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# SOURCES OF ANTHROPOGENIC $^{14}C$ TO THE NORTH SEA

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**ABSTRACT.** The Sellafield nuclear fuel reprocessing plant on the northwest coast of England is the largest source of anthropogenic radiocarbon to the UK coastal environment. In a mid-1990s study of $^{14}C$ distribution around the UK coast, the pattern of dilution with increasing distance from Sellafield appeared to be perturbed by anomalously high $^{14}C$ activities in marine biota in the coastal environment of northeast England. This present study was undertaken during 1998 and 1999 to determine whether this $^{14}C$ enhancement was due to Sellafield or the nuclear power plants on the east coast. Seawater, seaweed (*Fucus* sp.), and mussel (*Mytilus edulis*) samples that were collected from the vicinity of the Torness and Hartlepool advanced gas-cooled reactor (AGR) nuclear power stations were all enhanced above the contemporary regional background activity derived from natural production and atmospheric nuclear weapons testing. We used previously published dilution factors and transfer times for $^{99}Tc$ between Sellafield and various points on the UK coast to determine likely Sellafield-derived $^{14}C$ contributions to the activities at the nuclear power plant sites. The results suggest that the activities observed at Torness, which are only marginally enhanced above the natural background activity, are possibly due to discharges from Sellafield; however, the significant $^{14}C$ enhancements at Hartlepool are not Sellafield-derived. Furthermore, since both reactors have the same fundamental design, the low activities at the Torness AGR imply that the activities at Hartlepool are not from the AGR, suggesting that there is an input of $^{14}C$ to the marine environment in the vicinity of Hartlepool which is probably non-nuclear-power related. However, there is no other authorized site in the area that could account for the observed $^{14}C$ enrichments; therefore, further research is required to ascertain the source of this $^{14}C$.

## INTRODUCTION

A 1995 investigation of $^{14}C$ activities in the UK coastal marine environment (Cook et al. 1998) analyzed the dissolved inorganic carbon (DIC) component of seawater, seaweed (*Fucus* species), and flesh from mussels *(Mytilus edulis)*, crab (*Cancer pagurus*), flatfish (plaice), and roundfish (cod). The study revealed a pattern of dilution of $^{14}C$ activity in the DIC with increasing distance from Sellafield (the largest source of anthropogenic $^{14}C$ to the coastal waters of the British Isles) in a clockwise manner around the British Isles, i.e., in the direction of the prevailing currents. The trend was perturbed by anomalously high $^{14}C$ activities at Whitby, located on the east coast of the British Isles (Figure 1). As the $^{14}C$ activities in the north of Scotland were significantly lower than those found at Whitby, Cook et al. (1998) concluded that the net activities found at Whitby were possibly due to releases from the nearby nuclear power producing facility at Hartlepool (Figure 1), which was one of the first advanced gas-cooled reactors (AGR) commissioned in the British Isles.

The use of Sellafield-derived radionuclides as a tracer for water movement is well established, and these studies have shown that the Sellafield discharges are carried north from the Irish Sea, through the North Channel (Figure 1), along the west coast of Scotland, around the north coast of Scotland, and subsequently south along the east coast of the UK (Jefferies et al. 1973, 1982; Prandle 1984; Dahlgaard 1995; Nines 1990; Kershaw and Baxter 1995). Recent studies using Sellafield-derived $^{99}Tc$ indicate transport times of approximately 9 and 24 months to Pentland Firth and Lowestoft (270 km south of Whitby), respectively (Leonard et al. 1997; McCubbin et al. 2002) (Figure 1). In order to investigate whether or not the activities above ambient background at Whitby were an anomaly or were a common feature close to AGR facilities, this study was carried out to investigate $^{14}C$ activities in the vicinity of Hartlepool and another AGR facility at Torness (Figure 1) in south-

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east Scotland. Samples were obtained from the immediate vicinity of the Hartlepool AGR in 1998 and 1999, and from the vicinity of the Torness AGR in 1999.


Figure 1 Map of the United Kingdom indicating sampling and other sites referenced in the text

## METHODS AND MATERIALS

The following suite of samples was collected from the vicinity of both the Hartlepool and Torness nuclear power plants: DIC extracted from 100 L of water, seaweed (*Fucus* sp.), and mussels (*Mytilus edulis*).

The water samples were acidified, purged with nitrogen, and the $CO_2$ collected and converted to benzene for radiometric analysis, as described in Gulliver et al. (2001). The seaweed samples were washed thoroughly and then dried. The mussels were boiled briefly to aid removal of the flesh and then freeze-dried. These samples were then combusted and the $CO_2$ converted to benzene (Gulliver et al. 2001).

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At Torness, the samples were collected close to the coolant water outflow pipeline. At Hartlepool, the samples were collected from Parton Rocks, approximately 3 miles from the nuclear plant.

