

## TRANSPORT OF SELLAFIELD-DERIVED $^{14}\text{C}$ FROM THE IRISH SEA THROUGH THE NORTH CHANNEL

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**ABSTRACT.** Since the early 1950s, the Sellafield nuclear fuel reprocessing plant in Northwest England has released radio-carbon into the Irish Sea in a mainly inorganic form as part of its authorized liquid effluent discharge. In contrast to the trend in which the activities of most radionuclides in the Sellafield liquid effluent have decreased substantially,  $^{14}\text{C}$  discharges have increased since 1994–95. This has largely been due to a policy change favoring marine discharges over atmospheric discharges.  $^{14}\text{C}$  is radiologically important due to its long half life, mobility in the environment, and propensity for entering the food chain. Current models for radionuclide dispersal in the Irish Sea are based on a reversible equilibrium distribution coefficient ( $k_d$ ), an approach which has been shown to be inadequate for  $^{14}\text{C}$ . Development of predictive models for the fate of Sellafield-derived  $^{14}\text{C}$  requires a thorough understanding of the biogeochemical fluxes between different carbon reservoirs and the processes controlling the net flux of  $^{14}\text{C}$  out of the Irish Sea, through the North Channel. In this study, both an empirical and a halving time approach indicate that close to 100% of the  $^{14}\text{C}$  that is discharged from Sellafield is dispersed beyond the Irish Sea on a time-scale of months in the form of DIC, with little transfer to the PIC, POC, and DOC fractions, indicating that the “dilute and disperse” mechanism is operating satisfactorily. This is consistent with previous research that indicated little transfer of  $^{14}\text{C}$  to Irish Sea sediments. While significant  $^{14}\text{C}$  enhancements have been observed in the biota of the Irish Sea, this observation is not necessarily in conflict with either of the above as the total biomass has to be taken into account in any calculations of  $^{14}\text{C}$  retention within the Irish Sea.

### INTRODUCTION

Radiocarbon is produced in the structural materials, moderator, fuel and coolant of nuclear reactors via a number of activation reactions but principally by the  $^{14}\text{N}(\text{n},\text{p})^{14}\text{C}$  reaction (Kelly et al. 1975; Hayes and MacMurdo 1977; Bush et al. 1983; Otlet et al. 1992). The timing of release of this nuclear fuel cycle-derived  $^{14}\text{C}$  to the environment depends upon the location of production within the reactor. The  $^{14}\text{C}$  produced in the structural materials is sequestered there until reactor decommissioning, while the  $^{14}\text{C}$  in the fuel, moderator and coolant systems is released to the environment during normal reactor operations and fuel reprocessing. Global atmospheric releases of  $^{14}\text{C}$  by the nuclear industry to 1989 were estimated to be 1383 TBq (UNSCEAR 1993) which represents <1% of the total  $^{14}\text{C}$  activity produced by nuclear weapons testing. In contrast, Begg (1992), using installed power capacity data (IAEA 1991), estimated the potential annual production of  $^{14}\text{C}$  by the nuclear industry to be approximately 1470 TBq which is similar to the level of natural production (approximately 920 TBq per annum) estimated by Bush et al. (1983).

The radiological importance of  $^{14}\text{C}$  derives from its long half-life (5730 yr), mobility in the environment and propensity for entering the food chain which result in a high collective effective dose. Although the  $^{14}\text{C}$  that is discharged from nuclear fuel cycle activities represents a small proportion of the total activity released, it contributes 56% of the collective effective dose from all radionuclides discharged from reactors worldwide (UNSCEAR 1993).

