Muir, G. K. P., Cook, G. T., Tripney, B. G., MacKenzie, A. B., Stewart, H., and Tierney, K. M. (2015) Temporal trend in the transfer of Sellafield-derived 14C into different size fractions of the carbonate component of NE Irish Sea sediment. *Radiocarbon*, 57(3), pp. 347-354.

Version: Published

http://eprints.gla.ac.uk/103488/

Deposited on: 18 May 2014

TEMPORAL TREND IN THE TRANSFER OF SELLAFIELD-DERIVED ^{14}C INTO DIFFERENT SIZE FRACTIONS OF THE CARBONATE COMPONENT OF NE IRISH SEA SEDIMENT

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ABSTRACT. From 1994 onwards, radiocarbon discharges from the Sellafield nuclear fuel reprocessing plant have been made largely to the northeast Irish Sea. They represent the largest contributor to UK and European populations of the collective dose commitment derived from the entire nuclear industry discharges. Consequently, it is important to understand the long-term fate of ^{14}C in the marine environment. Research undertaken in 2000 suggested that the carbonate component of northeast Irish Sea sediments would increase in ^{14}C activity as mollusk shells, which have become enriched in Sellafield-derived ^{14}C, are broken down by physical processes including wave action and incorporated into intertidal and subtidal sediments. The current study, undertaken in 2011, tested this hypothesis. The results demonstrate significant increases in ^{14}C enrichments found in whole mussel shells compared to those measured in 2000. Additionally, in 2000, there was an enrichment above ambient background within only the largest size fraction (>500 μm) of the intertidal inorganic sediment at Nethertown and Flimby (north of Sellafield). In comparison, the present study has demonstrated ^{14}C enrichments above ambient background in most size fractions at sites up to 40 km north of Sellafield, confirming the hypothesis set out more than a decade ago.

INTRODUCTION

The Sellafield nuclear complex, located on the Cumbrian coast of northwest England (Figure 1), has been in operation since the early 1950s. Current operations include reactor decommissioning, fuel reprocessing, and storage of nuclear materials, including radioactive wastes. During operations, effluent is discharged to the Irish Sea and the atmosphere, or disposed of as solid waste to the low-level waste repository at Drigg, situated south of Sellafield (Nuclear Decommissioning Authority 2011). The discharges of liquid radioactive waste are via twin pipelines extending 2.1 km into the eastern Irish Sea. Peak liquid discharges of transuranic radionuclides, including [241]Am and [241]Pu (Figure 2), and fission and activation products including [137]Cs (Figure 3) occurred during the early to mid-1970s (Gray et al. 1995), with annual discharges during this period being 2 to 3 orders of magnitude greater than contemporary values (RIFE 1996–2012; MacKenzie et al. 1999). The environmental fate of transuranic radionuclides and fission/activation products has been the focus of extensive research, primarily as a consequence of their contribution to critical group exposure (e.g. MacKenzie et al. 2004). This research has demonstrated that an area of fine sediment, offshore from Sellafield and known as the Sellafield mud-patch, has acted as a sink for the discharges. Particle reactive transuranic species, with high distribution coefficients (K_d), e.g. [241]Am[3+], were almost totally retained (Aston and Stanners 1982; Pentreath et al. 1984), in contrast to [137]Cs (single oxidation state of +1, and low K_d) for which there was approximately 10% retention and approximately 90% distribution in solution beyond the Irish Sea (MacKenzie et al. 1994; McCartney et al. 1994). Rather than the current discharges, the mud-patch sediment is now the major source term for radionuclide distribution to the wider environment (including intertidal areas) in a general northwards direction, either through redissolution or redistribution of the fine sediment (Aston et al. 1981; Aston and Stanners 1982; Pentreath et al. 1984; Hunt 1985; MacKenzie et al. 1987, 1994, 1999, 2004; McDonald et al. 1990; Cook et al. 1995, 1997).