## RESULTS AND DISCUSSION

$^{14}C$ activities (Bq kg$^{-1}$ C) for the DIC and biota samples are presented in Table 1 as net activities above the relevant ambient background, which depends on the year of collection and species. The background activities were derived from Burtonport (Figure 1), a site on the west coast of Ireland which is not significantly influenced by discharges from the Sellafield plant (Cook et al. 1998; Gulliver et al. 2001). Also included for reference are net $^{14}C$ activities (Bq kg$^{-1}$ C) for the samples collected in 1995 from Whitby on the NE coast of England (Cook et al. 1998).

Table 1 Net $^{14}C$ activities (Bq kg$^{-1}$ C ± 1 σ) in DIC, seaweed, and mussels collected from the vicinity of the Hartlepool and Torness AGRs (1998 and 1999) and from Whitby (1995).

| Location | Sampling date | DIC | Mussels | Seaweed |
|---|---|---|---|---|
| Whitby[a] | 1995 | 23 ± 5 | 11 ± 3 | 23 ± 3 |
| Hartlepool | 1998 | 104 ± 4 | 561 ± 4 | 55 ± 3 |
| Hartlepool | 1999 | 94 ± 5 | 913 ± 4 | 50 ± 4 |
| Torness | 1999 | 16 ± 4 | 38 ± 3 | 27 ± 2 |

[a]From Cook et al. (1998).

The $^{14}C$ activities of all samples are enhanced above ambient background, while the Hartlepool samples are all more highly enriched than those from either Torness or Whitby. The questions raised by these results are the following:

- What is the source of the enhancements above ambient background at both Torness and Hartlepool?
- Why is there such a difference between the 2 sites?

Based on low $^{14}C$ activities of samples from northern Scotland, Cook et al. (1998) discounted the influence of Sellafield-derived $^{14}C$ discharges on the activities found at Whitby in NE England. However, it can be observed from Figure 2 that there was a large increase in $^{14}C$ discharges from Sellafield in 1994/95, around the time of their study. Sellafield-derived $^{99}Tc$ has much in common with Sellafield-derived $^{14}C$. Both have been discharged since 1952; they have similar, recent discharge histories (Figure 2), including a peak in discharges during 1994/95; they have long half-lives ($^{99}Tc$ has a half-life of $2.3 \times 10^5$ yr); and they behave in a conservative manner in the marine environment. In particular, $^{99}Tc$ has a low $K_d$ in sediments that are low in organic matter, such as those of the Irish Sea. Recent studies using Sellafield-derived $^{99}Tc$ as a tracer of water movement give transit times of 15–18 months from Sellafield to the east coast of the UK (Leonard et al. 1997; McCubbin et al. 2002). The 1994/95 peak in $^{99}Tc$ discharge activities and the propagation of this peak through UK coastal waters was studied in detail, with results indicating that the leading edge of the peak had traveled around the north coast of Scotland and as far south as latitude 52° north within 9 months (McCubbin et al. 2002). A further study has shown that the leading edge reached Arctic waters between 1998 and 1999 (Kershaw et al. 2004). If we use transport of Sellafield-derived $^{99}Tc$ as a proxy for Sellafield-derived $^{14}C$ transport in UK coastal waters, this implies that the activities found at Torness and Hartlepool in 1998 and 1999 could not be due to the peak in $^{14}C$ discharges from Sellafield in 1994/1995. In addition, if there was any contribution of $^{14}C$ to the activities found at Hartlepool or Torness, then it would be from discharges from Sellafield made approximately 15–18 months prior to the collection of samples for this study.

