown of the principal radioactive components of the effluents have been available since 1960, with more comprehensive data from 1978 onwards. Annual discharge figures have been published by the operator (BNFL) since 1979 and, in recent years, by the responsible Government Departments. Overall, the discharges of most radionuclides peaked in the mid- to late-1970s. As these discharges have been reviewed in detail elsewhere (Kershaw *et al.*, 1992; Gray *et al.*, 1995; Dunster, 1998), they will not be discussed in detail here. Suffice that their magnitude and composition have changed markedly in time, with the quantities of shorter-lived fission product nucllides such as ^{95}Zr, ^{106}Ru and ^{144}Ce declining steadily since the early 1970s, longer-lived nuclides such as ^{137}Cs peaking in the mid- to late 1970s and declining thereafter, and the major transuranics, ^{241}Am and ^{239,240}Pu peaking in the early- to mid-1970 and also declining thereafter (Figure 1). Factors responsible for such variations include the nature of the fuel, its burn-up time, the nature and duration of storage prior to reprocessing, the method of reprocessing and the nature of the effluent treatment plants, all of which have varied throughout the history of the Sellafield plant. Generally, by 1992, discharges were roughly two orders of magnitude or more lower than they were at their peak. Of the longer-lived radionuclides, estimated total activities of 49 PBq of ^{3}H, 6 PBq of ^{90}Sr, 6 PBq of ^{134}Cs, 41 PBq of ^{137}Cs, 120 TBq of ^{238}Pu, 611 TBq of ^{239,240}Pu, 22 PBq of ^{241}Pu, 540 TBq of ^{241}Am and smaller quantities of other transuranium nuclides have been discharged to the Irish Sea during the period 1952-98 (Gray *et al.*, 1995; BNFL, 1980-1999).

In 1985, the Site Ion Exchange Effluent Plant (SIXEP) and the Salt Evaporator waste treatment plants were commissioned, resulting in a reduction of radiocaesium discharges to relatively constant low levels. Since 1994, with the commissioning of the Enhanced Actinide Removal Plant (EARP), discharges of actinides were similarly reduced to relatively constant low-levels. However, the processing of an accumulated backlog of waste by this plant resulted in increased discharges of certain radionuclides and, in particular, ^{99}Tc.

2.2. Chemical form of the effluent

It is evident that a thorough knowledge of the different chemical and physico-chemical (solution, colloid and particulate) species from the Sellafield source term is required in order to understand the consequent environmental behaviour and transport of radionuclides following discharges into the Irish Sea (Leonard *et al.*, 1995). Despite problems associated to continuous changes in the chemical composition of effluent discharges and methodological difficulties in separation techniques, it has been possible to crudely characterise the different chemical and physico-chemical forms present in Sellafield effluents, thereby providing an initial assessment of the contribution of these individual forms to the discharge.

Analyses carried out in 1982 (and assumed to be typical of routine releases) showed that some 99% of the Pu(alpha), ^{241}Am and ^{243,244}Cm in the sea tanks, and about 60% of the ^{237}Np, were associated with particulate (>0.22 μm) material. In pond water effluents, the corresponding percentages were found to be somewhat lower (Pentreath *et al.*, 1984). Differences in chemical speciation were also noted, with reduced Pu(alpha) predominating in the filtrate of the effluent from the sea tanks and oxidised Pu(alpha) predominating in the filtrate of the effluent from the cooling ponds. However, as the bulk of the effluent came from the sea tanks, only about 1% of the total would appear to have been in an oxidised form upon

discharge. The nuclides of Am and Cm were present only in the Am(III) and Cm(III) forms, (i.e., chemically reduced) while almost 50% of the combined ^{237}Np discharges was in the oxidised, Np(V), form.

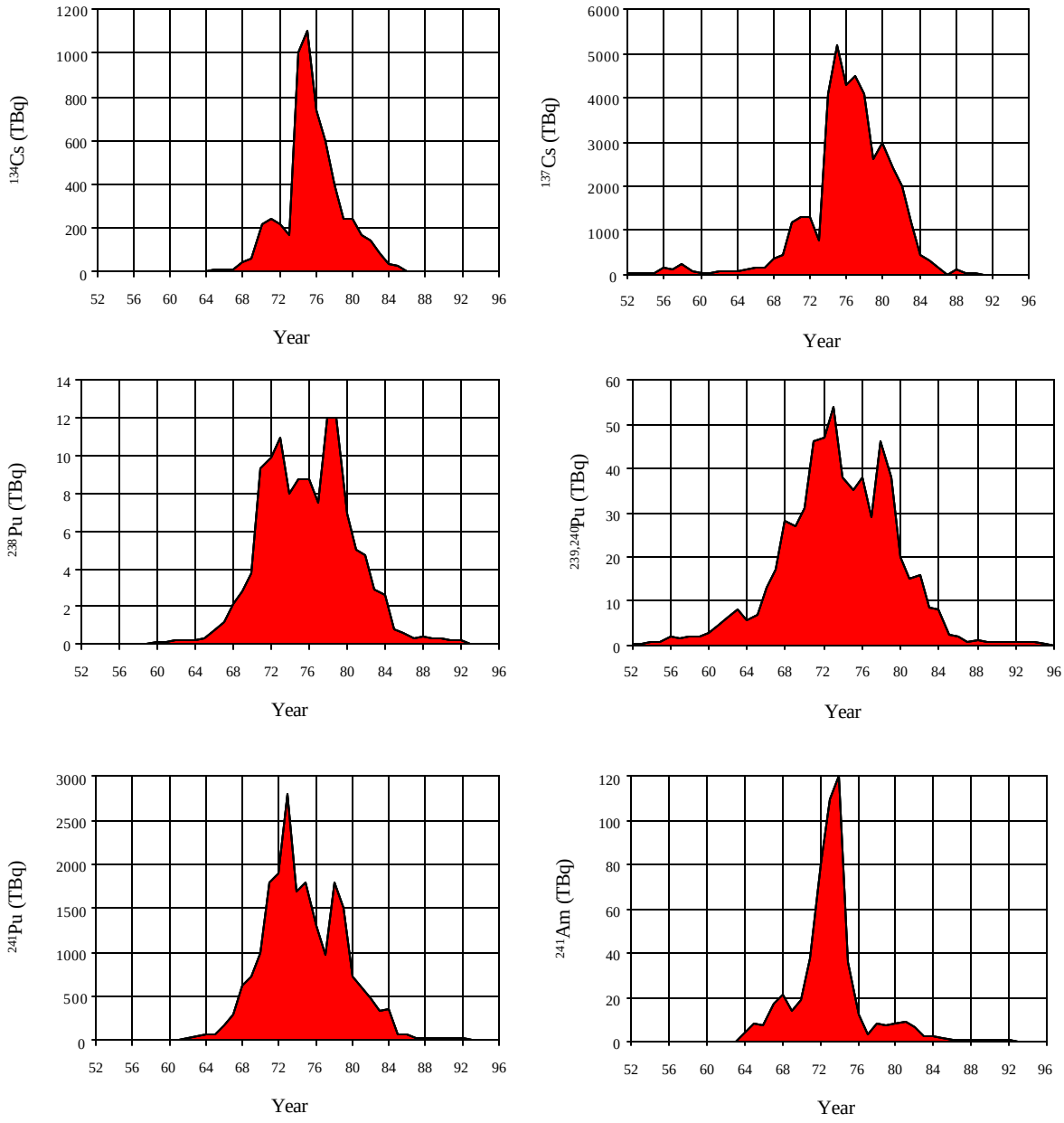

Figure 1. Sellafield pipeline discharges in the period 1952-94 for some radiologically important radionuclides (*source:* Gray *et al.*, 1995)

These observations have been supported by a more recent study carried out in 1991 on sea tank and SIXEP effluent streams (Leonard *et al.*, 1995). In the case of sea tank effluent, almost all of the activity of ^{239,240}Pu and ^{241}Am present was found to be associated with the iron floc material form upon the neutralisation of acid liquors containing ferrous sulphamate – a chemical used to control the valency of plutonium during fuel reprocessing. What little

plutonium was in the solution phase was determined to be in the reduced (tetravalent) form. Further, nuclides such as ^{137}Cs, ^{90}Sr and ^{99}Tc were found to be almost entirely in the solution phase. In SIXEP effluent, on the other hand, all of the radionuclides considered were almost entirely in a dissolved form, presumably as a result of the absence of particulate material in this waste stream.

Laboratory experiments to determine the colloidal size distribution of a suite of radionuclides in each of the effluent streams (SIXEP and seatank) have been carried out using ultrafiltration techniques (Leonard *et al.*, 1995). Overall, the results suggest that colloidal forms of individual radionuclides, originating from the solution phase, are more likely to occur in the SIXEP rather than in the sea tank effluent. Some typical data are given in Figure 2, and show clearly that in the SIXEP effluent significant fractions of the ^{90}Sr, ^{137}Cs, ^{239,240}Pu(V), ^{239,240}Pu(IV) and ^{241}Am are in a colloidally-bound form, as evidenced by the level of retention upon ultrafiltration (<3 kDa). In contrast, almost all of the ^{99}Tc in both waste streams is in a truly dissolved form.

'Hot particles', likely to contain fuel fragments, have also been identified in the effluent (Pentreath *et al.*, 1984) and have been shown to persist in the marine environment close to Sellafield for at least several months, with some being preserved in accreting estuarine sediments (Hamilton, 1981; Hamilton *et al.*, 1991).

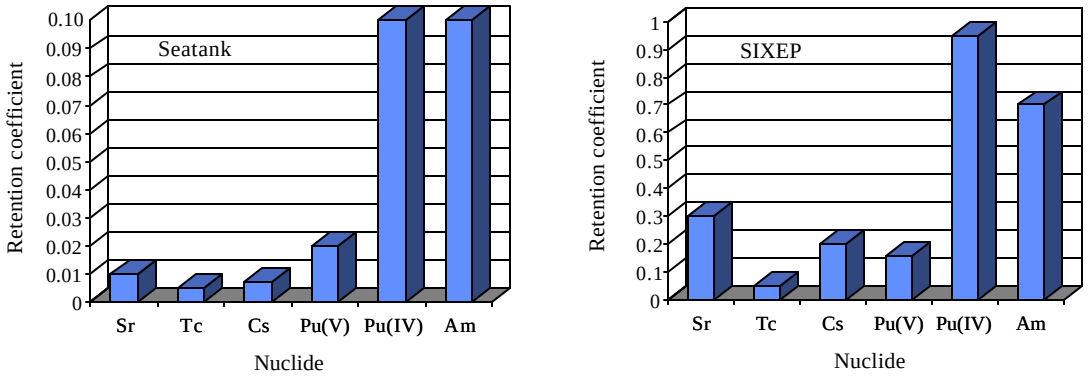

Figure 2. Radionuclide fractionation of sea tank and SIXEP effluent samples upon ultrafiltration (*source:* Leonard *et al.*, 1995). Note the order of magnitude difference in scale.

