

ANTHROPOGENIC ^{14}C MARINE GEOCHEMISTRY IN THE VICINITY OF A NUCLEAR FUEL REPROCESSING PLANT

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ABSTRACT. The British Nuclear Fuels plc reprocessing plant at Sellafield in Cumbria, Northwest England, discharges between 1 and 3 TBq of ^{14}C per annum as low-level waste via a pipeline into the Eastern Irish Sea. Our results demonstrate ^{14}C activities in excess of the current ambient level (*i.e.*, *ca.* 260 Bq kg⁻¹ carbon) in a range of both biotic and abiotic samples with evidence that a significant percentage of the discharges are in the form of dissolved inorganic carbon (DIC). There also appears to be a significant seasonal influence on the uptake of the DIC by the phytoplankton, most likely linked to their bloom periods. Results from different aspects of the research are integrated to provide an opportunity to consider the overall distribution and behavior of ^{14}C in the Eastern Irish Sea. We also discuss the results in terms of existing sediment chronologies and sedimentation rates and consider the possible consequences of phytoplankton blooms on the cycling of the other radionuclides.

INTRODUCTION

The authorized discharges into the Irish Sea of low-level liquid radioactive waste from the British Nuclear Fuels plc reprocessing facility at Sellafield in Cumbria, Northwest England, have been the subject of intensive study over the past few decades (Hetherington 1975; Baxter *et al.* 1979; Kershaw *et al.* 1983; Hunt 1985; McDonald *et al.* 1990; MacKenzie *et al.* 1994). This research has focused largely upon ^{238}Pu , $^{239,240}\text{Pu}$, ^{241}Am , ^{134}Cs and ^{137}Cs , which are among the major components of the discharges (BNFL 1977–1989; Cambray 1982). From these published data and radioactive decay and ingrowth calculations, one can determine that, by 1990, Sellafield had generated total environmental inventories of *ca.* 3×10^4 TBq of ^{137}Cs , 6.8×10^2 TBq of Pu- α and 8.9×10^2 TBq of ^{241}Am . Researchers have estimated that nearly all of the ^{241}Am , most of the Pu and *ca.* 10% of the radiocesium is retained within the sediments of the Irish Sea, in particular, in a near-shore deposit of mud and fine silt close to the discharge point (Hetherington 1975; Day and Cross 1981; Aston and Stanners 1982; Pentreath *et al.* 1984; Pentreath, Harvey and Lovett 1986). Kershaw (1986) used radiocarbon dating to examine two cores from this area of fine sediment and found near-constant ages ($12,500 \pm 1000$ BP) from the sediment surface to the base of the cores (*ca.* 150 cm). Later research (Kershaw, Swift and Denoon 1988) demonstrated rather irregular chronologies for the bulk sediment of several other cores (10,300–15,300 BP), but in contrast, what appear to be logical chronologies and much younger ages derived from dating the shells of the gastropod *Turritella communis* (Risso). Sedimentation rates based on the *T. communis* chronologies were 0.02–0.08 cm a⁻¹ over the past 3000–4000 yr at two sites, and a more variable and rapid rate of *ca.* 0.3 cm a⁻¹ at a third. During the period of high discharges in the 1970s, this area of sediment acted as a net sink for radionuclides. Because the present discharges, however, are currently around two orders of magnitude lower than during this period, the fine sedimentary material now constitutes a potential source of radionuclides to the wider environment. Radionuclide transport is either by physical dispersion of the contaminated particles or by redissolution of the radionuclides upon contact with seawater, which is now much less contaminated.

In contrast to the radionuclides discussed above, the annual marine ^{14}C discharges have been in the range of 1 to 3 TBq since monitoring began in 1984, and the estimated total environmental inventory of ^{14}C discharged to the Irish Sea is <50 TBq. Little information is available on the form or fate of the ^{14}C ; despite the low levels, ^{14}C contributes the highest estimated collective dose commitment to

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the UK population from the Sellafield marine discharges. This is due to its long half-life ($t_{1/2} = 5730$ yr) and high environmental mobility, although the dose is a small percentage of that derived from natural sources.

