

SELLAFIELD-DERIVED ANTHROPOGENIC ^{14}C IN THE MARINE INTERTIDAL ENVIRONMENT OF THE NE IRISH SEA

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ABSTRACT. The intertidal biota from Parton beach, close to the Sellafield nuclear fuel reprocessing plant, were all found to be enriched in radiocarbon relative to ambient background. The degree of enrichment appears to reflect the positions of the biota in the food chain once the dilution in seaweed from atmospheric uptake is taken into account. Close to the low-water mark, the order was mussels > limpets > anemones ≅ winkles > seaweed. The same order was observed close to the high-water mark, except that anemones were absent from this area. The activities in the biogeochemical fractions of the water column reflect the fact that discharges are primarily in the form of dissolved inorganic carbon (DIC), which is subsequently transferred to the particulate organic carbon (POC) and, to a lesser extent, the dissolved organic carbon (DOC), and finally, the particulate inorganic carbon (PIC). Analysis of intertidal sediment suggests that there is likely to be a gradual increase in the specific activity of ^{14}C in the inorganic component of this material as Sellafield contaminated organisms die and their shells are ground down by natural processes.

INTRODUCTION

The Sellafield nuclear fuel reprocessing plant on the Cumbrian coast of NW England (Figure 1) commenced operations in October 1950. The site is operated by British Nuclear Fuels plc (BNFL) and currently hosts a range of operations including magnox and thermal oxide fuel reprocessing, waste management, reactor decommissioning and 4 magnox reactors that form the Calder Hall nuclear power generating facility (BNFL 2002). During reprocessing operations, low-activity waste is discharged to the atmosphere, to sea, or is disposed of as solid waste, either on-site or at the Drigg disposal site to the south of Sellafield. The aquatic discharges which are the subject of this study are via a 2.1 km length pipeline into the NE Irish Sea. The aquatic discharges of most Sellafield-derived radionuclides reached a peak in the early to mid-1970s, and since then, as a result of the introduction of several waste treatment plants, have been reduced by 2 to 3 orders of magnitude from their peak values (MacKenzie 2000). Up to 1984, the reported aquatic ^{14}C discharges were estimated and subsequent to this time, they were measured, typically on a monthly basis. In contrast to most other radionuclides, the reported ^{14}C discharges remained approximately constant at <1–3 TBq per annum until 1994, at which time there were significant increases (BNFL 1997). These were due primarily to a change in discharge policy, whereby discharges previously released to the atmosphere were diverted (as a consequence of introducing a gas scrubber) to the aqueous route, and to a lesser extent, to an increase in reprocessing. This change to the aqueous route also has the positive effect of reducing the radiological impact of the discharges (Begg 1992). Although the total discharges of ^{14}C from Sellafield are small in comparison with many other radionuclides, they are of radiological importance. The marine critical group consists of people in the coastal area of Cumbria who are high consumers of fish and shellfish from the Sellafield area. The most recent BNFL data indicate that ^{14}C accounts for about 4% of the dose to that critical group. However, in terms of the collective dose derived from the aquatic Sellafield discharges, ^{14}C represents the largest contributor to the UK, European, and world populations (BNFL 2002).

A previous study (Begg et al. 1992), which was designed to investigate the biogeochemistry and cycling of ^{14}C in the Sellafield environment, demonstrated that for several tens of km to the north