2.3. Chemical form upon dilution with sea water

Laboratory experiments involving a 1:10^4 dilution of sea tank effluent samples into sea water revealed a progressive solubilisation of the predominantly particle-bound plutonium, over a period of almost a year, accompanied by a change in the oxidation state distribution of plutonium from the reduced to the oxidised form. In contrast, for the pond effluent, the degree of solubilisation was considerably less, and a shift to a lower oxidation state was observed (Pentreath *et al.*, 1986). Changes in ^{241}Am solubilisation were also observed, with ≈30% of the sea tank and pond effluent being solubilised over a period of 360 days.

Studies on the physico-chemical speciation of the transuranics in open waters throughout the Irish Sea have shown that a significant proportion of the total Pu and even more of the Am are associated with the particulate phase (Pentreath *et al.*, 1986; Kershaw *et al.*, 1992; Mitchell *et al.*, 1991a; 1995; 1999). This contrasts with ^{90}Sr, ^{137}Cs and ^{99}Tc, which show comparatively little affinity for suspended particulate or, indeed, seabed sediment.

Measurements of the oxidation state distribution of Pu in filtered water sampled throughout the Irish Sea show little variation, spatially or temporally, with the bulk of the Pu being in the oxidised, Pu(V), state (Lovett and Nelson, 1981; Nelson and Lovett, 1978; Pentreath *et al.*, 1986; Mitchell *et al.*, 1991a; 1995).

Although, as discussed above, small amounts of ^{90}Sr, ^{137}Cs, ^{239,240}Pu(V) in colloidal form were identified in SIXEP effluent, further experiments by Leonard *et al.* (1995), following the dilution of effluent into sea water under laboratory conditions, indicated that these form did not persist in sea water. On the other hand, the same researchers did find evidence to suggest that colloidal forms of ^{239,240}Pu(IV) and ^{241}Am do persist in sea water. Despite these observations, carefully-designed field experiments using tangential flow ultrafiltration and selective sorption techniques have shown no evidence of any colloidal component for plutonium and americium in open Irish Sea waters (Downes, 1999; Mitchell *et al.*, in press). This suggests that transport of radionuclides via a colloid pathway is unlikely, at least in the immediate vicinity of the release. However, further work is require to investigate possible transformations in physico-chemical behaviour over much longer time scales and further away from the discharge, as it is possible that the interaction of radionuclides with other colloidal sources, such as naturally-occurring complexing ligands, may contribute to the overall transport of radionuclides (Leonard *et al.*, 1995).

3. Artificial radionuclide distributions and dispersion processes

The initial dispersion of radionuclides from Sellafield is influenced by a number of factors including variations in the discharge rate, the chemical form of the radionuclides in the effluent, local hydrographic conditions and the distribution and composition of bottom sediments. Data collected since the early 1960s show that those nuclides which are relatively soluble in sea water (e.g., ^{99}Tc, ^{137}Cs, ^{90}Sr, ^{3}H) are advected principally in a northerly direction, associated with the mean flow of water currents, leaving the Irish Sea via the North Channel, with a mean transit time of about one year (Bowden, 1955; Jefferies *et al.*, 1982; McKay *et al.*, 1986). The main flow passes northward via input of Atlantic water from St. George's Channel to the west of the Isle of Man. A minor component of the flow enters the eastern Irish Sea to the north of Anglesey and moves anti-clockwise round the Isle of Man before rejoining the main flow to exit through the North Channel (Howarth, 1984). A much smaller proportion of the discharges is dispersed into the southern Irish Sea, mainly due to turbulent diffusion driven by the tides and wind.

To illustrate, the distribution of ^{137}Cs in filtered surface sea water in the Irish Sea over the period 1980-85 is given in Figure 3. As a result of this dispersion pattern, much of the ^{137}Cs activity discharged from Sellafield has now left the Irish Sea, following a well-defined path around the west and north of Scotland, where it enters the North Sea (Jefferies *et al.*, 1973; Baxter *et al.*, 1979; Mauchline, 1980; McKinley *et al.*, 1981). From here it is advected northwards via the Norwegian coastal current (Holm *et al.*, 1983; Kautsky, 1988) before branching off northern Norway. One branch passes eastwards into the Barents Sea (Vakulovski *et al.*, 1985) and the other passes through the Fram Strait into the Nansen Basin (Smith *et al.*, 1990). Comparison of time-series concentration data with the actual discharges have allowed the transit times to each of these zones to be established: 2–3 years to East Scotland; 3–4 years to West Norway and 4–5 years to the Barents Sea (Livingston *et al.*, 1982; Dahlgaard *et al.*, 1986; Hallstadius *et al.*, 1986; Kershaw and Baxter, 1995; Dahlgaard, 1995). Despite its predominantly conservative behaviour, a certain proportion of the radiocaesium has been absorbed onto sediments, and a considerable inventory still resides in the Irish Sea.

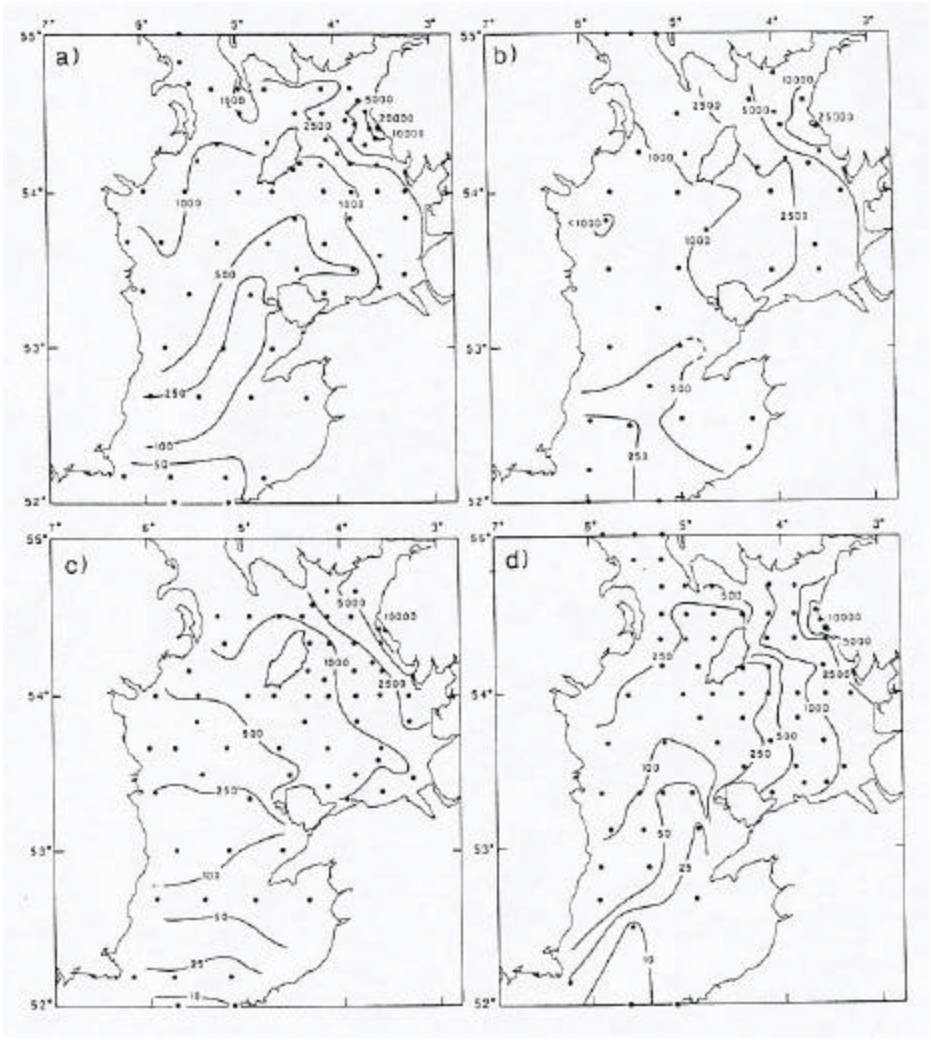

Figure 3. Concentration of ^{137}Cs (Bq m^{-3}) in filtered water from the Irish Sea: (a) April 1980; (b) March 1981; (c) November 1982; (d) March–April 1985 (*from:* Jefferies and Steele, 19889)

Another fission product, ^{99}Tc, is less susceptible to removal from the water column than caesium and, therefore, offers a better tool to study the movement of conservative radionuclides within and from the Irish Sea. Indeed, the relatively low level of the ^{99}Tc discharges over a long period prior to the commencement of the EARP operations in 1994, and the stepped nature of the ^{99}Tc discharge from this plant, provided a unique opportunity to study the dispersion of effluent (Leonard *et al.*, 1997). Prior to the introduction of EARP, levels of ^{99}Tc in filtered sea water were essentially uniform (1–4 mBq l^{-1}) for most of the Irish Sea, reflecting residual concentrations of a well-mixed system (Figure 4). Elevated levels were only observed in the immediate vicinity of the discharge point, and likely reflect the influence of discharges taking place at the time of collection. Along a large section of the Cumbrian and southern Scottish coastline, ^{99}Tc concentrations were greater by approximately an order of magnitude compared to levels in the west and south. The data from the post-EARP surveys showed a significant change in the magnitude of ^{99}Tc concentrations in sea water (Figure 4), with a marked migration of the effluent toward the Southern Scottish coast and the entrance of the North Channel in a relatively brief time period. On the basis of these surveys, an estimated transit time of ^{99}Tc to the North Channel of ≈6 months was derived, which suggests that the transport of dissolved radionuclides from Sellafield out of the Irish Sea can take place in a much shorter time scale than previously thought.