The information presented here is important for modeling the likely environmental impact of future discharges as well as for the chemical behavior and ultimate fate of carbon within a general near-shore environment. Previous studies (McCartney, Baxter and Scott 1988a,b) at this laboratory considered the local and global implications of releases to the atmospheric environment. We present here a study of the geochemistry and spatial and environmental distributions of the marine component of anthropogenic ^{14}C . Begg *et al.* (1992) considered the ^{14}C activity in a range of biological materials collected from the intertidal regions to the north and south of the discharge point. They selected the sample types on the basis of availability and carbon uptake mechanisms, as these could give a guide to the chemical form of the ^{14}C discharges. The sample types selected were: 1) mussels (*Mytilus edulis*)—mytilids are filter feeders on particulate organic material from the water column and, together with other filter-feeders, they represent a widespread and direct trophic link between the benthic and planktonic communities; 2) winkles (*Littorina littorea*)—winkles graze particulate material deposited on large seaweeds and algae-covered rocks, which could include phytoplankton, fecal deposits produced by mytilids and zooplankton; 3) seaweed (*Fucus* sp.)—during photosynthesis, seaweeds can directly utilize the dissolved inorganic carbon (DIC) fraction of the total carbon pool contained in seawater.

These studies indicated that ^{14}C concentrations depend on both the species of biota (and therefore on their feeding mechanism) and the distance from the point of discharge. In the immediate vicinity of Sellafield, mussels had enrichments of up to 7 times the current ambient ^{14}C level whereas seaweed had enhancements of *ca.* 3 times this activity. Winkles obtain carbon from the particulate phase and indirectly from the DIC by grazing on algae. This is reflected in a maximum activity of about five times the ambient level, intermediate between the mussel and seaweed activities. Thus, these results indicate the presence of ^{14}C -enriched material in both the particulate and dissolved phases.

To determine whether this was indeed the case, our attention turned to the water column and the analysis of the individual biogeochemical phases, *i.e.*, DIC and dissolved organic carbon (DOC) and particulate inorganic (PIC) and organic carbon (POC) fractions. Samples were collected during December 1989, February 1991 and June 1993. The following samples were also obtained: sediment cores were collected during December 1989 and November 1990; mixed plankton samples were collected during summer 1989; demersal (bottom feeding) fish species were collected during November 1989.

METHODS

^{14}C analyses were performed at Scottish Universities Research & Reactor Centre (SURRC) using liquid scintillation spectrometry (LSS) with the exception of small samples (<1 g carbon), which were sent as CO_2 for accelerator mass spectrometry (AMS) measurement at the University of Arizona, NSF-Arizona AMS Facility and Oxford University, Oxford Radiocarbon Accelerator Unit.

Sample Collection—Biota

Phytoplankton are the primary producers in the marine environment, deriving carbon for photosynthesis from the DIC fraction of seawater. They constitute, by far, the main source from which higher trophic levels derive energy. Mixed phytoplankton/zooplankton samples were collected in 1989 from the Northeast Irish Sea (Fig. 1) and stored in formalin, which had to be removed prior to ^{14}C

analysis. To accomplish this, the bulk of the formalin was removed by filtration and the plankton were soaked in distilled water for several days, with numerous changes of water. Because some formalin could remain associated with the plankton, the ^{14}C activities represent a minimum.