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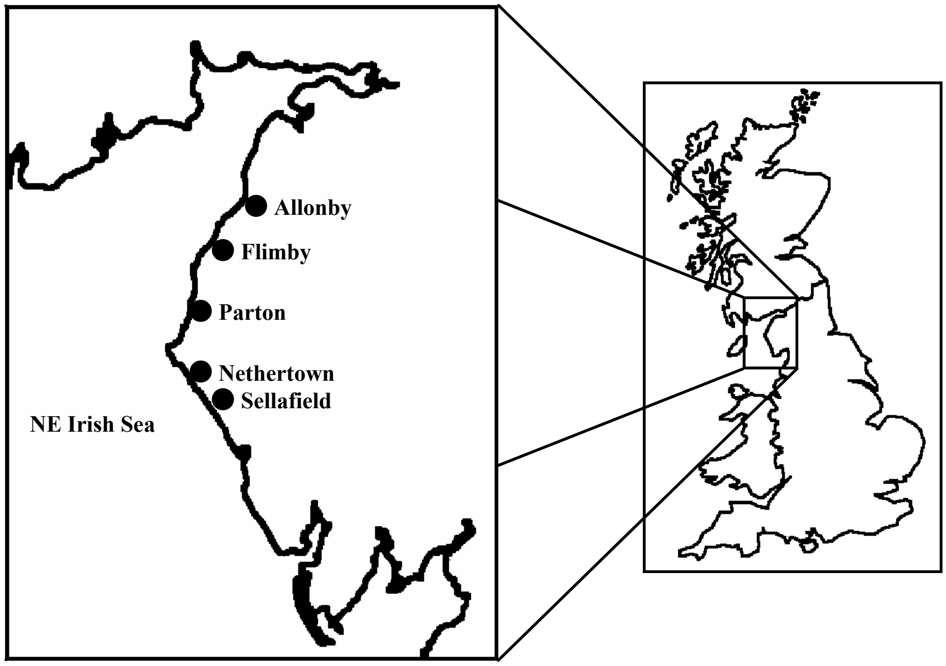

Figure 1 Map of sampling sites

and south of Sellafield, specific activities of ^{14}C were enhanced in a range of biota and that the relative degree of enhancement was always mussels (*Mytilus edulis*) > winkles (*Littorina littorea*) > seaweed (*Fucus* sp.). Begg al. (1992) also confirmed, from analysis of water column samples, that the aquatic ^{14}C discharges were predominantly in the form of dissolved inorganic carbon (DIC). However, the following questions remained unanswered by this study:

- Why did mussels, which are filter feeders, have the greatest ^{14}C enrichments, yet there was no evidence of ^{14}C enrichment in the particulate organic carbon component of the water column?
- Why did seaweed (*Fucus* sp.) have the lowest enhancements when they are primary producers, deriving their carbon directly from the DIC component, which was the only consistently enriched fraction of the water column?

In an attempt to provide a better explanation for these trends, it was decided that we should sample a greater range of intertidal biota, namely, seaweed (*Fucus* sp.), winkles (*Littorina littorea*), mussels (*Mytilus edulis*), anemones (*Actinia equina*), and limpets (*Patella vulgata*), together with water column samples. Also, whenever possible, biota samples were taken for analysis from close to both the low- and high-water marks. Seawater samples were collected periodically from the Sellafield area and ^{14}C analysis was undertaken on the particulate inorganic carbon (PIC), particulate organic carbon (POC), dissolved inorganic carbon (DIC), and dissolved organic carbon (DOC) fractions. Finally, intertidal sediment was collected from a number of locations to assess any potential long-term effects deriving from the breakdown of ^{14}C-contaminated shell from organisms such as winkles, mussels, limpets, etc.

MATERIALS AND METHODS

We collected the biota samples from Parton beach, approximately 20 km north of Sellafield, having chosen this site because of its proximity to the plant and for the range of biota that it supports. All biota samples were collected in May 2000 and were chosen for their variable feeding habits, in order to explore further the relationship between different feeding behaviors and variations in ^{14}C concentrations within the intertidal environment. Mussels, winkles, seaweed, limpets, and common beadlet anemone were all collected from close to the low-water mark. Close to the high-water zone, the same species were collected with the exception of the anemone, which was completely absent from this zone. We collected approximately 1 kg fresh weight of each invertebrate species, and upon our return to the laboratory, washed them thoroughly in dilute HCl and then in distilled water. We then boiled the mussels, winkles, and limpets for a few minutes to aid removal of the soft tissue from the shell and then re-washed them. We then freeze-dried and homogenized all samples in preparation for analysis. Seaweed samples were washed in dilute HCl, then in distilled water, oven-dried at 60 °C, and then homogenized. Radiometric ^{14}C analysis was carried out according to the method detailed in Begg et al. (1992).