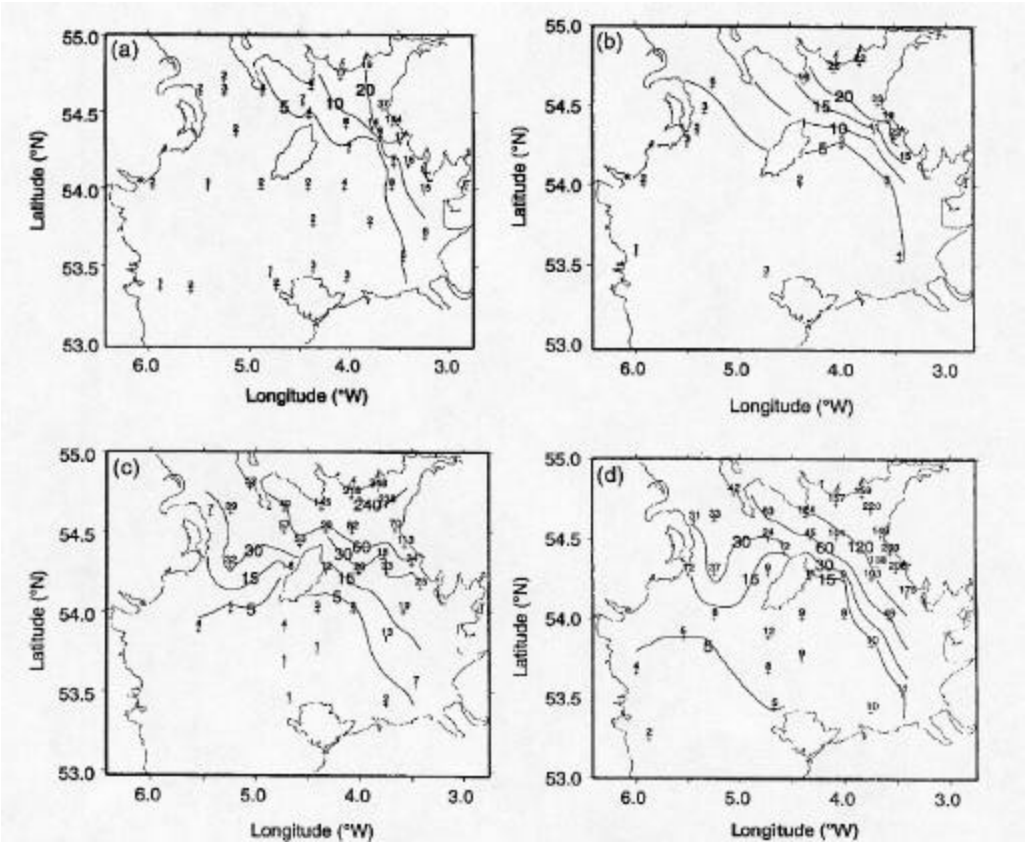

Figure 4. Contour map of the ^{99}Tc distribution in the Irish Sea, together with sea water sampling positions and concentrations (mBq l^{-1}). (a) Pre-EARP, December 1992; (b) Pre-EARP, December 1993; (c) Post-EARP, May 1994; (d) Post-EARP, December 1994 (*from:* Leonard et al., 1997)

In contrast, non-conservative radionuclides, such as Pu and Am, are quickly removed from the water column by direct precipitation or by absorption on suspended particulate matter. The behaviour and distribution of these nuclides is closely linked to that of the finer-grained seabed sediments. In the Irish Sea, muddy sediments are confined to two main areas: an extensive belt of muds and muddy sands parallel to the Cumbrian coast, and a large and relatively deep (≈ 100 m) basin lying between the Isle of Man and the coast of Ireland. Studies have confirmed that these muddy areas are a significant (temporary) sink for many of the radionuclides discharged from Sellafield, with by far the largest repository being the first mentioned (Hetherington, 1978; Pentreath *et al.*, 1986). These sediments are subject to tidal and wave mixing, as well as to extensive reworking by benthic organisms. Although the accretion rate of offshore muds in the vicinity of Sellafield appears to be low (Kershaw *et al.*, 1988a), it has been shown that bioturbation gives rise to a redistribution of the sediment (and associated radionuclides) to depths in excess of a metre (Kershaw *et al.*, 1983, 1984; 1988b Swift and Kershaw, 1986; Pentreath, 1987). A further source of disturbance is trawling, which can cause the resuspension of considerable quantities of surface sediment and result in the homogenisation of the radionuclide signal in the upper few tens of centimetres (Kershaw *et al.*, 1999). In the western Irish Sea mud basin, there is evidence that deposition of fine sediments is still taking place (Belderson, 1964; Condren *et al.*, 1996; Mitchell *et al.*, 1999).

A number of studies have been carried out on the distribution of transuranic nuclides in filtered waters of the Irish Sea (Hetherington *et al.*, 1975; Pentreath *et al.*, 1984; McKay and Walker, 1990; Walker and McKay, 1991; Mitchell *et al.*, 1991a; 1995; 1999; Woodhead and Pentreath, 1992; Cooke *et al.*, 1997, Leonard *et al.*, 1999). Although the concentration of

plutonium and americium differed depending on the dates of the surveys, a marked decrease in concentrations with distance from Sellafield was apparent in all occasions. In the immediate vicinity of the Sellafield pipeline (within 10 km), levels were greater by approximately an order of magnitude compared with concentrations observed to the west of the Isle of Man (Leonard *et al.*, 1999). The highest concentrations of plutonium and americium were generally observed close to the Cumbrian and southern Scottish coastline (Figure 5). Levels close to Sellafield were variable, and concentrations were not necessarily greatest at the point of discharge.

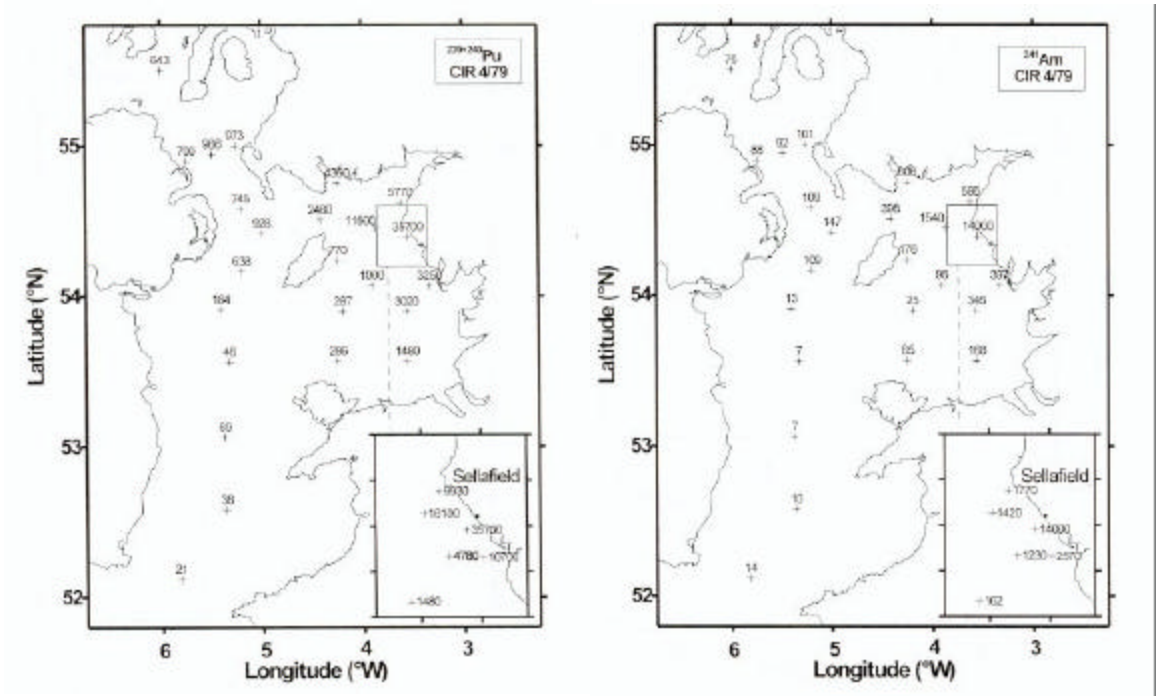

Figure 5. Distributions of dissolved ^{239,240}Pu and ^{241}Am (mBq m^{-3}) in surface waters of the Irish Sea in 1974 (*from:* Leonard *et al.*, 1999)

A similar distribution pattern was found for ^{239,240}Pu and ^{241}Am concentrations in suspended particulate in a survey carried out in December 1995, with concentrations falling markedly with distance from Sellafield (León Vintró, 1997). A clear difference was observed, however, between the dispersion of particulate Pu and Am in a north-westerly and south-westerly direction, with a clear bias towards the former. This confirms the observation made in previous studies (Aston and Stanners, 1981; McDonald *et al.*, 1990; Boust *et al.*, 1996) that the advection of particle-bound plutonium might be more active in this direction. Indeed, it had been shown that by the mid-1980s, the movement of contaminated silt was the dominant mechanism of supply of Sellafield-discharged plutonium and americium to the south-west coast of Scotland (McDonald *et al.*, 1990). For this particular survey, 53–95% of the plutonium and 89–98% of the americium were found to be associated with the solid phase (>0.45 μm). This contrasts with the partitioning observed in samples collected throughout the Irish Sea during previous surveys, where the percentages of plutonium and americium in a particulate form were significantly lower (Mitchell *et al.*, 1995). Similar variations within the same general zone have been reported by other workers (McKay and Pattenden, 1993; Vives i Batlle, 1993) and attributed to differences in suspended particulate loads at the time of sampling. Suspended loads in the Irish Sea typically lie within the range of 1 to 10 mg l^{-1}. The importance of suspended load in controlling the partitioning of transuranic nuclides

between the filtered and suspended particulate fractions is illustrated in Figure 6, where the proportions of plutonium and americium in solution are plotted against suspended particulate loading. It is clear from these graphs that temporal and spatial variations in the partitioning of plutonium and americium between the dissolved and particulate phases in open Irish Sea waters can largely be explained by variations in the particulate loading. These, of course, depend on a number of factors including seasonal changes, weather conditions, depth, nature of the underlying sediments, etc.