The Sellafield nuclear fuel reprocessing plant (Figure 1) has discharged radionuclides into the Irish Sea, under authorization, since 1952 (Gray et al. 1995) and  $^{14}\text{C}$  has been a component of this low-level liquid radioactive waste. Sellafield's low-level aquatic discharge records indicate that the activities of most radionuclides released to the environment reached a maximum in the mid to late 1970s and have decreased substantially since then as new “clean-up” technologies such as the enhanced actinide removal plant (EARP) and the site ion-exchange plant (SIXEP) began operations. SIXEP is

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Figure 1 Locations of sampling sites (s) and Sellafield nuclear fuel reprocessing plant (●)

highly efficient at removal of cationic species such as  $^{137}\text{Cs}^+$  from the waste stream, however, its efficacy for removal of anionic species such as  $^{14}\text{CO}_3^{2-}$  and  $^{99}\text{TcO}_4^-$  is poor. Figure 2 illustrates the difference in the discharge histories between  $^{14}\text{C}$  and  $^{137}\text{Cs}$ . The recent increases in  $^{14}\text{C}$  releases (since 1994) are due largely to a change in policy from mainly atmospheric discharges to mainly marine discharges and to an increase in fuel through-put as THORP (thermal oxide reprocessing plant) began operations.

The behavior of Sellafield derived radionuclides such as  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$  and  $^{239,240}\text{Pu}$  in the Irish Sea, has been extensively studied (Hetherington and Jefferies 1974; Jefferies et al. 1982; Pentreath et al. 1984; MacKenzie et al. 1987; Cook et al. 1997; Hunt et al. 1998; MacKenzie 2000) and in many cases their behavior can be adequately described using a reversible sorption ( $K_d$ ) approach to aqueous phase-solid phase partitioning. For example, during the 1970s approximately 10% of the discharged  $^{137}\text{Cs}$  was reversibly sorbed onto the sediments of the Irish Sea (Hetherington 1975; Pentreath et al. 1984) but now that the discharge activities have fallen, it is undergoing re-dissolution

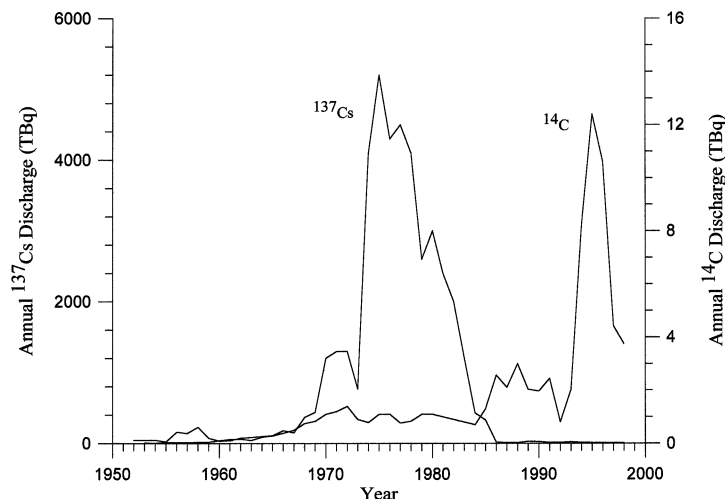


Figure 2 Annual  $^{14}\text{C}$  and  $^{137}\text{Cs}$  discharges from Sellafield

back into the overlying water column, with approximately 70–75% lost from surface sediments by 1992 (MacKenzie et al. 1998).

Recently, a significant research effort has focused on the behavior of  $^{14}\text{C}$  in the Irish Sea (Begg et al. 1991, 1992; Cook et al. 1995, 1998; Wolstenholme et al. 1998). The  $^{14}\text{C}$  in Sellafield liquid effluent is in an inorganic form and has been shown to have an immediate impact on the activity of the dissolved inorganic carbon (DIC) fraction of the seawater (Begg et al. 1991).  $^{14}\text{C}$  enhancements above ambient background have also been demonstrated in the biota and sediments of the Northeast Irish Sea (Begg et al. 1992; Cook et al. 1995; Wolstenholme et al. 1998), but the rates and mechanisms of the transfer processes involved are not well understood and the relatively simple  $K_d$  approach applied to species such as  $^{137}\text{Cs}$  and  $^{241}\text{Am}$  cannot be applied to  $^{14}\text{C}$  because, as noted by the IAEA in 1985:

(the)... problem of defining  $K_d$  and concentration factor for  $^{14}\text{C}$  (is that)... the total concentration of...C includes dissolved organic carbon, carbonate, bicarbonate and  $\text{CO}_2$ . There are insufficient data to indicate what fraction, if any, would not equilibrate with  $^{14}\text{C}$ , and to what extent any of the forms of C, would or would not become biologically incorporated.