In comparison, relatively few investigations have been undertaken into the environmental fate of Sellafield-derived aqueous ^{14}C discharges. Over the lifetime of the Sellafield site, total aqueous [14]C

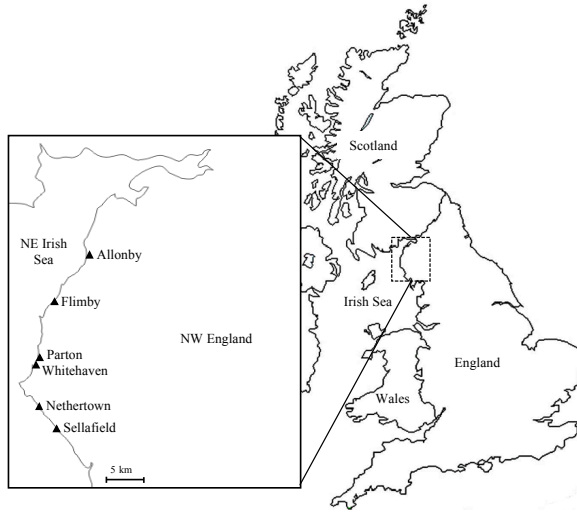1. Corresponding author. Email: Graham.Muir@glasgow.ac.uk.

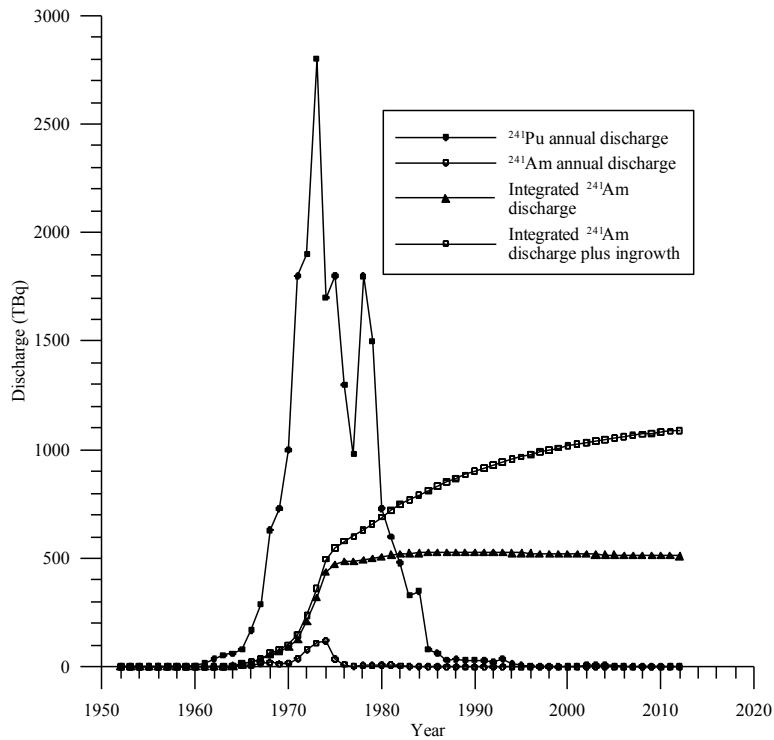Figure 1 Map of the UK and the Sellafield area (inset)

Figure 2 ^{241}Am and ^{241}Pu discharges from Sellafield and associated ^{241}Am ingrowth from ^{241}Pu (1952–2012).

discharges have been small in comparison with other radionuclides, but in contrast to the pattern of most others, they have increased by about an order of magnitude since the early 1990s (Cook et al. 2004a). They ranged between approximately 1 and 3 TBq per annum until 1994, when, partly due to a change in discharge policy that diverted the majority of the atmospheric discharges to the aqueous stream, discharges increased significantly, with a maximum of 17.0 TBq occurring in 2003 (Figure 3). The total ^{14}C discharged to the marine environment between 1994 and 2011 was 153 TBq,