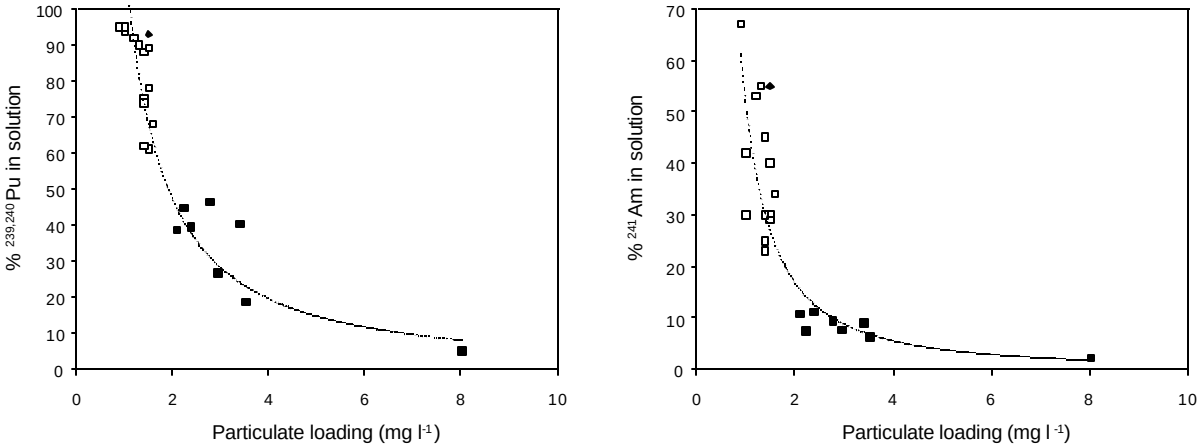

Figure 6. Relationship between dissolved ^{241}Am and particulate loading: ( ) León Vintró, 1997; (□) Walker and McKay, 1991; (◆) Vives i Batlle, 1993

Despite the rapid decline in transuranic concentrations in sea water with distance from the discharge point, the Sellafield signal has been sufficiently large for enhanced concentrations to be detected readily in the North Sea and along the Norwegian Coastal Current and into the Barents Sea (Kershaw and Baxter, 1995). On this evidence, it is clear that plutonium from Sellafield has been transported in solution over distances up to 2500 km from the source.

4. Temporal evolution of radionuclide concentrations in key marine compartments

4.1. Sea water

Surveys of the concentration of radiocaesium in filtered water from throughout the Irish Sea have been conducted on a regular basis for many years (MAFF, 1967–99; Cunningham and O'Grady, 1986; Cunningham *et al.*, 1988; O'Grady and Currivan, 1990; McGarry *et al.*, 1994; Pollard *et al.*, 1996; Long *et al.*, 1998a; Ryan *et al.*, 2000). The extensive data available show that general levels reached a peak in the years between 1975 and 1978 and have been declining more or less steadily ever since in line with the reduction in annual discharges. At their peak, ^{137}Cs concentrations in the north-eastern Irish Sea, close to the Sellafield discharge point, attained 5-50 Bq l^{-1} and were more than three orders of magnitude higher than representative fallout levels at similar latitudes. By 1994, concentrations in the north-eastern Irish Sea had declined to between 0.1 and 1.0 Bq l^{-1}. As an illustration, measured ^{137}Cs concentrations in the vicinity of Sellafield (St Bees and Seascale) as measured and tabulated by Hunt and Kershaw (1990) are shown in Figure 7. A semi-empirical model,

proposed by Nicholson and Hunt (1995), has been applied to the data in order to derive concentrations for the years prior to sampling (Condren, 1998). A similar trend is observed in the western Irish Sea, with ^{137}Cs levels peaking in the mid 1970s and steadily declining thereafter (Figure 8). In general, concentrations prevailing in the western Irish Sea in any given year are ≈20 times lower than those in the vicinity of the discharge point.

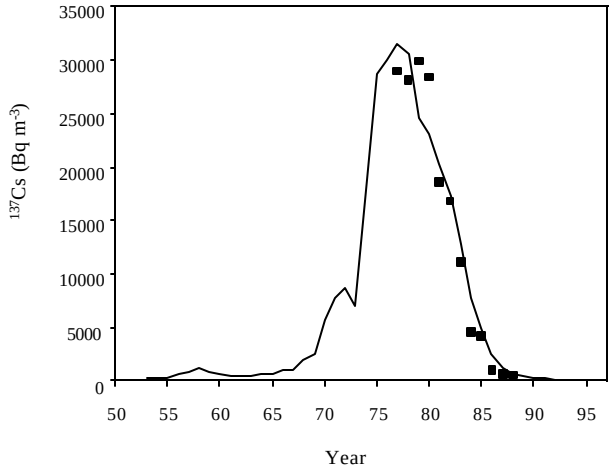

Figure 7. Predicted and measured ^{137}Cs concentrations in sea water in the Sellafield offshore vicinity (experimental data from: Hunt and Kershaw, 1990)

Detailed studies of the ^{137}Cs concentrations in sea water in relation to the Sellafield discharge rate have revealed that most of the radiocaesium present in the water column since the mid-1980s does not arise directly from the discharges, but instead is due to the progressive remobilisation of ^{137}Cs from historically contaminated sediments (Hunt and Kershaw, 1990; Condren, 1998). These observations are in good agreement with budget calculations by Poole *et al.* (1997), which indicate that the predominant contribution (72–80%) of ^{137}Cs in the water column of the Irish Sea arises from the flux from the seabed.

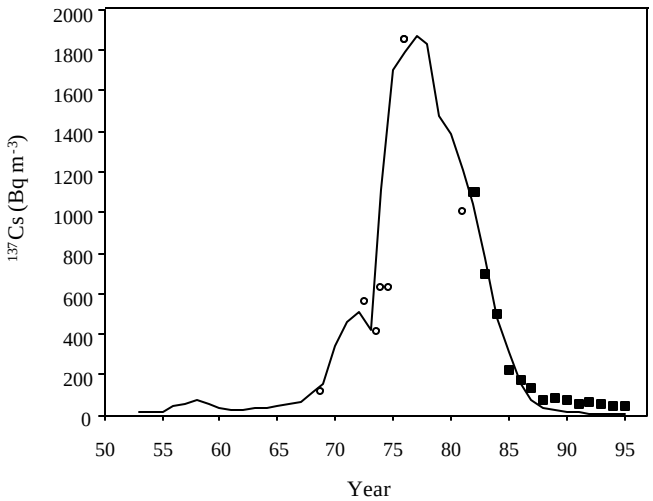

Figure 8. Predicted (—) and measured ( ) ^{137}Cs concentrations in sea water in the western Irish Sea (experimental data from Long *et al.*, 1996). The predictions have been validated with experimental data (○) from MAFF (1968-81)

Extensive data on the distribution of ^{239,240}Pu in filtered seawater throughout the Irish Sea have recently been reported by Leonard *et al.* (1999). By comparing the combined median concentrations obtained from surveys carried out in the 1970s with a similar combined median value for post-1985 surveys, these authors have shown that plutonium concentrations in the coastal region of Cumbria have fallen by a factor of ≈4 between the two surveying periods. In contrast, comparatively little change in transuranic concentrations appears to have taken place over the same period (Mitchell *et al.*, 1999; Leonard *et al.*, 1999).

4.2. Seabed sediments

The spatial and temporal variations of a wide range of radionuclides in the surface sediments of the Irish Sea have been determined from a large number of samples collected over the period 1968-96 (McCartney *et al.*, 1994; Poole *et al.*, 1997; Kershaw *et al.*, 1999). The most noticeable feature of the trends in the concentrations of these radionuclides in sediments sampled close to the outfall in this period is a general reflection of the reduction of discharges since 1978, the effect being more pronounced for sites closer to the source.

Presently, concentrations of radionuclides such as ^{137}Cs, ^{239,240}Pu and ^{241}Am in muddy sediments near the outfall are in the ranges 500–1,500 Bq kg^{-1}, 500–600 Bq kg^{-1} and 700–900 Bq kg^{-1}, respectively (MAFF, 1967-99). Time-series analyses in surface sediment samples collected in this zone show mean availability times of 2–5 years for radiocaesium and 5–8 years for plutonium (Hunt, 1985; MacKenzie *et al.*, 1994; Nicholson and Hunt, 1995). As an illustration, the evolution of measured ^{137}Cs and ^{241}Am concentrations in sediments from Newbiggin in the Ravenglass Estuary near Sellafield is shown in Figure 9, together with the predictions of a simple, semi-empirical model (Nicholson and Hunt, 1995). As expected, the rate of reduction is least for nuclides which exhibit strong sediment-seeking properties. Further afield, this (temporal) reduction in transuranium concentrations is not always evident. Indeed, comparison of the most recent data set of ^{239,240}Pu and ^{241}Am concentrations in surface sediments with earlier surveys shows the concentration isopleths to have been displaced northwards, with a progressive decrease in both surface concentrations and inventories near the Cumbrian coast and increases at more distant sites (McCartney *et al.*, 1994; Kershaw *et al.*, 1999). Similar behaviour was previously observed in the case of ^{137}Cs (McCartney *et al.*, 1994).