To determine the fate of Sellafield-derived  $^{14}\text{C}$ , some knowledge of the quantity of  $^{14}\text{C}$  removed from the Irish Sea, as well as that retained in the area in the different carbon reservoirs, is required. Begg (1992) concluded that  $^{14}\text{C}$  behaves in a mainly conservative manner in the DIC fraction of seawater with some evidence of transfer of  $^{14}\text{C}$  from the DIC into the other biogeochemical fractions of the water column (particulate inorganic carbon or PIC, particulate organic carbon or POC, and dissolved organic carbon or DOC) although there was no systematic trend between the activity of the DIC in the water column and that of the other fractions. This was supported by Wolstenholme (1999) who estimated that of the total  $^{14}\text{C}$  discharged between 1952 and 1995, only 0.08% had been incorporated into the organic carbon fraction of the sediments of the Northeast Irish Sea and between 0.03 and 0.9% had been incorporated into the inorganic fraction.

To model the behavior of  $^{14}\text{C}$  in the Irish Sea, the physical properties of the system, such as water depth, current movements etc., must be known and indeed, this is a well studied area of coastal

water. The Northeast Irish Sea is a shallow basin with an average water depth of 30–50 m, large tidal ranges and a small residual northwards current. Removal of water and associated radionuclides from the Irish Sea occurs via these small residual currents through the North Channel (Ramster and Hill 1969; Howarth 1982). The water flux through the North Channel is highly variable and previous studies have shown a range of fluxes varying between 1.72 and 7.76 km<sup>3</sup> day<sup>-1</sup> (Bowden 1950; McKay and Baxter 1985; Jefferies et al 1982; Howarth 1982; Brown and Gmitrowicz 1995; Knight and Howarth 1999; Young et al. 2000). There is also a counter current running in from the Atlantic Ocean along the Irish Coast although it is small in comparison to the total flux (Brown and Gmitrowicz 1995). Furthermore, there is evidence of periodic changes in the direction of water movement resulting in an influx of high salinity water from the Atlantic (Howarth 1982).

To estimate the removal of <sup>14</sup>C from the Irish Sea in the form of DIC, DOC, PIC, and POC, a two-year study of these activities in the water column was carried out. In addition, dissolved <sup>137</sup>Cs activities were also studied during this period as an indicator of the behavior of a further radionuclide with a conservative behavior, however, it should be noted that the discharge history of <sup>137</sup>Cs is quite different from that of <sup>14</sup>C (Figure 2).

## MATERIALS AND METHODS

Portpatrick, on the east coast of the North Channel (Figure 1), was chosen as the sampling site for this study. Near-shore water was collected into 10 liter polyethylene screw-cap containers that had been thoroughly cleaned prior to use. On sampling, the containers were rinsed three times with seawater before being completely filled, leaving no head-space.

Water samples were collected on a monthly basis from August 1997 until July 1999. One hundred liters were collected each month except November 1997, February 1998, May 1998, August 1998, November 1998 and February 1999 when 200 liters were collected for additional analysis of the PIC, POC, and DOC fractions.

The usual definition of particulate matter in seawater is material that is retained on a 0.22 µm filter. To avoid carbon contamination, glass fiber filters had to be employed in this study and these are not available at this pore size. Therefore, we have used 0.7 µm glass fiber filters (Whatman GF/F) which are the smallest pore size available. Upon returning to the laboratory, the samples were filtered through pre-combusted glass fiber filters and approximately 80 liters of filtered water were then analyzed for DIC by addition of 1 liter of 4M HCl. The evolved CO<sub>2</sub> was cryogenically trapped and converted to benzene for liquid scintillation counting via lithium carbide and acetylene syntheses (Barker 1953).