103 TBq of which were discharged between 2001 and 2011 (MAFF 1994, 1995; RIFE 1996–2012). The latter value is particularly relevant as it represents the timeframe of this study.
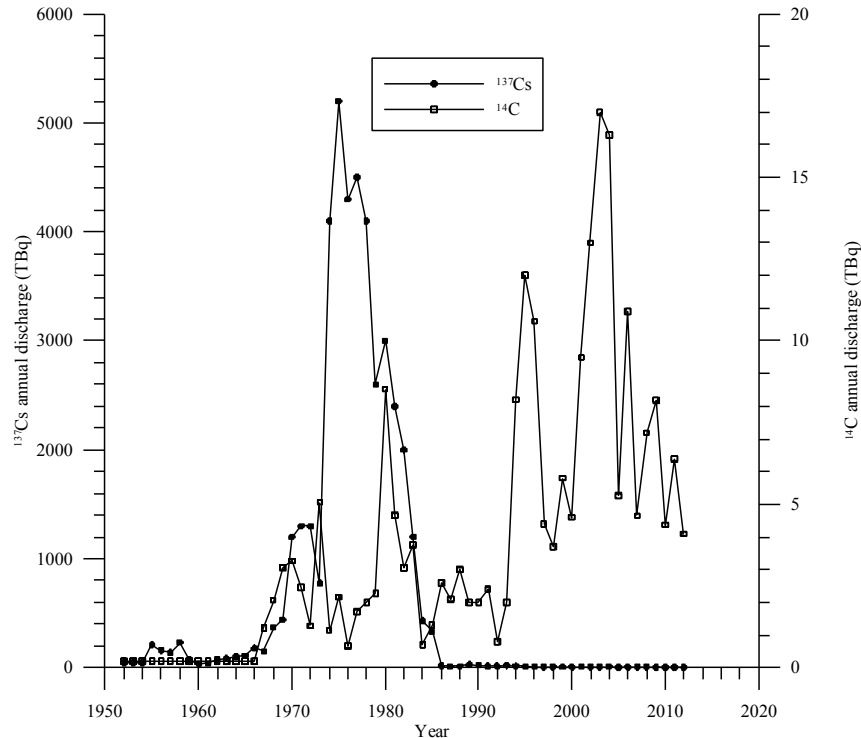

Figure 3 ^{137}Cs and ^{14}C aquatic discharges from Sellafield (1952–2012)

Sellafield ^{14}C is discharged into an existing "background" pool of ^{14}C derived from both natural production and atmospheric testing of nuclear weapons. Cook et al. (1998) investigated ^{14}C activities in marine dissolved inorganic carbon (DIC) and biota at sites remote from ^{14}C inputs around the UK and Ireland and subsequently defined the background activity as 247.6 ± 1.0 Bq kg^{-1}. This value was proposed as a best estimate of natural production/nuclear weapons testing "background" for 1995. We subsequently redefined the background in 2014 and obtained an almost identical value (248.6 ± 0.8 Bq kg^{-1}). Therefore, a value of 248 Bq kg^{-1}, which represents what we consider to be a good estimate of the background ^{14}C activity, is indicated in the figures in order to demonstrate the Sellafield ^{14}C inputs more clearly.

The dispersion and sediment uptake processes affecting ^{14}C are significantly different from those of other radionuclides. Sellafield-derived ^{14}C is discharged into the Irish Sea primarily as inorganic carbon (Begg et al. 1992; Cook et al. 1995), which is highly soluble in seawater and rapidly becomes part of the DIC reservoir. While most is distributed beyond the Irish Sea in solution *via* the North Channel (Wolstenholme et al. 1998; Gulliver et al. 2001), a small proportion is retained by the following mechanisms: (1) The DIC is readily utilized by primary producing organisms (phytoplankton and macroalgae) through fixation of CO_3^{2-} and HCO_3^{-} during photosynthesis (Lalli and Parsons 1993). The ^{14}C is then transferred through the food chain from plants to animals. (2) The DIC is also taken up directly in the formation of the carbonate shells of marine mollusks (McConnaughey et al. 1997; Gillikin et al. 2006); therefore, ^{14}C is also transferred into the inorganic components of the marine biota. Thus, its behavior cannot be described by a sorption coefficient (K_d). Ultimately, a