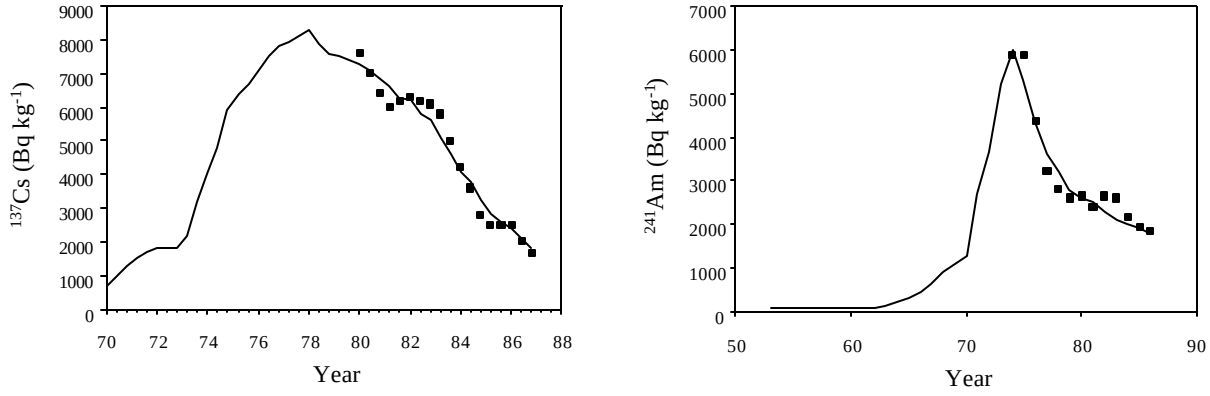

Figure 9. Temporal evolution of measured (■) and model-predicted (—) concentrations of ^{137}Cs and ^{241}Am in surficial sediments sampled at Newbiggin in the Ravenglass Estuary (experimental data from Nicholson and Hunt, 1995)

Time-series analyses of western Irish Sea surface sediments have shown that, while ^{137}Cs concentrations have fallen by a factor of between 2 and 5 since their peak in 1980-81, ^{239,240}Pu concentrations have remained relatively constant (Figure 10). Model predictions for the western Irish Sea mud basin indicate that, were discharges to remain at their present levels, ^{137}Cs concentrations will continue to decrease with mean availability times in the range of 7-17 years, while little or no reduction in ^{239,240}Pu concentrations is likely to occur for some decades to come (Mitchell *et al.*, 1999). At the present time, general transuranic concentrations in western Irish Sea muddy sediments are almost two orders of magnitude lower than the corresponding concentrations in the north-eastern Irish Sea, while ^{137}Cs concentrations are little more than an order of magnitude lower.

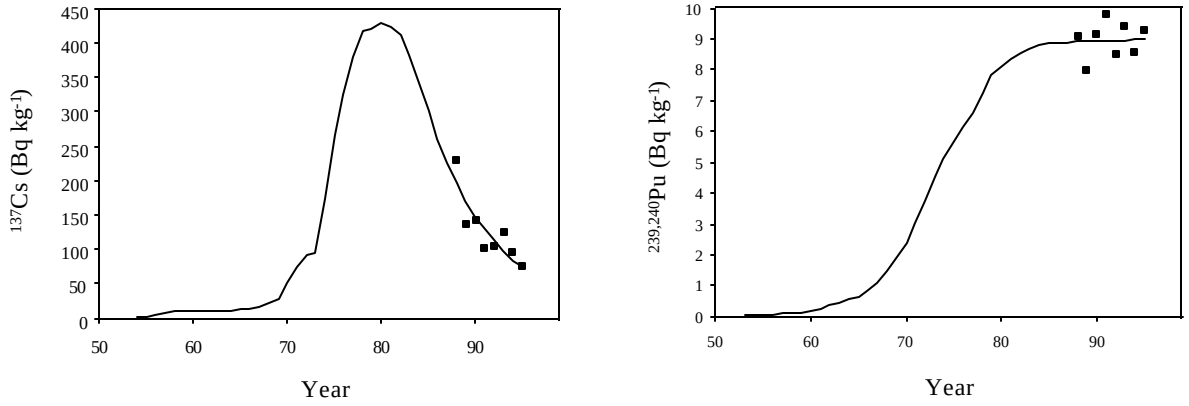

Figure 10. Temporal evolution of measured (■) and model-predicted (—) concentrations of ^{137}Cs and ^{239,240}Pu in surficial sediments sampled in the western Irish Sea (*from:* Mitchell *et al.*, 1999)

4.3. Plutonium isotopic ratios in surficial sediment and sea water

^{238}Pu/^{239,240}Pu and ^{241}Pu/^{239,240}Pu activity ratios in filtered sea water, suspended particulate and surficial sediments throughout the Irish Sea have been found to be consistently lower than those reported for recent discharges, and closely reflect those of the time-integrated or cumulative Sellafield discharge (MacKenzie *et al.*, 1994; Mitchell *et al.*, 1999; MAFF, 1980-99). This observation can best be explained by 'pre-depositional' mixing of contemporary and past discharges (Hunt, 1985; Kershaw *et al.*, 1990; MacKenzie *et al.*, 1994), rather than 'post-depositional' mixing *in situ.* As already stated, mixing is believed to take place mainly in the muddy sediments close to the Sellafield outfall, where a combination of bioturbation and very low sediment accumulation rates leads to extensive vertical mixing (Kershaw *et al.*, 1988a; Kershaw, 1986; Kershaw *et al.*, 1990).

To illustrate, annually recorded ^{238}Pu/^{239,240}Pu and ^{241}Pu/^{239,240}Pu activity ratios in surficial western Irish Sea sediments (corrected for the contribution of weapons fallout and averaged over three time-series stations in the western Irish Sea) are shown in Figure 11, together with activity ratios derived from annual and cumulative discharges (decay-corrected to the year in question). The mean ^{238}Pu/^{239,240}Pu derived from this data, at 0.174 ± 0.015 (n = 30; Mitchell *et al.*, 1999), is identical to previously reported values for marine materials (*Fucus*, sediment and water) in the western Irish Sea (Mitchell *et al.*, 1987; 1995), but somewhat lower than that observed along the Cumbrian and south-western Scottish coasts over the same period, namely 0.212 ± 0.009 (n = 186; MAFF, 1989-99). Although small, this difference appears to be real and has been attributed to the slow rate at which plutonium is dispersed following discharge, largely as a result of active sediment mixing and re-working in

the general vicinity of the release point. Moreover, surficial sediments near Sellafield are labelled with a relatively high proportion of plutonium from recent discharges, characterised by a higher $^{238}Pu/^{239,240}Pu$ activity ratio. Further afield, the proportion from earlier discharges increases, resulting in lower $^{238}Pu/^{239,240}Pu$ activity ratios.

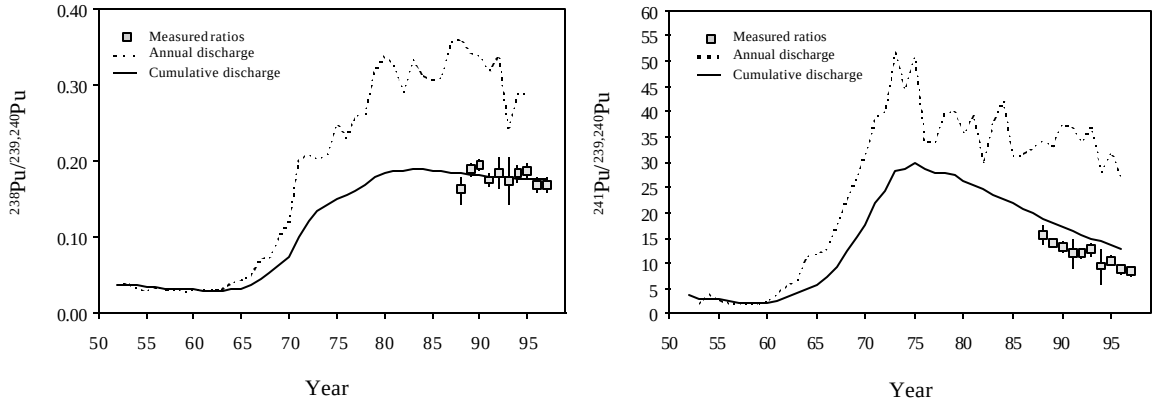

Figure 11. Measured $^{238}Pu/^{239,240}Pu$ and $^{241}Pu/^{239,240}Pu$ activity ratios ($\pm 2\sigma$) in surficial sediments in the western Irish Sea (averaged over three stations) compared with ratios in annual and cumulative discharges (*from:* Mitchell *et al.*, 1999)

The $^{238}Pu/^{239,240}Pu$ activity ratio in filtered sea water and suspended particulate in the western Irish Sea were found to be 0.199 ± 0.015 (n = 12) and 0.18 ± 0.03 (n = 12), respectively (Mitchell *et al.*, 1999). These ratios clearly indicate that the plutonium in the water column at the present time is mainly sourced from sediments rather than from contemporary discharges in which the ratio is ≈0.30 or higher. As suggested above, dispersion of plutonium re-worked from a mixed pool of contaminated sediment, either by resolubilisation or resuspension, appears to be the most likely mechanism controlling plutonium concentrations in the western Irish Sea. Within the limits of analytical uncertainty, it is possible, on the basis of the measured ratios, to make a rough estimate of the contribution of eastern and western Irish Sea sediments to present sea water concentrations. Taking the western and eastern Irish Sea sediment pools (top 10 cm approximately) to have $^{238}Pu/^{239,240}Pu$ ratios of 0.174 and 0.212, respectively, the measured mean ratios for the dissolved and suspended particulate fractions given above would suggest that most of the dissolved plutonium and comparatively little of the particulate plutonium in the western Irish Sea is sourced directly from eastern Irish Sea sediments. In other words, most of the particulate plutonium in western Irish Sea waters has its origins in the resuspension of local bed sediment. That most of the plutonium sourced from the eastern Irish Sea is transferred to the western Irish Sea in a soluble form after resolubilisation from seabed sediments is compatible with the observations of other workers, who have suggested that particle transport dominates in the north-eastern Irish Sea but not further afield (Cook *et al.*, 1997)

4.4. Biota

From the beginning of operations at Sellafield, samples of marine biological materials have been assayed for their radionuclide content as part of monitoring and surveillance programmes. The temporal evolution of ^{106}Ru, ^{137}Cs, Pu–α and ^{241}Am concentrations in *Porphyra* seaweed, fish (cod and plaice), lobster, crab, winkles and mussels sampled in the Cumbrian coast, as compiled and tabulated by Jackson *et al.* (2000) are presented in Figures

12 to 17 (original data taken from Bratt and Banbury, 1952; Dunster, 1952; 1956; 1958; Wix *et al.*, 1960; 1965; Pentreath and Lovett, 1976; Hamilton and Clifton, 1980; Myers, 1980a,b; Kershaw *et al.*, 1992; McDonald *et al.*, 1991, 1993; MAFF, 1967-99; BNFL 1978-99). It is clear from these data that, in general, radionuclide concentrations in seaweed, fish and shellfish sampled in the NE Irish Sea peaked in the early- to mid-1970s, declining thereafter. The reductions in the levels of major fission and activation products have been greater than those of the transuranium nuclides due to the apparently longer lag period between the decrease in transuranic discharges and its reflection in environmental materials (Hunt, 1985).