For the analysis of the PIC fraction, 4M HCl was added to a reaction vessel containing a filter paper holding the particulate material. This quickly hydrolyzed the PIC to CO<sub>2</sub> which was cryogenically trapped and reduced to graphite for AMS analysis according to the method of Slota et al. (1987). For POC analysis, the filter paper was then rinsed with reverse osmosis water to remove the residual HCl, dried and transferred to a quartz combustion tube containing copper oxide and silver wire. This was evacuated, flame sealed and placed in a furnace at 850 °C overnight. The following day, the combustion tube was opened under vacuum, the CO<sub>2</sub> was cryogenically trapped and converted to graphite (Slota et al. 1987).

The DOC was extracted from 500 mL of filtered seawater using a freeze-drying method developed during the course of this study and very similar to that developed by Burr et al. (2001). When the sample was completely dry, oxygen gas was added under vacuum and the flask was flame sealed.

The sample was then combusted overnight at 500 °C. The resulting gases were separated under vacuum using a pentane trap and transferred to a secondary combustion tube containing 0.5 g copper oxide and 0.1 g manganese oxide (Fry et al. 1996) to remove any remaining HCl and oxides of sulphur. The secondary combustion tube was flame sealed and then combusted at 500 °C for two hours. The resultant  $\text{CO}_2$  was extracted from the secondary combustion tube and reduced to graphite (Slota et al. 1987). All graphite samples were measured at the NSF-University of Arizona Accelerator Mass Spectrometry (AMS) Facility.

Dissolved  $^{137}\text{Cs}$  was extracted from 20 liters of seawater from which the DIC had been removed using a KCFC cartridge according to the method of MacKenzie et al. (1979). The  $^{137}\text{Cs}$  activity on the KCFC cartridge was then determined by gamma spectroscopy using an N-type intrinsic germanium detector.

## RESULTS

Results for 1)  $^{14}\text{C}$  activities above ambient background in the DIC, PIC, DOC, and POC fractions in seawater at Portpatrick, and 2) dissolved  $^{137}\text{Cs}$  activities in seawater at Portpatrick are presented in Table 1. Table 2 presents background biota and DIC data from Burtonport on the west coast of Ireland. This site is remote from Sellafield and is mainly influenced by Atlantic Ocean water. The 1995 values from Burtonport are from Cook et al. (1998).

## DISCUSSION

While the published values for the flux of water through the North Channel are variable, a value of  $5 \text{ km}^3 \text{ day}^{-1}$  has been used successfully in previous studies of radionuclide transport from the Irish Sea through the North Channel (Pentreath et al. 1984; Cook et al. 1997) and was adopted for calculations within the present study.

During the 2 year duration of this study, the total discharges of  $^{137}\text{Cs}$  were 15.6 TBq. Using an average  $^{137}\text{Cs}$  activity of  $36.3 \pm 2.0 \text{ mBq L}^{-1}$ , the total activity lost through the North Channel in the dissolved phase over the two year study period was  $133 \pm 7 \text{ TBq}$ , well in excess of the total discharge and consistent with continuing re-dissolution from the underlying sediment, in agreement with previous observations (Hunt and Kershaw 1990; McCartney et al. 1994; Cook et al. 1997).