proportion of the ^{14}C is deposited either directly or indirectly, as organic carbon or carbonates, into a mixed-age sediment pool (Begg et al. 1992; Cook et al. 1995, 2004b; Wolstenholme et al. 1998; Gulliver et al. 2001; MacKenzie et al. 2004). ^{14}C contributes only 4.2% to the current total dose rate received by critical consumer groups of marine fish and shellfish from the Cumbrian coast; nevertheless, due to its long half-life (5730 yr), ready entry into the food chain, and high environmental mobility, the aqueous ^{14}C discharges from Sellafield are the major contributor to the collective dose commitment to UK and European populations from the entire nuclear industry (BNFL 2002). Also, according to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 2008), the largest collective total dose estimates remain associated with effluent release of ^{14}C. Thus, an improved understanding of the behavior of Sellafield ^{14}C discharges over decadal periods is required to improve our understanding of its long-term (century to millennium) fate. The specific objective of the present study was to collect samples from the original sampling locations visited in 2000 to test the hypothesis that mollusk shells, which were demonstrated to be enriched in ^{14}C, are being broken down in the environment, leading to retention and a gradual increase in the ^{14}C activity of the inorganic component of NE Irish Sea sediments.

EXPERIMENTAL

Samples were collected during June 2011 from the four sites previously sampled in May 2000 (Nethertown, Parton, Flimby, and Allonby) (Figure 1). Approximately 5 kg of surface sediment, including whole shells from a range of species (mussels, winkles, and limpets) and shell fragments, were collected randomly from several intertidal locations at each of the four sites. Whole shells and large shell fragments (>2 mm) were separated from the sediment, which was mixed, oven-dried, and dry-sieved into three size fractions (<63 μm, 63–500 μm, and >500 μm–2 mm).

Shell fragments and sieved sediments were well mixed, subsampled, and subsequently hydrolyzed with 1M HCl to liberate CO_2, which was cryogenically purified under vacuum using liquid N_2, in preparation for accelerator mass spectrometry (AMS) analysis. Subsamples (3 mL) of CO_2 were converted to graphite according to the procedure described by Slota et al. (1987) and measured on a 250kV single-stage accelerator mass spectrometer (SSAMS) according to Freeman et al. (2008, 2010) and with quality assurance standards as described in Naysmith et al. (2010). Results were calculated relative to the international standard oxalic acid II. AMS results, reported as fraction modern values, were converted to specific activities (Bq kg^{-1} C) using the calculation regime for enhanced activity samples described by Mook and van der Plicht (1999). $\delta^{13}C$ measurements were made offline by isotope ratio mass spectrometry using a VG Sira 10.

In 2000, only Nethertown beach yielded sufficient fine (<63 μm) material for radiometric ^{14}C analysis. Sediment fractions were analyzed and reported as ^{14}C-specific activities in either <500-μm or >500-μm size fractions (Cook et al. 2004b). The introduction of AMS has facilitated the analysis of milligram quantities of fine sediment fractions. It was therefore possible to analyze three size fractions collected in 2011. The <500-μm fraction reported in Cook et al. (2004b) is analogous, and is reported as the 63–500-μm fraction in this study.