We collected 100-L seawater samples periodically between November 1989 and February 1999 at a location close to the Sellafield plant. The samples were filtered through 0.7-µm glass fiber filters and the filtered particles stored at –22 °C prior to analysis. Where DOC analysis was carried out, the water samples were poisoned with mercuric chloride and stored in the dark at 4 °C prior to analysis. The 4 biogeochemical fractions (PIC, POC, DIC, and DOC) were analyzed according to the methods of Gulliver et al. (2001) and Wolstenholme (1999).

The sediment samples were collected from the intertidal area at 4 locations (Nethertown, Parton, Flimby, and Allonby) as depicted in Figure 1. Large fragments or indeed whole shells (mainly mussel and winkle) lying on the sediment surface were also collected. The sediment was sieved into 2 size fractions, <500 and >500 µm. These fractions and the shells were hydrolyzed with 4M HCl and analyzed for ^{14}C according to the method of Begg et al. (1992)

RESULTS

Figure 2 depicts the ^{14}C activities in intertidal biota from Parton once the contributions from weapons testing fallout and natural production for that year (approximately 248 Bq kg^{-1} C) had been removed (Gulliver 2002). This illustrates that the intertidal biota are all enriched in ^{14}C and that the trend in activity is mussels > winkles > limpets > seaweed, close to the high-water mark, and mussels > limpets > anemone ≅ winkles > seaweed, close to the low-water mark. The trend in ^{14}C activities of mussels > winkles > seaweed is, therefore, the same as that demonstrated by Begg et al. (1992) and applies to biota collected at close to the high- and low-water marks.

Gross ^{14}C activities (i.e. not corrected for natural production and weapons testing fallout) for the 4 geochemical fractions of the water column are presented in Table 1. Gross values are given because the background contribution to the DOC, POC, and PIC activities are difficult to estimate since they are dependent on local variables such as contributions from terrestrial run-off, sediment resuspension, level of primary production, etc. The DIC value for background is not subject to such local variables and is approximately 250 Bq kg^{-1} C (Gulliver et al. 2001). With the exception of 1 sampling date (Feb 1995), the DIC component was always enriched above ambient background. Also, from July 1995 onwards, the POC fraction was consistently enriched above the ambient background for the DIC, which can be taken as a maximum value for any of the geochemical components. The PIC values are highly variable and always depleted relative to the DIC and POC values for the corresponding sampling date. Three samples from 1997 and 1998 certainly are indicative of substantial enrichments above ambient background. DOC values were only measured until November 1996 and the result from this last sampling is certainly indicative of a substantial enrichment.