Ambient  $^{14}\text{C}$  background activity from natural production and nuclear weapons testing fallout will differ between the four biogeochemical fractions and must be subtracted from the measured activities to obtain  $^{14}\text{C}$  activities due to the Sellafield discharges. As the main source term for Irish Sea water is the North Atlantic (Bowden 1950), the DIC background was estimated from measurements on near-shore water samples and seaweed from Burtonport, on the west coast of Ireland (Figure 1). Burtonport is a suitable sampling site as the water exiting the Irish Sea, through the North Channel, is tightly entrained along the west coast of Scotland and studies have shown that there is negligible activity from Sellafield discharges to the west of Malin Head (McKinley et al. 1981; Jefferies et al. 1973). The seaweeds are primary producers that use DIC directly and therefore produce an integrated  $^{14}\text{C}$  value. The weighted mean "background" value for 1997–99 was  $251 \pm 1 \text{ Bq kg}^{-1} \text{ C}$  (Table 2). The ambient background values for the other three biogeochemical fractions of the water column are more difficult to estimate as they are more dependent on local variables such as contributions from terrestrial run-off, sediment resuspension, level of primary production, etc., so  $^{14}\text{C}$  activities for the PIC, POC, and DOC fractions in Atlantic waters are not necessarily applicable to the Irish Sea. We concluded that the best available estimates for Irish Sea PIC, POC, and DOC backgrounds were from those samples collected in 1989 at the St. George's Channel and the North Chan-

Table 1  $^{14}\text{C}$  activities ( $\text{mBq L}^{-1}$ ) above ambient background in the DIC, DOC, PIC, and POC phases and dissolved  $^{137}\text{Cs}$  activities ( $\text{mBq L}^{-1}$ ) in seawater at Portpatrick

| Date          | DIC             | DOC  | PIC                                     | POC                                   | $^{137}\text{Cs}$ |
|---------------|-----------------|------|-----------------------------------------|---------------------------------------|-------------------|
| August 1997   | $2.60 \pm 0.06$ |      |                                         |                                       | $47.9 \pm 1$      |
| Sept. 1997    | $2.76 \pm 0.06$ |      |                                         |                                       | $43.5 \pm 2$      |
| Oct. 1997     | $3.09 \pm 0.07$ |      |                                         |                                       | $45.9 \pm 2$      |
| Nov. 1997     | $2.64 \pm 0.06$ |      | $4.3\text{E}^{-3} \pm 9.0\text{E}^{-4}$ | $1.5\text{E}^{-2} \pm 8\text{E}^{-4}$ | $44.4 \pm 2$      |
| Dec. 1997     | $4.43 \pm 0.07$ |      |                                         |                                       | $52.7 \pm 2$      |
| Jan. 1998     | $6.12 \pm 0.07$ |      |                                         |                                       | $50.2 \pm 2$      |
| Feb. 1998     | $4.37 \pm 0.08$ | 0.38 | $6\text{E}^{-4} \pm 4.8\text{E}^{-4}$   | $1.2\text{E}^{-2} \pm 7\text{E}^{-4}$ | $37.3 \pm 2$      |
| March 1998    | $3.66 \pm 0.06$ |      |                                         |                                       | $36.3 \pm 2$      |
| April 1998    | $2.73 \pm 0.06$ |      |                                         |                                       | $27.7 \pm 2$      |
| May 1998      | $2.91 \pm 0.06$ | 0.12 | $9\text{E}^{-4} \pm 3.3\text{E}^{-4}$   | $2.4\text{E}^{-2} \pm 8\text{E}^{-4}$ | $28.5 \pm 3$      |
| June 1998     | $3.30 \pm 0.12$ |      |                                         |                                       | $27.3 \pm 2$      |
| July 1998     | $3.17 \pm 0.10$ |      |                                         |                                       | $30.4 \pm 2$      |
| August 1998   | $3.31 \pm 0.05$ | 0.04 | $1.9\text{E}^{-3} \pm 5.0\text{E}^{-4}$ | $1.9\text{E}^{-2} \pm 7\text{E}^{-4}$ | $32.6 \pm 1$      |
| Sept. 1998    | $2.96 \pm 0.06$ |      |                                         |                                       | $38.9 \pm 1$      |
| Oct. 1998     | $2.62 \pm 0.13$ |      |                                         |                                       | $30.5 \pm 1$      |
| Nov. 1998     | $3.25 \pm 0.05$ |      | $2.2\text{E}^{-3} \pm 3.1\text{E}^{-4}$ | $8.2\text{E}^{-3} \pm 4\text{E}^{-4}$ | $33.8 \pm 1$      |
| Dec. 1998     | $3.60 \pm 0.05$ |      |                                         |                                       | $50.6 \pm 1$      |
| Jan. 1999     | $3.05 \pm 0.08$ |      |                                         |                                       | $46.8 \pm 1$      |
| Feb. 1999     | $2.16 \pm 0.05$ |      | $3.7\text{E}^{-3} \pm 1.3\text{E}^{-3}$ | $2.9\text{E}^{-2} \pm 1\text{E}^{-3}$ | $35.7 \pm 2$      |
| March 1999    | $2.16 \pm 0.06$ |      |                                         |                                       | $32.9 \pm 1$      |
| April 1999    | $2.10 \pm 0.07$ |      |                                         |                                       | $21.5 \pm 1$      |
| May 1999      | $3.81 \pm 0.08$ |      |                                         |                                       | $19.8 \pm 2$      |
| June 1999     | $1.40 \pm 0.06$ |      |                                         |                                       | $20.7 \pm 1$      |
| July 1999     | $2.72 \pm 0.11$ |      |                                         |                                       | N/A <sup>a</sup>  |
| Mean $\pm$ SE | $3.12 \pm 0.19$ | 0.18 | $2.3\text{E}^{-3} \pm 5.5\text{E}^{-4}$ | $1.8\text{E}^{-2} \pm 3\text{E}^{-3}$ | $36.3 \pm 2$      |