RESULTS AND DISCUSSION

^{14}C activities in sediments and biota collected during studies undertaken in 2000 and 2011 are presented in Table 1. Figure 4 compares ^{14}C data for whole shells and shell fragments collected in 2011. The results demonstrate significant enhancements above the natural production/nuclear weapons testing background in all species present [common limpet (*Patella vulgata*), common winkle (*Littorina littorea*), and common mussel (*Mytilus edulis*)] and at all sites. There is also a general decrease in activity with increasing distance from Sellafield.

Table 1 Gross ^{14}C activities (Bq kg^{-1} C ± 1σ) in sediment size fractions and whole shell/shell fragments from four sampling sites situated north of Sellafield in 2000 and 2011.

| | Sampling site | | | |
| | Nethertown | Parton | Flimby | Allonby |
Sampling period	(Bq kg^{-1} C)	(Bq kg^{-1} C)	(Bq kg^{-1} C)	(Bq kg^{-1} C)
2000				
Mussel shell/fragments	2001.7 ± 6.8	1092.5 ± 4.4	856.7 ± 3.6	768.9 ± 3.0
Sediment >500 μm–2 mm	360.4 ± 1.6	127.7 ± 0.8	259.9 ± 1.3	153.6 ± 0.9
Sediment 63–500 μm	148.6 ± 1.3	22.3 ± 2.7	68.9 ± 1.8	70.5 ± 0.8
Sediment <63 μm	188.3 ± 1.9	nd	nd	nd
2011				
Mussel shell/fragments	2596.2 ± 7.4	1430.8 ± 4.3	1280.1 ± 3.7	972.2 ± 3.0
Winkle shell/fragments	1286.2 ± 3.8	1643.4 ± 4.8	1176.4 ± 3.1	355.2 ± 1.0
Limpet shell/fragments	2898.7 ± 9.0	1227.7 ± 3.8	598.3 ± 1.8	na
Sediment >500 μm–2 mm	2080.0 ± 6.5	429.0 ± 1.5	679.5 ± 2.4	337.8 ± 1.2
Sediment 63–500 μm	921.6 ± 3.3	162.7 ± 0.6	304.4 ± 0.9	127.3 ± 0.5
Sediment <63 μm	898.3 ± 3.2	463.2 ± 1.7	399.5 ± 1.4	89.0 ± 0.3

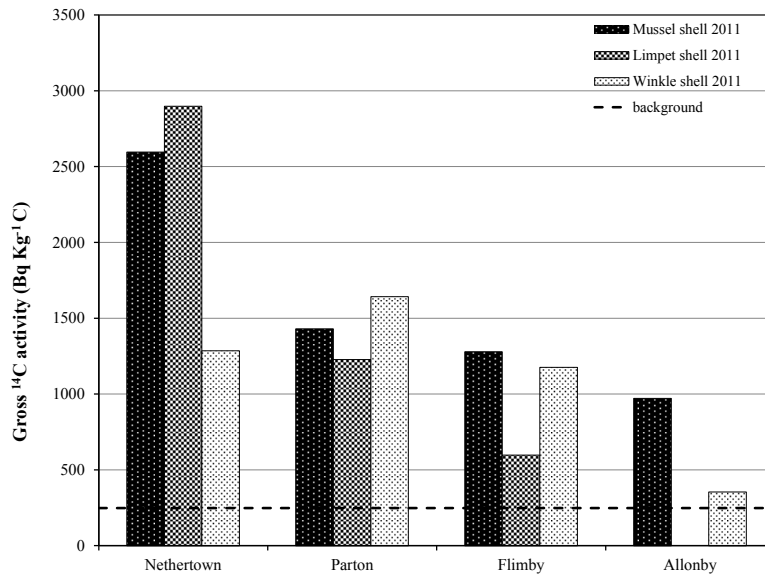Notes: nd – no data; na – no species available.

Figure 4 Mussel, limpet and winkle shell gross specific ^{14}C activities from Nethertown, Parton, Flimby, and Allonby, collected in May 2011.