Concentrations in both fish and shellfish diminish with distance from Sellafield, the rate of reduction being least for nuclides which are relatively mobile in seawater. A similar pattern is observed for algae. Variations in concentrations between fish species sampled in a given area are comparatively small and can be explained in terms of residence time in the area as well as feeding habits (MAFF, 1967-99). On the other hand, substantial variations are observed between shellfish species. For example, molluscs tend to concentrate the transuranics to a considerably greater extent than do crustaceans, which in turn accumulate them more than fish. At the present time, typical fresh weight concentrations of ^{137}Cs in white fish, lobster, crab, winkles and mussels sampled in the Sellafield coastal area are 7 Bq kg^{-1}, 3.4 Bq kg^{-1}, 2.9 Bq kg^{-1}, 12 Bq kg^{-1} and 4 Bq kg^{-1}, respectively, while the corresponding Pu–α concentrations are 0.02 Bq kg^{-1}, 0.54 Bq kg^{-1}, 0.54 Bq kg^{-1}, 13 Bq kg^{-1} and 11 Bq kg^{-1}, respectively (Jackson *et al.*, 2000).

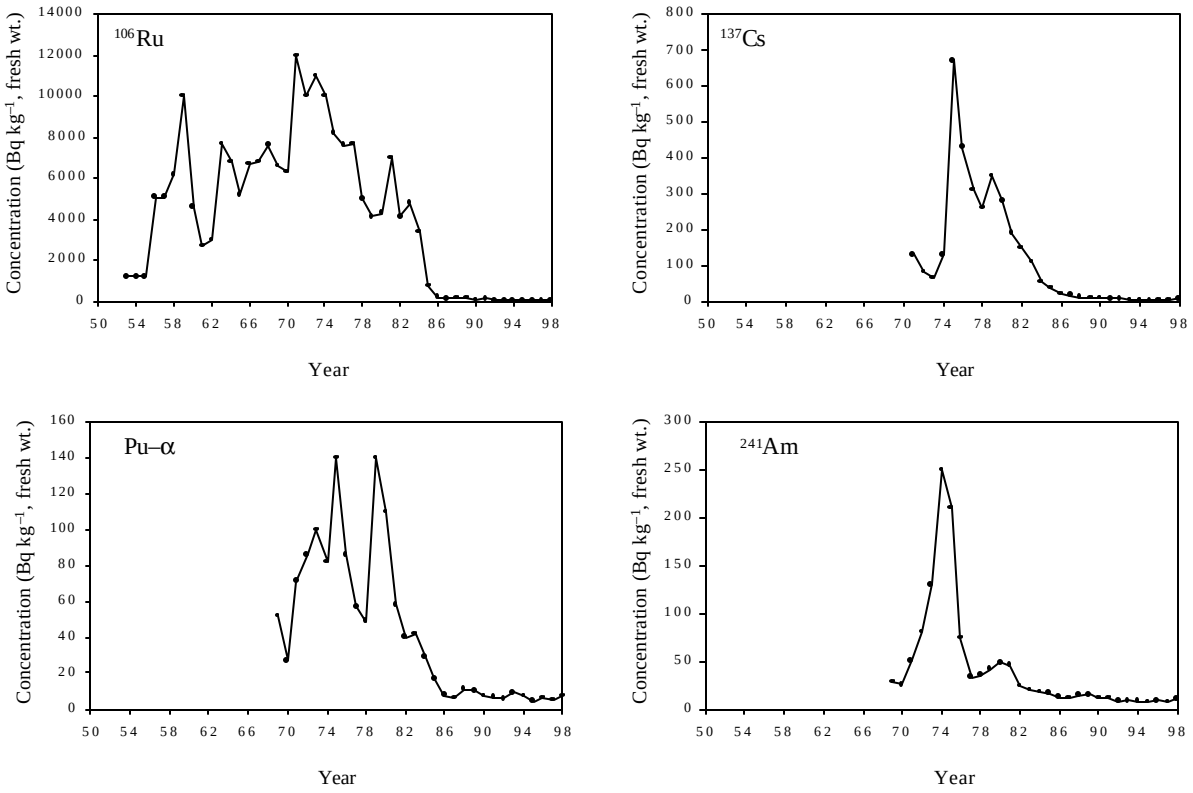

Figure 12. Concentrations of ^{106}Ru, ^{137}Cs, Pu–α and ^{241}Am in *Porphyra* seaweed in the Cumbrian coast

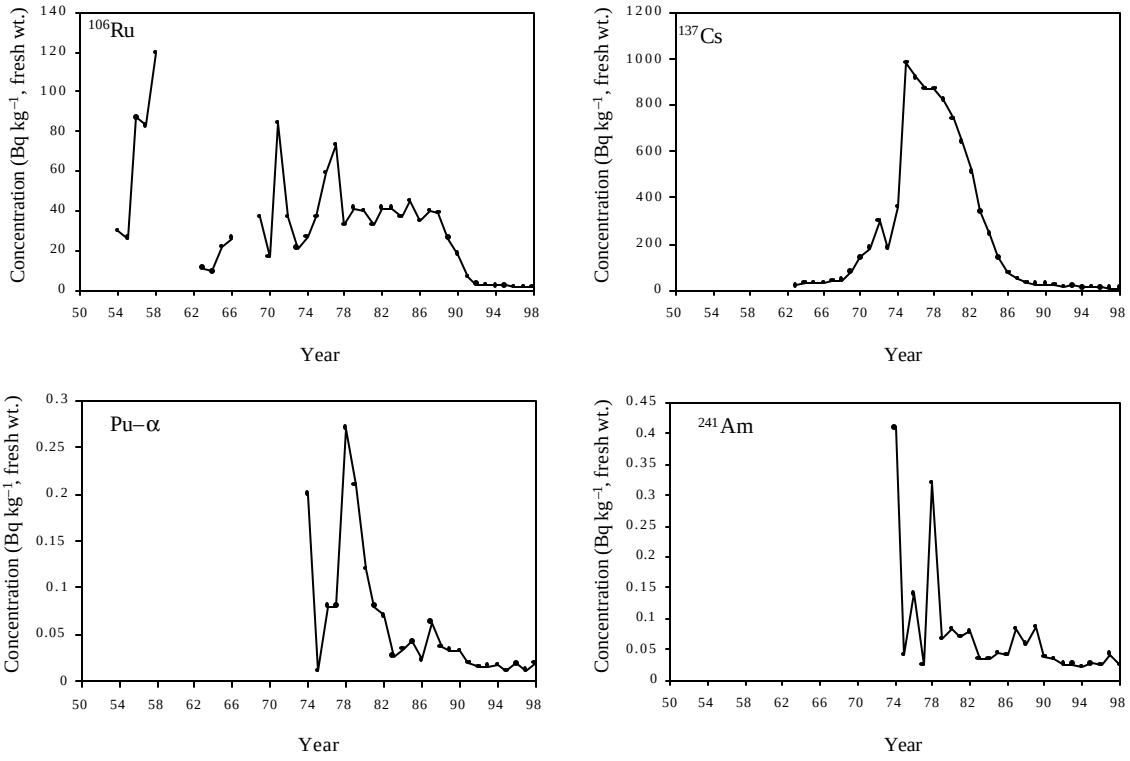

Figure 13. Concentrations of ^{106}Ru, ^{137}Cs, Pu–α and ^{241}Am in Cumbrian coastal fish (cod and plaice)

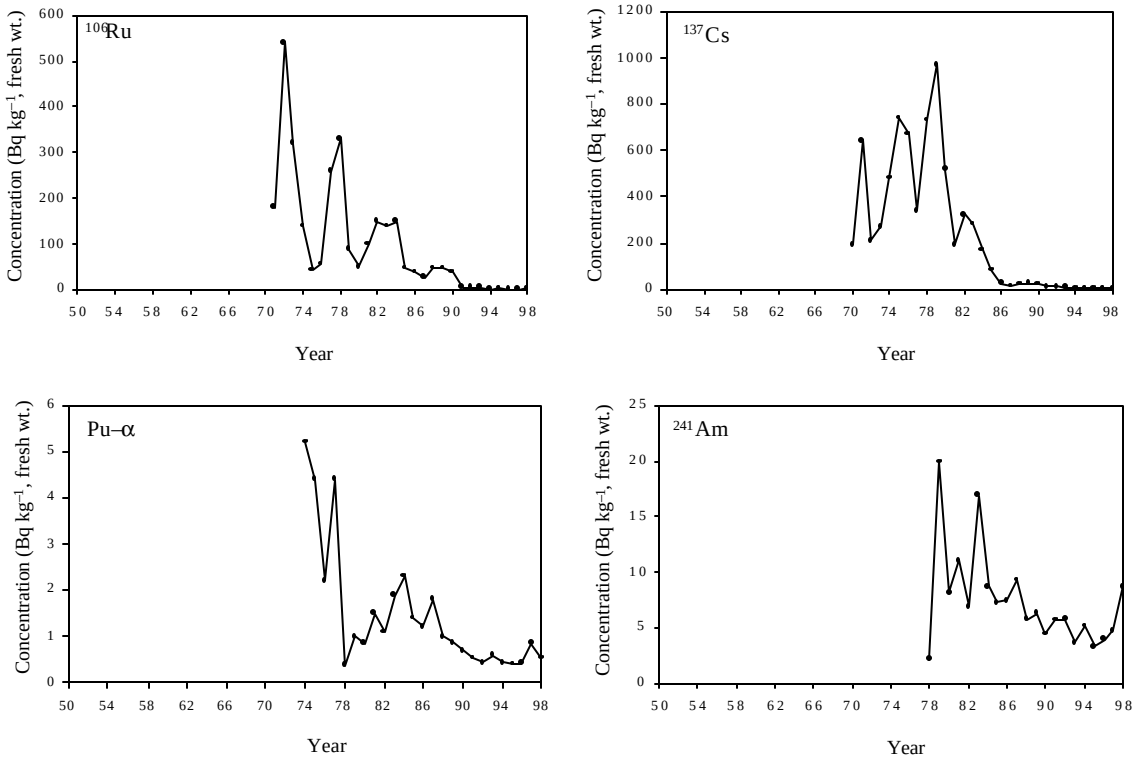

Figure 14. Concentrations of ^{106}Ru, ^{137}Cs, Pu–α and ^{241}Am in Cumbrian coastal lobster