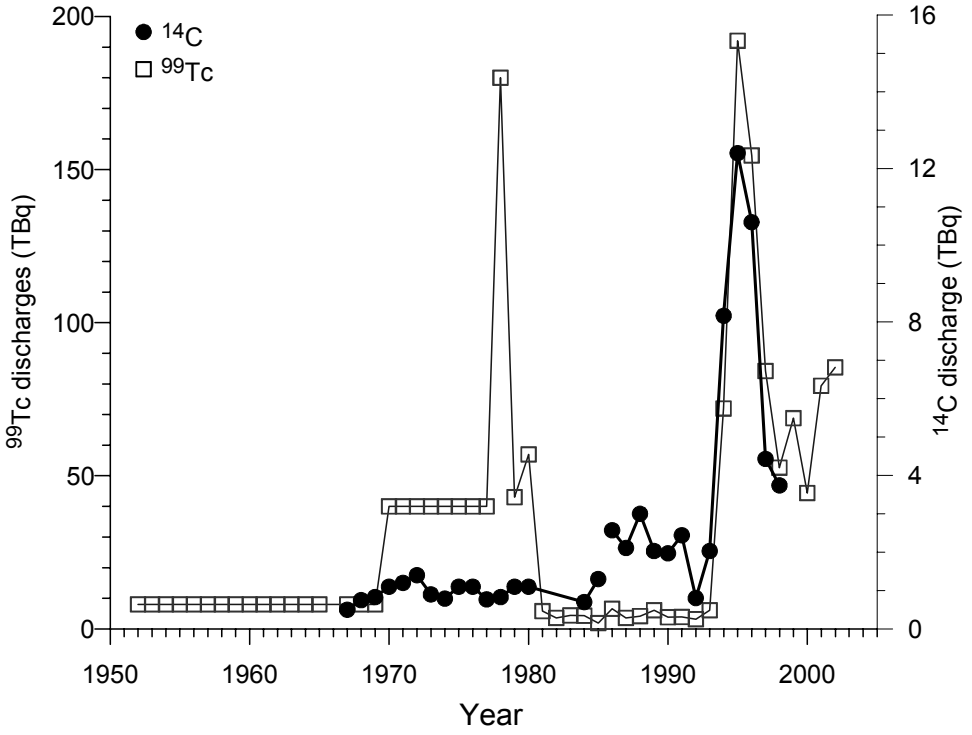


Figure 2 $^{14}$C and $^{99}$Tc discharges from the Sellafield nuclear fuel reprocessing plant (1952–2000)

From Tables 1 and 3, it can be observed that $^{14}$C activities in the vicinity of Hartlepool fall within the range measured at the North Channel. While it is unlikely that transport of $^{14}$C in the DIC would not result in dilution, it is hard to be definitive about this without data from an additional site situated between Sellafield and the NE coast of England. Unfortunately, this study did not investigate $^{14}$C activities in the marine environment from northern Scotland, unlike the study of Cook et al. (1998). However, estimates of DIC $^{14}$C activity at Cape Wrath (Figure 1) can be made using (i) dilution factors between the North Channel and Cape Wrath calculated by McCubbin et al. (2002) for 1993 to 1996 using $^{99}$Tc (see Table 2), and (ii) DIC $^{14}$C activities from the North Channel collected between August 1997 and July 1999 (Table 3). The North Channel DIC $^{14}$C activities reflect discharges from Sellafield made 3–6 months prior to the collection date, and so are representative of Sellafield discharges that would reach Torness and Hartlepool 15–18 months after the date of discharge. Due to variations in current flux and $^{14}$C discharge from Sellafield, there are obvious assumptions in using dilution factors for 1993 to 1996 and $^{14}$C data for 1997 to 1999; however, neither the dilution factors (4.7 to 12.2) nor the DIC $^{14}$C activities at the North Channel (101–271 Bq kg$^{-1}$) vary greatly over this period. Therefore, the estimates (8.2 to 57.7 Bq kg$^{-1}$ C) (Table 2) based on these data are useful in providing the only probable range of DIC $^{14}$C activities for Cape Wrath during this time. The net $^{14}$C activity for the DIC at Torness falls within this range, suggesting that at least part of the enhancement above ambient background may be Sellafield-derived. In contrast, the net activities in the DIC at the Hartlepool site are greater than this range. In view of the dilution that $^{14}$C undergoes during transport and the fact that Torness is significantly closer to Sellafield than Hartlepool, it would seem unlikely that the enhancements above ambient background at Hartlepool are Sellafield-derived, although some contribution from Sellafield cannot be ruled out. As both the Hartlepool and Torness nuclear plants have the same fundamental design, the low activities at the Torness AGR

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imply that the significantly higher activities found at Hartlepool are not from the AGR, although on the evidence available some contribution cannot be ruled out.

Net activities in mussels from Hartlepool were 561 ± 4 Bq kg$^{-1}$ C in 1998 and 913 ± 4 Bq kg$^{-1}$ C in 1999. These activities are significantly higher than those found at the North Channel; indeed, the net mussel activity in 1999 falls within the range of activities found at Nethertown (Table 3), close to Sellafield (Figure 1). Comparison of the Hartlepool data and data from a site very close to Amersham International plc (Cook et al. 1998) indicates certain similarities. The Amersham data had an excess $^{14}C$ ratio of approximately 10:1 for mussels:seaweed, which was interpreted as indicating a significant organic component in the discharges. The enhancements in mussels relative to seaweed at Hartlepool are 11:1 for 1998 and 18:1 for 1999. These ratios are greater than the Amersham ratio, implying that the discharges close to Hartlepool also have an organic component. The activities in mussels are similar in magnitude at the Amersham and Hartlepool sites, implying a similar sized discharge to Amersham (1–2 TBq during this period), close to the Hartlepool sampling site, or indeed, a greater sized discharge at distance. However, the only other authorized nuclear site in the area was a small test reactor that ceased operations in 1996, and subsequent discharges are under the heading of beta/gamma and ranged from $9.69 \times 10^{-8}$ to $9.79 \times 10^{-7}$ TBq between 1996 and 1999 (CEFAS 1997–2000). The magnitude of these discharges is much too small to account for the observed enrichments at Hartlepool. We therefore have no obvious explanation for these anomalously high $^{14}C$ activities.