Figures 5a and 5b demonstrate that mussel shells collected in 2011 are significantly enriched in ^{14}C at all sites compared to the same species collected in 2000, reflecting increased ^{14}C discharges post-2000. The gross mean activity for mussel shells was 1569 Bq kg^{-1} in 2011, compared to 1179 Bq kg^{-1} in 2000, representing a mean overall increase of ~34%. Proportionally, mussel shell ^{14}C activities at Nethertown, Parton, Flimby, and Allonby were enhanced by 30%, 31%, 49%, and 26%, respectively, compared to previous activities.
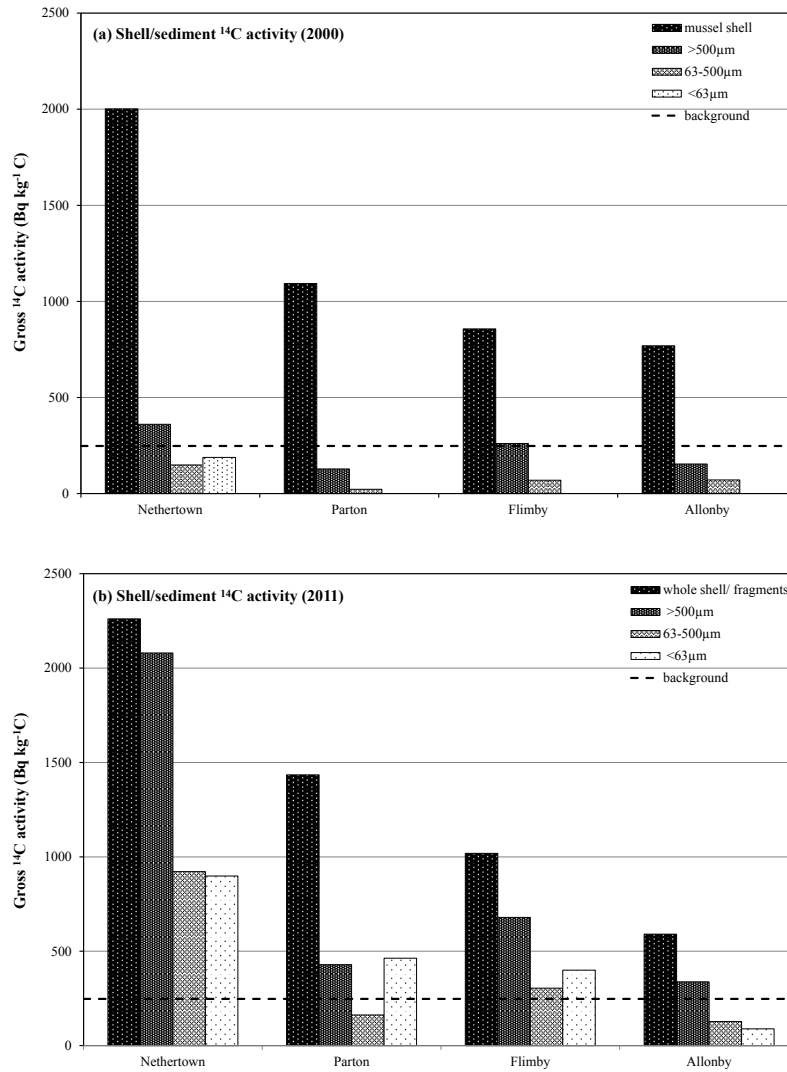
Figure 5 (a) Gross ¹⁴C-specific activities in different size fractions of the carbonate component of NE Irish Sea intertidal sediment measured at four sites in 2000. (b) Gross ¹⁴C-specific activities in different size fractions of the carbonate component of NE Irish Sea intertidal sediment measured at the same four sites in 2011.