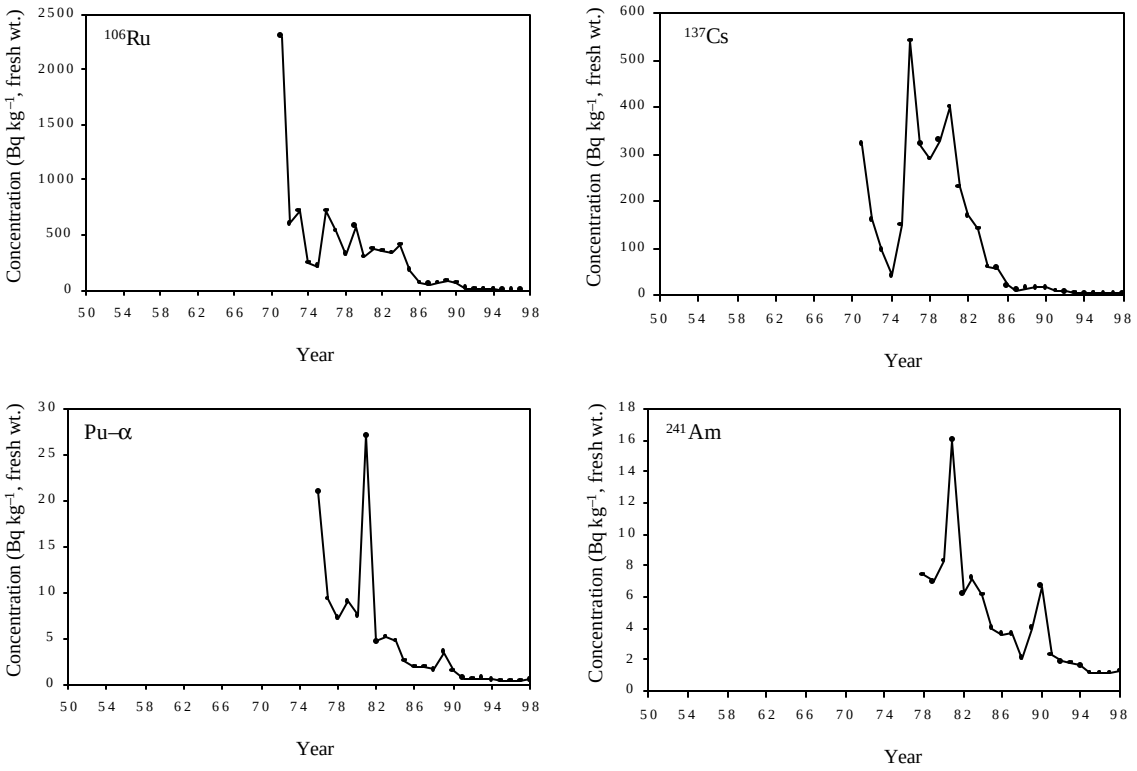

Figure 15. Concentrations of ^{106}Ru, ^{137}Cs, Pu–α and ^{241}Am in Cumbrian coastal crab

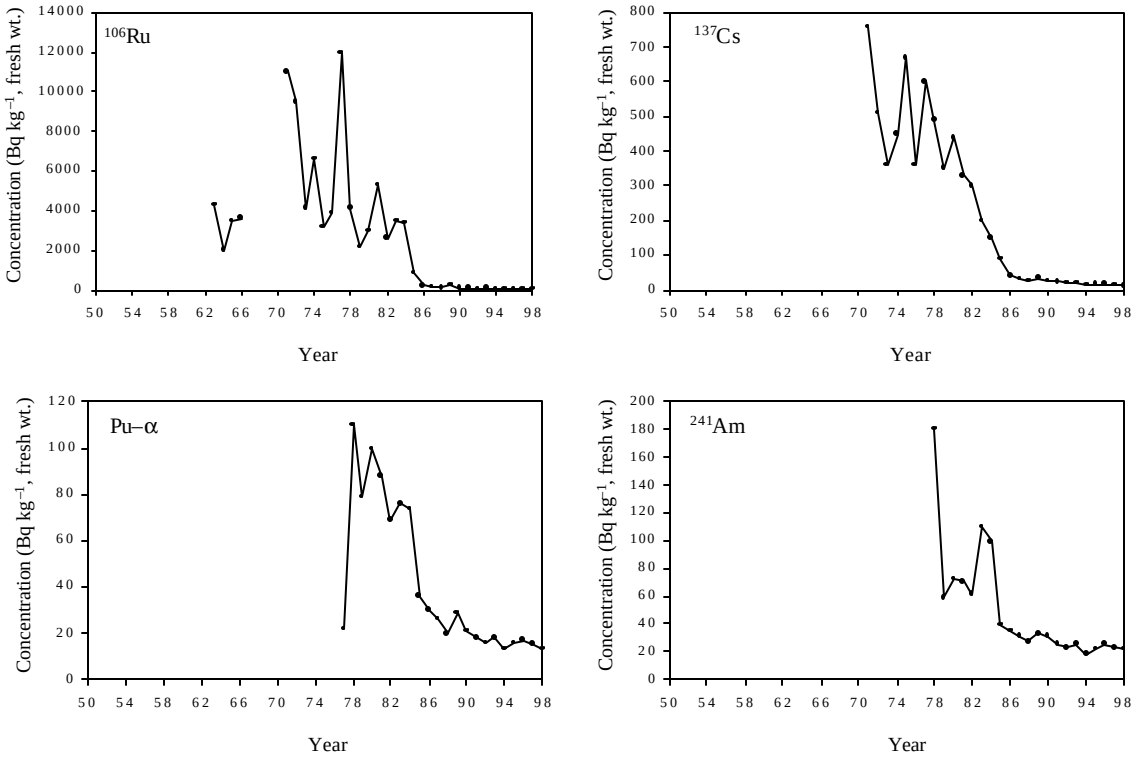

Figure 16. Concentrations of ^{106}Ru, ^{137}Cs, Pu–α and ^{241}Am in Cumbrian coastal winkles

Surveys carried out in the period 1979-95 have shown that radiocaesium concentrations in the main species of fish and shellfish taken mainly in the western reaches of the Irish Sea and landed at Irish east coast ports have declined from a mean of approximately 70 Bq kg^{-1} (fresh wt.) in 1979 to a mean of less than 2 Bq kg^{-1} (fresh wt.) by 1995. A somewhat smaller decrease has been observed for ^{239,240}Pu, with concentrations persisting at levels similar to those prevailing at the beginning of the 1990s (Ryan *et al.*, 1999).

Availability times in biota are generally less than those for sediments, reflecting the relative contributions of recent and historic discharges to present activity concentrations (Nicholson and Hunt, 1995). Further reductions in transuranium and other radionuclide concentrations in biota may be anticipated in the coming years, particularly in the north-eastern Irish Sea.

5. Radionuclide inventories in the Irish Sea

Attempts have been made to quantify the total environmental inventory of caesium and transuranic nuclides in the water column and sediments of the Irish Sea. Poole *et al*. (1997) presented estimates of ^{137}Cs inventories in the subtidal sediments of the Cumbrian coast, the eastern Irish Sea and the whole of the Irish Sea for 1988 and 1995. The distribution of ^{137}Cs (kBq m^{-2}) for both years are shown in Figure 17. A summary of the data is presented in Table 1. The total estimated inventory of ^{137}Cs in the subtidal sediments of the whole of the Irish Sea in November 1988 was 1532 TBq, representing ≈5% of the corresponding cumulative, decay-corrected, ^{137}Cs discharge. The estimated 1995 inventories of ^{137}Cs in the whole of the Irish Sea, the eastern Irish Sea, and the Cumbrian coastal area were 73%, 69% and 72%, respectivelym of the 1988 inventories decay-corrected to 1995. The similarity of the proportion in all three areas indicates that the reduction was consistent across the whole of the Irish Sea. The reduction in the estimated ^{137}Cs inventory in the sediments of the Irish Sea between 1988 and 1995 was 573 TBq, of which 223 TBq can be attributed to decay. Since decay occurs both in the sediments and after release of ^{137}Cs to the water column, the actual release to the water column over the 6.5 year period must have been between 350–573 TBq, representing 54–88 TBq y^{-1}. Since the integrated Sellafield discharge over the same period was 126 TBq, the sediment inventory from historic discharges was the dominant source to the water column, contributing between 74% and 82% of the total input (Poole, 1997).

Table 1. Cumulative ^{137}Cs discharge data and subtidal sediment inventory data for the whole of the Irish Sea, the Cumbrian coast and the eastern Irish Sea (*from:* Poole *et al.*, 1997)

Year	^{137}Cs inventory (TBq)			Annual	% of total discharge		
	All Irish Sea	E Irish Sea	Cumbrian coast	discharge (TBq)	All Irish Sea	E Irish Sea	Cumbrian coast
1988	1532	1098	420	308976	5.0	3.5	1.4
1988*	1309	938	359				
1995	959	648	258	26469	3.6	2.5	1.0

*data decay-corrected to 1995

Similar calculations for ^{239,240}Pu and ^{241}Am inventories in subtidal sediments for 1978, 1983, 1988 and 1995 have also been reported by Kershaw *et al.* (1999), and are summarised in Table 2. The data clearly indicate that subtidal sediments of the Irish Sea remain the single

most important repository of plutonium and americium discharged from Sellafield. The subtidal radionuclide distribution has changed, with a progressive decrease in both surface concentrations and inventories near the Cumbrian coast and increases at more distant sites (Kershaw *et al.*, 1999). Efforts have also been made to provide a budget for these radionuclides in intertidal sediments and the water column. Using a number of sources (Eakins *et al.*, 1988; 1990; Garland *et al.*, 1989; Toole, 1992; Kershaw, 1997; Jones *et al.*, 1998; 1999) Kershaw *et al.* (1999) have arrived to a figure of ≈70 TBq for the inventories of plutonium and americium in the intertidal sediments of the Irish Sea. For the water column, an inventory of ≈56 TBq is quoted for Pu–α. There remains a significant shortfall between the estimated inventories and the reported, decay-corrected discharge. Although part of this can be accounted for by the total loss through the North Channel, it is likely that a significant proportion of the unaccounted fraction is situated in subtidal and intertidal sandy deposits below the sampling depth achieved in most published studies (Kershaw *et al.*, 1999).