Table 2 $^{99}Tc$ dilution factors between the North Channel and Cape Wrath (from McCubbin et al. 2002) and estimated $^{14}C$ activities in the DIC fraction of the water column at Cape Wrath.

| Survey date | $^{99}Tc$ dilution factor | Estimated $^{14}C$ activity (Bq kg$^{-1}$ C) |
|---|---|---|
| December 1993 | 12.2 | 5.5–22.2 |
| December 1994 | 8.0 | 8.4–22.9 |
| December 1995 | 9.1 | 7.4–29.8 |
| December 1996 | 4.7 | 14.3–57.7 |

## CONCLUSIONS

There are $^{14}C$ enrichments in biota around the Hartlepool and Torness AGRs. Both have the same fundamental design, and so would be expected to have the same $^{14}C$ formation and discharge pathways; however, the activities around Hartlepool are significantly greater.

Hartlepool is significantly further away from Sellafield than Torness, implying that the activities are not Sellafield-derived.

Using dilution factors for Sellafield-derived $^{99}Tc$ between the North Channel and Cape Wrath and $^{14}C$ data from the North Channel, it was possible to calculate likely $^{14}C$ activities at Cape Wrath. These data demonstrated that the $^{14}C$ discharges from Sellafield could be a significant contributor to the net DIC activities found in the vicinity of the Torness AGR but are not responsible for the net DIC activities found in the vicinity of the Hartlepool AGR.

The activities in mussels were significantly greater than those in the DIC and seaweed, implying the possibility of an organic $^{14}C$ discharge in the vicinity of Hartlepool; however, the only other authorized discharge in the area appears too small to account for the activities.

Table 3 Net $^{14}C$ activities (i.e. above ambient background) in mussels (*Mytilus edulis*) and the DIC fraction of the water column (Bq kg$^{-1}$ C ± 1 σ) collected at Nethertown and the North Channel.

| | Nethertown | | North Channel | |
|---|---|---|---|---|
| Collection date | Mussels | DIC | Mussels | DIC |
| Aug. 97 | 813 ± 4 | 440 ± 5 | 120 ± 4 | 120 ± 4 |
| Sept. 97 | 956 ± 7 | 1012 ± 7 | 116 ± 4 | 117 ± 4 |
| Oct. 97 | 1045 ± 4 | 2779 ± 9 | 111 ± 4 | 116 ± 4 |
| Nov. 97 | 1104 ± 4 | 1335 ± 7 | 118 ± 4 | 111 ± 4 |
| Dec. 97 | 1188 ± 5 | 3283 ± 9 | 111 ± 4 | 171 ± 4 |
| Jan. 98 | 1197 ± 6 | 2956 ± 10 | 101 ± 4 | 271 ± 5 |
| Feb. 98 | 1210 ± 6 | 3208 ± 15 | 117 ± 4 | 164 ± 4 |
| Mar. 98 | 1216 ± 5 | 8057 ± 25 | 110 ± 5 | 150 ± 4 |
| Apr. 98 | 1300 ± 5 | 2379 ± 9 | 116 ± 4 | 109 ± 4 |
| May 98 | 1983 ± 8 | 3212 ± 11 | 108 ± 3 | 123 ± 4 |
| June 98 | 1918 ± 8 | 5807 ± 27 | 115 ± 4 | 130 ± 6 |
| July 98 | 1827 ± 8 | 4656 ± 15 | 108 ± 4 | 136 ± 5 |
| Aug. 98 | 1812 ± 7 | 998 ± 5 | 106 ± 4 | 126 ± 4 |
| Sept. 98 | 1596 ± 6 | 598 ± 5 | 105 ± 4 | 122 ± 4 |
| Oct. 98 | 1569 ± 9 | 344 ± 4 | 109 ± 4 | 101 ± 6 |
| Nov. 98 | 1555 ± 7 | 1453 ± 7 | 102 ± 4 | 124 ± 4 |
| Dec. 98 | 1526 ± 10 | 520 ± 4 | 102 ± 4 | 136 ± 4 |
| Jan. 99 | 1444 ± 7 | 1400 ± 7 | 106 ± 4 | 115 ± 4 |

It is also unlikely that the Hartlepool AGR is the source of organic $^{14}C$ as there is no evidence of an organic $^{14}C$ discharge in the vicinity of Torness and there is no known pathway for a significant production/discharge of organic $^{14}C$ from these reactors.

A further and more extensive investigation into $^{14}C$ distributions in the marine environment around Hartlepool is required to determine the location, discharge activities, and impact of the anthropogenic source of $^{14}C$ in this area.

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