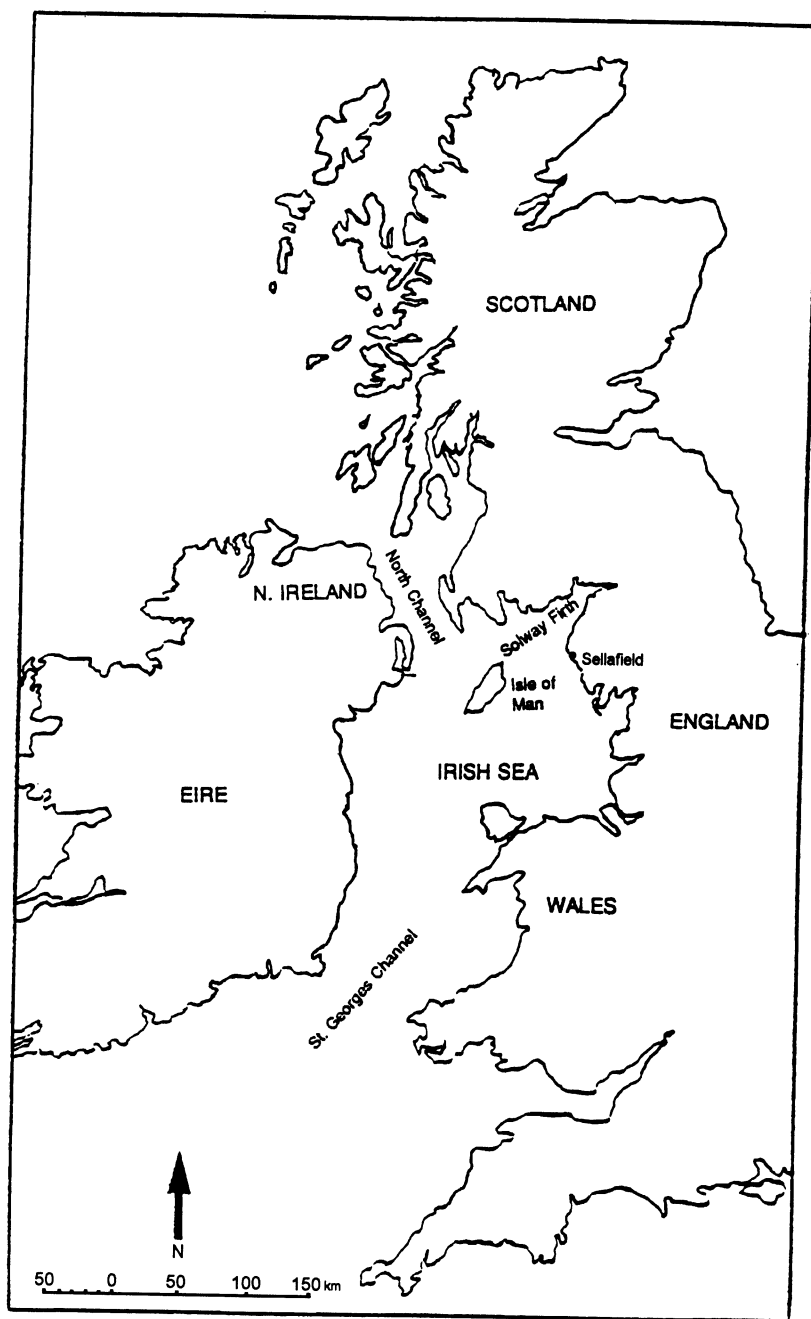


Fig. 1. Map showing the location of Sellafield within the Irish Sea basin and the proximity to the North Channel and St. Georges Channel

Plaice and dab, both demersal fish species that derive energy by feeding in the surface layers of sediments, were collected in November 1989 from an area *ca.* 2 km south of the Sellafield discharge pipe. These were filleted, partly cooked using a microwave oven and dried at 60°C.

Sample Collection—Abiota

We collected water samples (200 liters) from five sites in the Irish Sea and isolated four biogeochemical fractions: POC, PIC, DIC, DOC. Of these sites, 3 were sampled in December 1989—1 in St. Georges Channel and 1 in the North Channel—to provide an indication of the ^{14}C specific activity in the biogeochemical fractions as the water enters and exits the area, respectively (Fig. 1). The third site was just off the Sellafield pipeline, close to the point of release. We resampled this site in February 1991 and again in June 1993 for comparison with the trends observed in the initial results. Two other sites were sampled in February 1991—one just off the North Solway coast and the second just to the north of the Isle of Man. We sampled these two sites to learn about the rate at which the ^{14}C specific activity in the various fractions might change within the immediate zone of ^{14}C contamination from Sellafield.

In December 1989, we collected a sediment sample using a Kaston corer from the “mud patch” close to the end of the Sellafield pipeline to determine if, and in what form, Sellafield-derived ^{14}C was being incorporated into the bottom sediments. This was subsequently subsampled using a plastic tube (8 cm in diameter) to 25-cm depth and divided into 5-cm sections for analysis of both the organic and inorganic ^{14}C activities. We collected a gravity core in November 1990 from the “mud-patch” *ca.* 20 km south of Sellafield and again, we determined ^{14}C in both the organic and inorganic fractions from a range of depths.