All intertidal sediment fractions collected in 2011 are enhanced in ¹⁴C relative to comparable samples collected in 2000 (Figures 5a and 5b). At each of the four sites, there is an obvious trend of decreasing ¹⁴C activity with decreasing particle size with: [whole shells/shell fragments] > [>500-µm size fraction] > [63–500 µm size fraction]. Only the <63-µm size fraction does not follow this trend. There is also a general trend of decreasing ¹⁴C activity with increasing distance from Sellafield.

In 2000, only two sediment fractions, Flimby >500 µm and Nethertown >500 µm, had activities enhanced above the ambient background level. In contrast, in 2011, all but three samples [Allonby (<63 µm and 63–500 µm) and Parton (63–500 µm)] have ¹⁴C activities above the ambient background level. Sites previously depleted in ¹⁴C, with activities comparable to subtidal carbonate sediments (Begg 1992; Cook et al. 1995), now show ¹⁴C enrichments, consistent with increased ¹⁴C

discharges from Sellafield and an increased period for breakdown of shells containing Sellafield-derived [14]C. This again satisfies the hypothesis of cumulative ^{14}C transfer to finer fractions, suggested by Cook et al. (2004b). Average ^{14}C activity increases in 2011 sediments were 710, 301, and 656 Bq kg[−1] for the <63 µm, 63–500 µm, and >500 µm–2 mm fractions, respectively, on comparable sediments measured in 2000. Fivefold increases in mean ^{14}C activities compared to previous (2000) activities were measured in the <63-µm and 63–500 µm fractions, respectively, and 3.5 times in the >500-µm fraction. These increases are much greater than the increase in discharges post-2000 compared to pre-2000 and are indicative of the cumulative effect of the breakdown process. The Nethertown (>500 µm) fraction has the highest ^{14}C activity from any site (2080 ± 6.5 Bq kg[−1]), reflecting its close proximity to the discharge point from Sellafield and the fact that coarse material has been in the intertidal environment for a shorter period of time, most likely encompassing the period of high discharges. In 2000, only one <63-µm measurement was available (Nethertown). In 2011, this fraction is ~5 times the 2000 activity (188.3 ± 1.9 compared to 898.4 ± 3.2 Bq kg[−1]). The finest fraction (<63 µm) activities again show a trend of decreasing activity with distance from Sellafield, consistent with the northwards flowing currents and well-established northwards movement of sediment (Mackenzie et al. 1987, 1994, 1998, 1999; McDonald et al. 1990; Cook et al. 1997). However, at Parton and Flimby, the activities are very similar and are greater than those of the 63–500 µm fractions, which would seem to argue against the concept that the larger fractions should have the higher activities. This can potentially be explained by two mechanisms: There is (1) greater northwards movement of the <63-µm fraction compared to the coarser material or (2) there is more rapid breakdown of the shell material at these two sites.

CONCLUSIONS

Over the period from 2000 to 2011, mollusk shells and all size fractions of the carbonate component of sediment collected at four intertidal locations north of the Sellafield nuclear fuel reprocessing plant show significant increases in ^{14}C activity, indicative of incorporation of ^{14}C discharged from Sellafield into DIC, ^{14}C transfer to shell material in living mollusks, subsequent death of these organisms, environmental breakdown of shell material, and finally, incorporation into sediment. The intertidal sediments provide clear temporal evidence of gradual enrichment and retention of ^{14}C in all size fractions, particularly in the finer fractions, which were previously depleted relative to ambient background levels. Intertidal mollusks are highly efficient at assimilating and "fixing" [14]C from the DIC component of seawater directly into shell carbonate and this process, over the short-to-medium term, will be a major factor in ^{14}C accumulation in NE Irish Sea intertidal sediment. Predicating a steady state concurrent with Sellafield ^{14}C discharges is difficult, given the environmental mobility and rapid accumulation of ^{14}C noted over the decadal period of this study. Nevertheless, the observed accretion of ^{14}C in biota and sediment is of long-term radiological significance as this material is subject to lateral offshore transport, mixing and resupply to coastal locations, with potential implications for near- and far-field accumulation.

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