<sup>a</sup>N/A = not analyzedTable 2 Background  $^{14}\text{C}$  activities ( $\text{Bq kg}^{-1}\text{ C}$ ) in the DIC and an assortment of biota at Burtonport on the west coast of Ireland

| Sample date | DIC         | Mussel<br>( <i>mytilus edulis</i> ) | Seaweed<br>( <i>fucus sp.</i> ) | Crab<br>( <i>cancer pagurus</i> ) | Roundfish<br>(various) | Flatfish<br>(various) |
|-------------|-------------|-------------------------------------|---------------------------------|-----------------------------------|------------------------|-----------------------|
| 1995        | $247 \pm 3$ | $245 \pm 2$                         | $248 \pm 2$                     | $263 \pm 2$                       | $252 \pm 3$            | $265 \pm 2$           |
| 1997        | $254 \pm 4$ | $249 \pm 3$                         | $252 \pm 3$                     | $263 \pm 3$                       | $265 \pm 2$            | $248 \pm 2$           |
| 1998        | $252 \pm 2$ | $247 \pm 3$                         | $254 \pm 2$                     | $279 \pm 3$                       | $254 \pm 3$            | $287 \pm 3$           |
| 1999        | $249 \pm 2$ | $245 \pm 1$                         | $250 \pm 1$                     | $270 \pm 2$                       | $250 \pm 2$            | $244 \pm 1$           |

nel and analyzed by Begg (1992). The values used for PIC, POC and DOC background activities were  $144 \pm 10$ ,  $125 \pm 6$ , and  $176 \pm 4\text{ Bq kg}^{-1}\text{ C}$ , respectively. Some Sellafield  $^{14}\text{C}$  activity would have been incorporated into these samples but we estimate that it was negligible as  $^{14}\text{C}$  discharges at that time were much lower than current discharges. For this reason, the calculated excess  $^{14}\text{C}$  values contributed by Sellafield will be a minimum. It should be noted that the calculation of activity removed from the Irish Sea in the DOC fraction is based on only 3 analyses and, as yet, the method

employed is still under development and not fully validated. Therefore, the value determined for this estimated removal should be treated with some caution.