Sample Analysis Procedures

Of the fractions isolated from the water column, only the DIC is present in sufficient concentration to allow LSS analysis of ^{14}C . DOC, POC and PIC analysis must be carried out by AMS. To obtain sufficient carbon from the DIC fraction for LSS analysis, we collected 200 liters of surface water into previously acid-washed barrels. We then filtered the water through precombusted glass fiber filters (Whatman GFA) and acidified and purged it with CO_2 -free nitrogen. We then bubbled the resultant CO_2 through 4M NaOH (carbonate-free) for 8 h in a closed system. To obtain sufficient DOC for ^{14}C analysis, we collected 25 liter water samples. The preconcentration of DOC from the surface water was carried out using macroporous non-ionic XAD resins (XAD-2, XAD-4 and XAD-8), which have been used extensively for extracting trace amounts of organic compounds from natural and waste waters (Leenheer 1981), groundwaters and seawater (Stuermer and Harvey 1977). Begg (1992) and Begg *et al.* (1991) give a full account of this procedure.

Filter papers containing the particulate material were washed with distilled water (~2 liters) to remove any salts that may have presented problems during wet oxidation of the POC. Prior to oxidation of the POC, the PIC was hydrolyzed with 50% H_3PO_4 . The residue then underwent oxidation with chromic acid/excess potassium dichromate to yield CO_2 from the POC material. All sediment samples, bottom and intertidal, were slurried in distilled water to remove excess salts, filtered and dried prior to analysis. The sedimentary inorganic carbon was then hydrolyzed using 2 M HCl before the organic material was wet-oxidized.

For those samples analyzed by conventional radiometric techniques, the CO_2 produced by either sample combustion (biota) or hydrolysis (DIC and sediments) was collected and converted to ben-

zene for LS counting *via* lithium carbide and acetylene production. The remaining samples (POC, PIC, DOC, some DIC and sediments) were converted to CO_2 and sent for AMS analysis.

RESULTS AND DISCUSSION

We report our results according to Stuiver and Polach (1977) as percent modern (pM) $\pm 1 \sigma$, corrected for fractionation effects and decay of the modern reference standard (SRM-4990C, NIST HOxII) or as conventional ^{14}C ages (BP) $\pm 1 \sigma$. Table 1 gives results for the biogeochemical fractions of the water column. The December 1989 results indicate that all the fractions are depleted relative to the ambient activity ($\sim 115.4 \text{ pM}$ in the UK coastal marine environment at the time of sampling (Begg 1992)) with the exception of the DIC fractions. The St. Georges Channel sample is slightly enriched because Amersham International plc discharges ^{14}C into Cardiff Bay to the south of this sampling site.

TABLE 1. ^{14}C Activities in the Four Geochemical Fractions of Irish Sea Water Samples*

Location	Fraction			
	DIC	PIC	DOC	POC
<i>December 1989</i>				
Entrance (St. Georges Channel)	119.8 ± 0.8	55.0 ± 0.5	82.7 ± 0.9	51.9 ± 0.6
Sellafield	243.3 ± 1.7	47.9 ± 0.5	63.4 ± 0.8	73.1 ± 0.7
Exit (North Channel)	135.9 ± 0.3	63.8 ± 0.6	74.0 ± 0.8	57.5 ± 0.5
<i>February 1991</i>				
Sellafield	200.0 ± 0.6	59.1 ± 1.2	38.5 ± 0.7	80.4 ± 1.1
Solway Coast	183.9 ± 0.7	100.3 ± 1.5	33.2 ± 0.5	66.6 ± 0.7
North of Isle of Man	168.9 ± 0.7	51.4 ± 1.0	30.2 ± 0.5	59.4 ± 1.0
<i>June 1993</i>				
Sellafield	187.5 ± 1.0	67.0 ± 0.5	149.0 ± 0.8	188.3 ± 1.0

*All results reported as $\text{pM} \pm 1 \sigma$

These results for the biogeochemical fractions were very surprising because the highest activities observed in the intertidal biota samples were from mussels, which are exclusively filter feeders. Thus, we expected to see an enrichment of the POC and perhaps of the DOC fractions. The inconclusive nature of the results obtained for these samples led to us collecting a second suite of samples in February 1991. These results (Table 1) completely substantiate the general trends observed in the 1989 samples. The results derived from the samples collected in June 1993 (Table 1) are quite different from those of the previous two sampling periods, and imply that the activities in the biogeochemical fractions of the water column are seasonally dependent. Only the PIC fraction is depleted relative to the modern ambient activity. In addition, the POC and DIC fractions are not significantly different from one another; this could indicate that POC derives directly from DIC. This also indicates fast carbon turnover that can be expected of phytoplankton. However, because we did not carry out any microscopic examination of the POC, this is entirely speculative. Table 2 lists the results of the mixed plankton and demersal fish analyses. ^{14}C activities in the plankton are comparable with those for most DIC samples, both in this study and that of Begg *et al.* (1991), and appear to corroborate our contention that plankton are central to seasonal changes in the ^{14}C distribution.