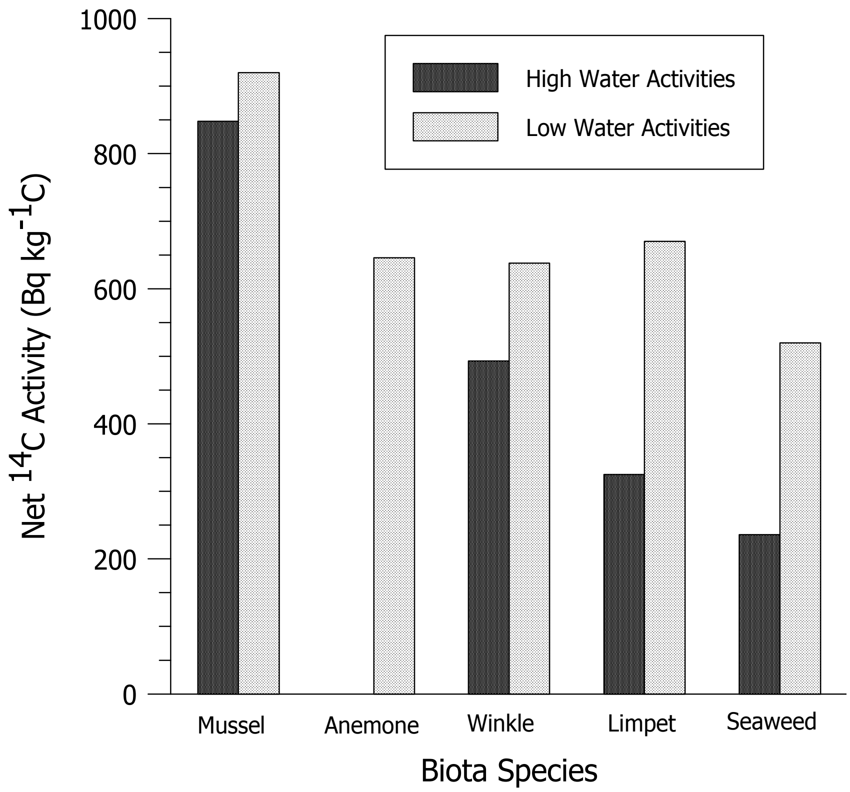

Figure 2 ^{14}C activities in a range of intertidal biota from Parton

Table 1 Gross ^{14}C activities (Bq kg^{-1} C) in the 4 geochemical fractions of the water column from the vicinity of Sellafield.

Collection date	DIC (Bq kg^{-1} C)	POC (Bq kg^{-1} C)	DOC (Bq kg^{-1} C)	PIC (Bq kg^{-1} C)
Dec '89[a]	550 ± 4	165 ± 2	143 ± 2	108 ± 1
Feb '91[a]	452 ± 2	182 ± 3	87 ± 2	134 ± 3
Feb '95[b]	247 ± 3	80 ± 1	218 ± 1	150 ± 2
July '95[b]	1099 ± 7	406 ± 3	107 ± 5	285 ± 3
Nov '95[b]	1538 ± 10	451 ± 2	76 ± 1	145 ± 1
Mar '96[b]	945 ± 4	478 ± 2	132 ± 1	144 ± 1
June '96[b]	1227 ± 5	612 ± 4	183 ± 1	180 ± 1
Nov '96[b]	1677 ± 9	614 ± 3	598 ± 3	201 ± 1
Nov '97	4553 ± 18	3616 ± 12	n/a	796 ± 11
Feb '98	1365 ± 4	631 ± 3	n/a	72 ± 10
May '98	1853 ± 43	1146 ± 4	n/a	860 ± 11
Aug '98	1160 ± 14	1024 ± 4	n/a	579 ± 10
Nov '98	486 ± 5	343 ± 2	n/a	103 ± 11
Feb '99	453 ± 2	296 ± 1	n/a	75 ± 10

[a]Begg 1992.
[b]Wolstenholme 1999.

The results for ^{14}C analysis of intertidal sediments are shown in Figure 3. The <500-µm size fraction data are consistent with sub-tidal inorganic sediment activities, while activities in the >500-µm fraction are consistently higher than the sub-tidal activities (Gulliver 2002). Finally, the whole shells/shell fragments are all significantly enriched with respect to the ambient background activity.