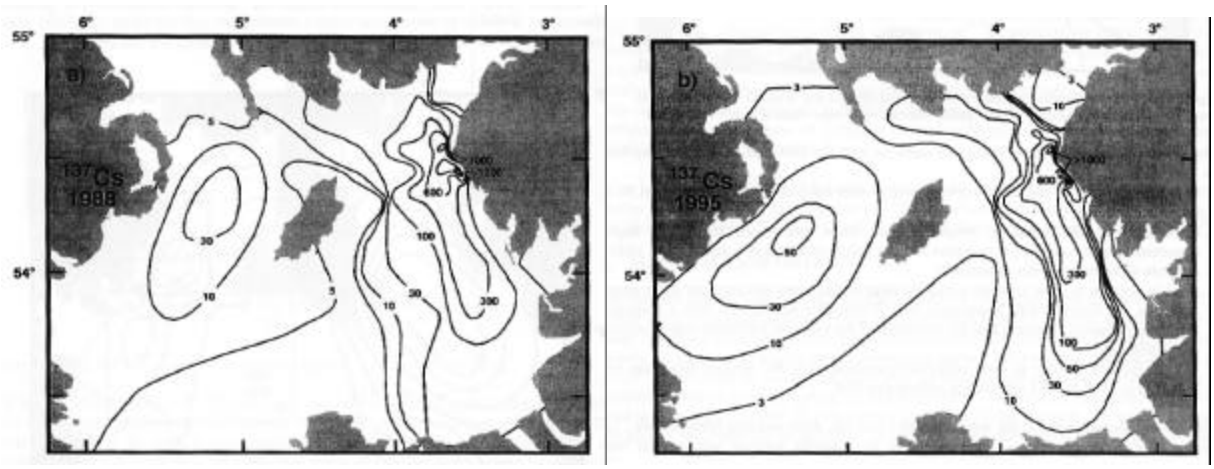

Figure 17. ^{137}Cs inventory (kBq m^{-2}) in the subtidal sediments of the Irish Sea in (a) 1988 and (b) 1995 (*from:* Poole *et al.*, 1997)

Table 2. Estimated inventories of ^{239,240}Pu and ^{241}Am in subtidal sediments of the Irish Sea, expressed as total activities (TBq) and as percentages of the total, decay-corrected inputs to the environment (from: Kershaw *et al.*, 1999)

	1978		1983		1988		1995	
	TBq	**%**	**TBq**	**%**	**TBq**	**%**	**TBq**	**%**
^{239,240}Pu								
All Irish Sea	248	52.3	n/a	n/a	341	58.1	360	60.8
E Irish Sea	196	41.3	236	41.2	286	48.7	284	47.9
Cumbrian coast	115	24.2	146	25.5	172	29.3	151	25.5
^{241}Am								
All Irish Sea	308	49.9	n/a	n/a	487	56.7	545	56.7
E Irish Sea	248	40.2	275	36.2	415	48.3	439	45.6
Cumbrian coast	144	23.3	163	21.5	250	29.1	242	25.2

6. Radiological implications of the Sellafield discharge

The assessment of the impact of discharges from Sellafield have always taken the irradiation of the local marine fauna and flora into consideration. Numerous field and laboratory studies, reviewed by Kershaw *et al.* (1992), have concluded that during the period of maximum discharges, dose rates around Sellafield were 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 might be expected to have an effect at the population level (Woodhead, 1980). As evidence from the above discussion, were the Sellafield discharges to remain at their present low level, dose rates are likely to decline significantly as a result of the dispersion and mixing of water and sediments.

A comprehensive review on doses to local human critical groups in Cumbria, based on the best available monitoring data, supplemented by modelled values where necessary, has recently been undertaken by Jackson *et al.* (2000). During the 1950s and 1960s, the highest doses were received by individuals consuming Cumbrian *Porphyra* and laverbread, with peak doses around 0.8 to 1.0 mSv y^{-1}. During the 1970s and 1980s the critical exposure group switched to consumers of local fish and shellfish, with peak doses possibly reaching 2.5 to 3.0 mSv y^{-1}. In recent years, doses to all critical groups have declined to less than 0.15 to 0.2 mSv y^{-1}. Thus, doses have at all times been within the appropriate limits set for members of the public.

Further afield, doses are even lower. To illustrate, the committed effective dose due to radiocaesium to typical and heavy consumers of fish and shellfish landed at north-eastern Irish ports for the years 1982 to 1999 are given in Figure 18 (Long *et al.*, 1998a, b; Ryan *et al*., 2000). The trend in the dose clearly reflects the pattern of ^{137}Cs levels in sea water and sediments of the western Irish Sea over the same period. The dose due to radiocaesium constitutes the majority of the dose arising from artificial radionuclides. For example, in 1995, the dose to a heavy consumer of fish and shellfish landed at north-eastern Irish ports due to radiocaesium was estimated to be 1.7 μSv y^{-1}, while that attributable to the actinides was only 0.3 μSv y^{-1} (Long *et al.*, 1998b). The total dose is less than 1% of the ICRP reccommended limit of 1000 μSv for members of the public, and is very small compared with the annual average dose of ≈3000 μSv received by Irish members of the public from all sources of radiation. These figures may also be put into perspective by comparing them with the annual dose arising from the naturally-occurring ^{210}Po, estimated to be 148 μSv y^{-1} (Pollard *et al.*, 1996).

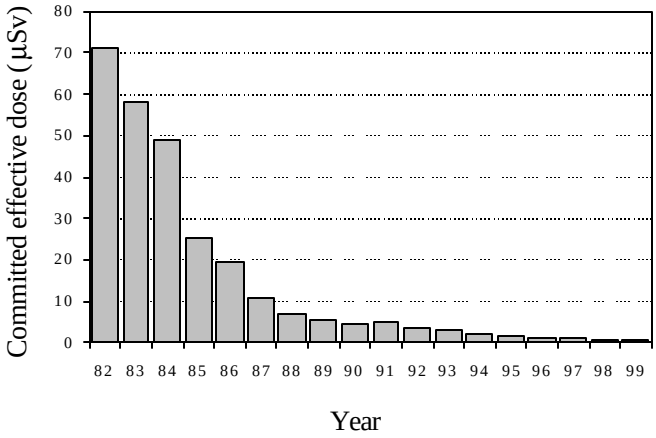

Figure 18. Committed effective dose due to radiocaesium in fish and shellfish landed at Irish north-east ports, 1982-99 (source: Long *et al.*, 1998a, 1988b; Ryan *et al.*, 2000)

7. Concluding remarks

It should be clear from this overview that a considerable body of knowledge has been accumulated over the years on the behaviour of radionuclides discharged from Sellafield, the transfer processes responsible for their redistribution and their environmental impact.

Numerous studies undertaken over the past three decades have shown conclusively that ^{137}Cs concentrations and inventories in the Irish Sea have already peaked and are now in steady decline. This reduction appears to be taking place in all of the Irish Sea, and there is unambiguous evidence that ^{137}Cs is being remobilised from the sediments at a rate of at least 50–90 TBq y^{-1}. Since present direct ^{137}Cs discharges from Sellafield are at a level of ≈8 TBq y^{-1}, the sediment inventory from historic discharges is now the dominant source to the water column. Much of the ^{137}Cs activity discharged from Sellafield has now left the Irish Sea, and has resulted in a significant increase in the ^{137}Cs inventory in the North Atlantic.

In the case of the transuranic nuclides, it is clear that subtidal sediments of the Irish Sea remain the single most important repository of plutonium and americium discharged from the Sellafield site, with most of the inventories contained in the eastern Irish Sea. Although little change has been observed in total inventories for these nuclides, comparison of past and recent surveys of radionuclide distributions indicate a progressive decrease in both surface concentrations and inventories near the Cumbrian coast and increases at more distant sites. This is supported by time-series data on the western Irish Sea, which confirm that transuranic concentrations in this zone are being driven by the rate at which these nuclides are being remobilised in the eastern Irish Sea and transferred to the western Irish Sea. Measurement of transuranium activity ratios in the western Irish Sea mud basin, as well as in filtered and suspended particulate sampled in east-west transects from Sellafield indicate that desorption of these nuclides from the pool of well-mixed sediments in the western Irish Sea, followed by transfer, primarily in soluble form, to the western Irish Sea and beyond are the dominant mechanisms involved. As for ^{137}Cs, following the considerable reduction in the rate of discharge, subtidal sediments now represent the biggest source of actinides to the Irish Sea. The rate of dissolution of plutonium and americium appears, however, to be very slow, and it is clear that environmental concentrations of these actinide radionuclides will persist in the Irish Sea environment for a considerable time. At the same time, the water flowing out of the Irish Sea is likely to remain one of the major sources of transuranium nuclides to the shelf seas of northwest Europe and beyond.

The importance of radionuclide remobilisation in the assessment of the environmental impact of radioactive discharges into the marine environment and the realisation that sediments can act not just a sink but also a potential source of contaminants to the overlying waters has prompted an intensification of interest on the study of the processes controlling remobilisation and transfer of radionuclides from historically contaminated sediments. A new multinational research programme in this area, funded by the European Commission, and involving twelve European institutions working in the field of marine radioecology, has now just started. The programme, entitled *Processes Regulating Remobilisation, Bioavailability and Translocation in Marine Sediments (REMOTRANS)*, will address, among others, questions such as: • To what extent does the translocation of remobilised radionuclides differ from that of direct discharges?; • What are the chemical and physico-chemical forms of remobilised radionuclides ?; • What is the impact of radionuclide remobilisation from aquatic sediments on bioavailability?; and • What is the best way to incorporate remobilisation processes in realistic predictive models for long-term dose assessment?

The Irish Sea is one of the principal sites selected for the study, and the answer to the question cited above should help to gain a deeper understanding on the long-term impact of the Sellafield discharges on the marine environment.

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