The lack of evidence for ^{14}C enrichment of particles during the winter months could have implications for particle uptake and environmental transfer of the particle reactive radionuclides present in

the discharges. In particular, several isotopes of Pu and ^{241}Am are important radiological components of the Sellafield discharges. It is well recognized that $^{234}\text{Th}/^{238}\text{U}$ disequilibrium studies can be used to estimate primary production in the open ocean, because ^{234}Th is removed very effectively from the water column onto plankton (Coale and Bruland 1985; Buesseler *et al.* 1992) and inorganic particle fluxes in this environment are low. Similarly, ^{234}Th becomes sorbed onto mineral particles and the $^{234}\text{Th}/^{238}\text{U}$ disequilibrium can also be used to study nearshore scavenging rates (McKee, De Master and Nittrover 1984). It is also feasible that plankton are an additional source of particles for removal of Am and Pu from the water column, as both have high K_D values for this partitioning (Fisher, Olson and Bowen 1980; Fisher, Bjerregaard and Fowler 1983). Thus, the environmental transfer of particle-reactive radionuclides could differ between the summer and winter months.

TABLE 2. ^{14}C Activities in Plankton and Demersal Fish Collected from the Northeast Irish Sea

Sample type	Activity (pM \pm 1 σ)
Mixed plankton (Sellafield)	270.0 \pm 2.1
Mixed plankton (St Bees Head)	252.0 \pm 1.6
Fish—dab species (Sellafield)	502.0 \pm 1.2
Fish—plaice species (Sellafield)	398.0 \pm 1.2

The ^{14}C activities in demersal fish are approximately twice those of the DIC, whereas the highest activity measured in mussels (Begg *et al.* 1992) is approximately a factor of three greater than the DIC. If the main transfer mechanism through the food chain is from DIC to fish and shellfish (mussels) *via* plankton, then we might expect all the sample types to have similar activities. Both mussels and fish will integrate the activities associated with plankton (and other organic detritus such as fecal pellets) derived from several bloom periods. If such a period coincided with a high ^{14}C discharge, this would result in the highest activities in phytoplankton. An activity of 479.1 pM was measured in the DIC within 1 km of Sellafield (Begg *et al.* 1991); this anomaly requires further investigation.

The ^{14}C results of organic carbon analyses for Core 1 (Table 3) show ages varying from 1350 BP at the top of the core (0–4 cm) to 3155 BP at the base (32–36 cm); however, this chronology is irregular and shows age reversals, which might be explained by an upper mixed box of younger ages and a lower box of older material. In contrast, the inorganic carbon ages are much greater and comparatively uniform throughout. These results are similar to those of Kershaw (1986) and Kershaw, Swift and Denoon (1988) for their bulk carbonate analyses. Thus, a discrepancy of *ca.* 9500 yr exists between the ages of the inorganic and organic carbon fractions at the top of the core and one of 8500 yr at the base. From this, it is difficult to draw conclusions based on logical chronologies and calculations of sedimentation rates.

TABLE 3. Radiocarbon Measurements in Northeast Irish Sea Sediment Core 1

Depth (cm)	Inorganic C (age \pm 1 σ)	Organic C (age \pm 1 σ)
0–4	10,810 \pm 110	1350 \pm 55
8–12	11,550 \pm 140	1675 \pm 55
16–20	11,840 \pm 100	3640 \pm 60
24–28	12,650 \pm 100	3410 \pm 70
32–36	11,840 \pm 150	3155 \pm 60