Using the average  $^{14}\text{C}$  excesses and a water flux of  $5 \text{ km}^3 \text{ d}^{-1}$ , this gives an estimated removal during the 2-year period of  $11.4 \pm 0.7 \text{ TBq}$  in the DIC fraction,  $0.07 \text{ TBq}$  in the POC fraction,  $<0.01 \text{ TBq}$  in the PIC fraction and approximately  $0.66 \text{ TBq}$  in the DOC fraction. This compares with total discharges over the study period of  $11.3 \text{ TBq}$ . However, this does not take into account the standing crop of  $^{14}\text{C}$  present at the beginning of this study. To encompass this, we also compared the trends in  $^{14}\text{C}$  activity at Portpatrick with the monthly  $^{14}\text{C}$  discharge data from 1992 onwards. Initially, this approach assumed a number of halving times for the removal of  $^{14}\text{C}$  in the form of DIC from the Irish Sea and assumed that transfer of  $^{14}\text{C}$  from the DIC into other carbon fractions was negligible in comparison with removal in the DIC phase. Using this approach, a one month halving time for removal of  $^{14}\text{C}$  from the Irish Sea gave the best fit to the Portpatrick DIC activities (Figure 3), consistent with rapid removal of  $^{14}\text{C}$  in the DIC. This approach gives an estimated removal of  $11.1 \text{ TBq}$  over the study period.

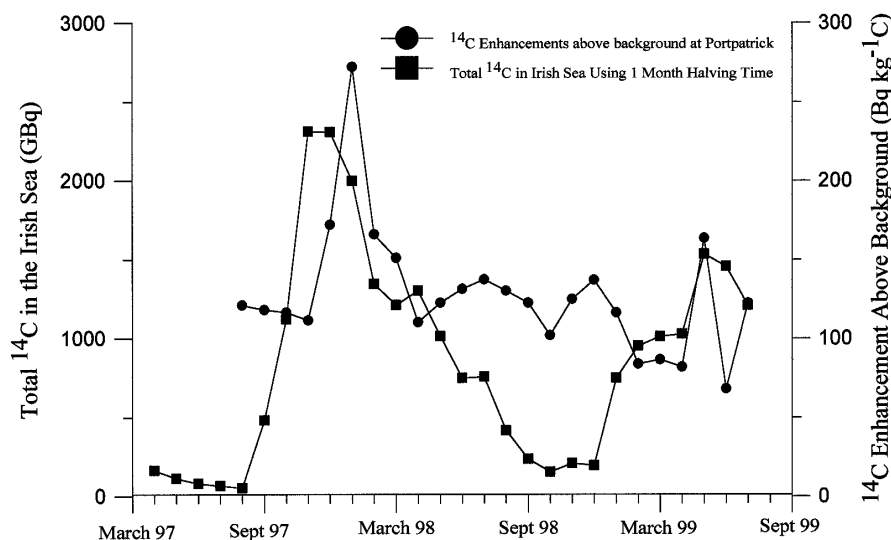


Figure 3  $^{14}\text{C}$  enhancements above ambient background in DIC at Portpatrick and total  $^{14}\text{C}$  in the Irish Sea. Based on Sellafield discharges with a one month halving time for residence in the Irish Sea.

## CONCLUSIONS

Both an empirical and a theoretical halving time approach indicate that close to 100% of the  $^{14}\text{C}$  that is discharged from Sellafield is dispersed beyond the Irish Sea on a time-scale of months, in the form of DIC, indicating that “dilute and disperse” theory is operating satisfactorily. This is consistent with the results of a limited study by Wolstenholme (1999) that indicated little transfer of  $^{14}\text{C}$  to the sediment. While significant  $^{14}\text{C}$  enhancements have been observed in the biota of the Irish Sea, this observation is not necessarily in conflict with either of the above as the total biomass has to be taken into account in any calculations of  $^{14}\text{C}$  retention. While previous calculations of the dose from  $^{14}\text{C}$  to the Sellafield seafood-consuming critical group indicate that the dose is very small ( $<4 \mu\text{Sv}$ )

(Cook et al. 1998), the effective operation of “dilute and disperse” theory raises the issue of whether  $^{14}\text{C}$  enhancements in biota could be limited by careful selection of when  $^{14}\text{C}$  discharges are made relative to the degree of primary production occurring in the water column.

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