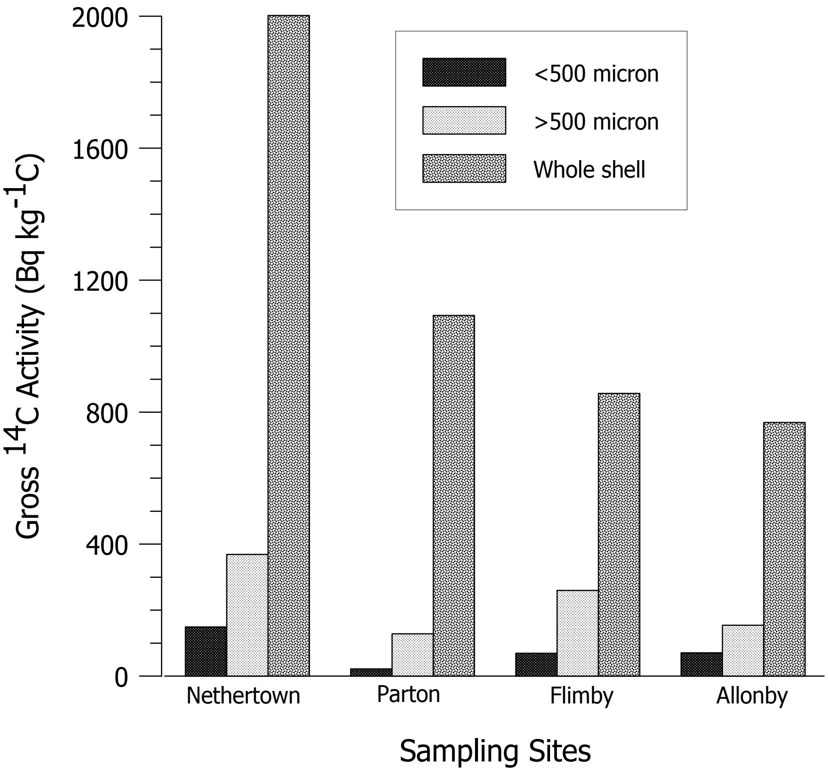

Figure 3 ^{14}C activities in different size fractions of the shell component of intertidal sediments

DISCUSSION

Biota

The explanation for mussels always having the highest activity lies in the fact that they are singularly suspension feeders, trapping fine food particles suspended in the water column. Water flows through an inhalant syphon and over the gills, where food particles in the form of phytoplankton and detritus are trapped on a mucous coating while the filtered water is expelled through an exhalant aperture. Particles that are too large to be digested or are recognized as pseudofeces are ejected (Barnes 1987). As (i) phytoplankton are the primary producers in the marine environment, deriving their carbon from the DIC component of the water, (ii) Sellafield marine ^{14}C discharges are almost entirely in the form of DIC, and (iii) the main food source of mussels is phytoplankton, it is reasonable to assume that the mussels are ingesting relatively active material due to this selective feeding habit as a primary consumer. It can also be noted from Figure 2 that there is little difference in activity between the low-water and high-water mark samples, consistent with mussels being filter feeders.

Seaweed (*Fucus* sp) consistently has the lowest ^{14}C activity of all biota analyzed. Since, like phytoplankton, seaweeds are primary producers that derive their carbon for photosynthesis from the DIC component of seawater, it could be expected that their activities should be similar to those of mussels (primary consumers). However, many seaweed species (including *Fucus*) have the ability to derive carbon from atmospheric CO_2 when they are exposed to the atmosphere. The activity of seaweed relative to mussels must then depend upon (i) the proportion of time that the seaweed is submerged and (ii) the ^{14}C activity of the DIC component of the water column relative to that of the atmosphere, which may be perturbed locally by the atmospheric Sellafield discharges. The fact that the seaweed ^{14}C activities are significantly lower than those in mussels implies that the ^{14}C activity of the atmosphere (Bq kg^{-1} C) is lower than that of the DIC. This can be substantiated by the fact that Isogai et al. (2002) demonstrated that the largest excess ^{14}C activity above ambient background in tree rings from the years 1994 to 1999 was 125 Bq kg^{-1} C in 1995 when the discharges to the atmosphere totalled 4.6 TBq for that year. These data were derived from analysis of a section of oak tree felled in September 1999 at a location 1.5 km east and slightly to the north of Sellafield. In contrast, the intertidal biota samples were collected in 2000 at Parton, which is approximately 20 km northwest from Sellafield, where the excess will undoubtedly be much reduced (the dominant wind direction is roughly from the southwest). Furthermore, the atmospheric discharges in that year totalled 2.9 TBq. Therefore, it can reasonably be assumed that the excess ^{14}C activity in the atmosphere in 2000 at Parton would have been significantly less than 125 Bq kg^{-1} C. It should also be noted from Figure 2 that the seaweed ^{14}C activity close to the low-water mark is approximately double that at the high-water mark, consistent with the fact that seaweed will derive CO_2 from the atmosphere when exposed. The activity of the seaweed close to the low-water mark is still significantly lower than that of the mussels. We would propose that this is because the seaweed close to low water is still exposed to the atmosphere for a significant period of time and also that the integration time of ^{14}C may differ for these 2 forms of biota.