The results for the inorganic carbon analyses of Core 2 (Table 4) indicate younger ages than in Core 1. Again, we can argue for a younger, surface mixed box (0–10 cm) and an older mixed box (15–25 cm) with an intermediate age at 10–15 cm. In this core evidence for ^{14}C enrichment of the organic fraction throughout the core (generally >200 pM) is clear, which again makes it difficult to invoke arguments for calculating sedimentation rates for this area. The core also provides unambiguous evidence of Sellafield-derived ^{14}C pollution to a minimum depth of 20–25 cm. Core 2 was collected just off the end of the discharge pipe and has consistently higher activities than Core 1, which was collected from *ca.* 20 km to the south. This could signify a local enrichment affecting both the inorganic and organic components. Kershaw, Swift and Denoon (1988) established their chronologies by dating the shells of *Turritella communis*, which yielded different ages from the surrounding bulk sediment. The authors argue that these shells are much less liable to biological mixing by the species of organisms inhabiting this sediment, and these chronologies are best estimates at present. However, if *T. communis* are still present in this area, which Jones (1952) and Pentreath and Jeffries (1971) dispute, they could be contaminated with Sellafield ^{14}C . The vertical mixing of such shells by physical means could give the effect of increasing age with depth and render the sedimentation rates invalid.

TABLE 4. Radiocarbon Measurements in Northeast Irish Sea Sediment Core 2

Depth (cm)	Inorganic C (pM $\pm 1\sigma$)	Inorganic C (age $\pm 1\sigma$)	Organic C (pM $\pm 1\sigma$)
0–5	53.8 \pm 0.7	4980 \pm 100	211.5 \pm 1.1
5–10	53.3 \pm 0.5	5060 \pm 80	192.2 \pm 1.3
10–15	36.8 \pm 0.4	8030 \pm 90	233.4 \pm 0.8
15–20	32.7 \pm 0.3	8980 \pm 70	256.1 \pm 1.5
20–25	32.7 \pm 0.4	8980 \pm 100	233.8 \pm 0.9

In their review of the geological structure of the Irish Sea, Smith, Parker and Kirby (1980) state that the area consists of several sedimentary basins, formed by subsidence due to faulting, which contain Carboniferous and Permo-Triassic rocks, overlain by sediments deposited since the Pleistocene (1.6×10^6 BP to 10^4 BP). During the glacial retreat, from *ca.* 12 ka BP, a series of proglacial lakes was formed, resulting in the deposition of highly stratified lacustrine sediments. Cronan (1969) postulates that the sediments throughout the Irish Sea are subject to wave and current action, and when combined with the shallow nature of the basin (20–40 m to the east of the Isle of Man), these processes play a prominent role in sediment redistribution. Our hypothesis for the observed trends in inorganic carbon data is that the ages are incurred by the mixing of old, reworked carbonate (>12 ka and quite possibly of infinite age) with biologically produced carbonate from the entire Holocene ($\delta^{13}\text{C}$ values were all typical of marine carbonate). The most recent production is enriched in ^{14}C due to inputs from nuclear weapons testing and Sellafield and Amersham (Cardiff) releases. Begg (1992) demonstrated significant ^{14}C enrichments in mussel and winkle shells from intertidal areas of the Irish Sea. The rate of redissolution of this inorganic material is slow, causing these apparently old ages because of the dominance of old material. The organic carbon derives from riverine inputs of terrigenous material and Holocene marine production. During the past four decades, this current production material was enriched with contributions from nuclear weapons testing and Sellafield and Amersham discharges. The trends are brought about by intense mixing, and the younger ages may be due to relatively fast oxidation of the organic material (compared to the dissolution rate of carbonate) in a relatively oxic environment. Thus, little old organic carbon will remain and the organic fraction will be dominated by recent inputs of material. The fact that Core 2 is enriched to

>200 pM suggests that the rate of turnover of the bulk of the organic carbon must be of the order of a few tens of years.

CONCLUSION

We deduce from our findings that a significant proportion of the marine discharges of ^{14}C are in the form of CO_2 / CO_3^{2-} / HCO_3^- . During the summer months, a sufficient quantity of this ^{14}C -enriched DIC is taken up by phytoplankton during their bloom periods, so that they are enriched relative to the current ambient activity. This activity is subsequently transferred to higher trophic levels and, either directly or indirectly, to the sediment. We have already observed that, in the winter, particle fluxes are dominantly inorganic while in the summer, a greater proportion of organic particles is present. This high organic particle flux could have important implications for the transfer of particle reactive radionuclides through the food chain. Finally, the differences in ^{14}C activity between the inorganic and organic components of the sediments cast serious doubts over the validity of deriving sedimentation rates for Irish Sea sediments based on either the bulk organic or inorganic carbon components.

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