The beadlet anemone feeds on a number of available food sources, including mussel flesh, zooplankton, small crustaceans, and worms. The food is captured on tentacles, paralyzed, and then carried to the mouth. The mussel flesh has been demonstrated here to be highly enriched in ^{14}C, while zooplankton, like mussels, are primary consumers which feed on phytoplankton and are also likely to be highly enriched. In contrast, it can be speculated on the basis of the relatively minor ^{14}C enrichments (and often lack of enrichment) of the organic component of the sediments within this area (Gulliver 2002) that the worms and small crustaceans which live in the surface sediment will show similar minor enrichments. Overall, the data would seem to reflect the position of the beadlet anemone in the trophic food chain as a secondary consumer.

Limpets and winkles can be dealt with together since their feeding behaviors are quite similar. Both species are herbivorous and at high tide they emerge from their protective shells to rasp algae (including *Fucus* sp.) and detritus that cover the rocks. Thus, their activities will reflect those of seaweed and detritus (including plankton); hence, the activities are intermediate between mussels and seaweed. Many of the winkles were collected from tidal pools and this could account for the fact that the high- and low-water activities are similar, i.e., they fed on seaweeds that were covered by seawater for a significant period.

Water Column

The water column results appear to reflect the change in discharge policy that took effect in 1994. After this time, the activities in the 4 biogeochemical fractions are typically greater than pre-1994. The POC component, the food source for mussels, is always depleted in ^{14}C relative to the DIC, which appears to make the trend in activities of the mussels and seaweed difficult to explain. However, the following 3 factors have to be taken into account as discussed above:

1. Seaweed derive carbon from the atmosphere when not covered by seawater;
2. The activity of ^{14}C in the atmosphere is significantly lower than the DIC of the water column;
3. The mussels are selective feeders that can exclude certain particles. When these factors are taken into account, it becomes possible to understand why the activities in mussels exceed those of seaweed.

Sediment

The results presented in Figure 3 suggest that as contemporary shell is broken down and is incorporated into the sediment, there will be a gradual increase in the ^{14}C activity of the inorganic component of the sediment with time. The current ^{14}C activity of the inorganic component is low, indicating that it is dominantly pre-Sellafield in origin. The rate at which the inorganic sediment ^{14}C activity increases will depend upon future trends in ^{14}C discharges, the mass of Sellafield-era shell material relative to the pre-Sellafield mass, the ^{14}C activity of the Sellafield-era shell material, and the rate at which natural processes break it down into fine material.

CONCLUSIONS

The ^{14}C activities in the biota appear to reflect their positions in the food chain once the dilution in seaweed from atmospheric uptake is taken into account. The activities in the biogeochemical fractions of the water column reflect the fact that discharges are primarily in the form of DIC, which is subsequently transferred to the POC and to a lesser extent the DOC and finally the PIC. Analysis of intertidal sediment suggests that there is likely to be a gradual increase in the specific activity of ^{14}C in the inorganic component of this material as Sellafield contaminated organisms die and their shells are ground down by natural processes.

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