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SPATIAL AND TEMPORAL IMPACTS OF ^{14}C RELEASES FROM THE SELLAFIELD NUCLEAR COMPLEX ON THE IRISH COASTLINE

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ABSTRACT. The Sellafield nuclear fuel reprocessing plant is estimated to be the largest single source of global anthropogenic radiocarbon discharge. This study addresses the impact of these releases on the Irish coastal marine environment. Spatial trends in the ^{14}C content of seaweed (*Fucus* spp.) were assessed by collecting and analyzing samples from well-distributed locations around the Irish coastline. Temporal trends were studied by comparing ^{14}C concentrations in present-day samples with levels found in archive material collected at the same locations during research campaigns conducted in the mid-1980s and mid-1990s. The impact of ^{14}C discharged from Sellafield was found to be most apparent in seaweeds from the northeastern Irish coast. This indicates that the pattern of residual currents and, in particular, the south to north transfer of water known to predominate in the Irish Sea, largely controls the spatial distribution of ^{14}C releases. Maximum ^{14}C discharge levels to the marine environment from Sellafield (between 12 and 13 TBq yr^{-1}) were mirrored by peak concentrations found in seaweed from the mid-1990s and in present-day samples (highest recorded value of 130.4 pMC). Concentrations of ^{14}C in seaweed from the west coast of Ireland correspond closely with values measured for seaweeds from the Atlantic coast of northwest Spain and do not appear to be significantly affected by Sellafield discharges.

INTRODUCTION

The radiological importance of radiocarbon derives from its long half-life (5730 yr), mobility in the environment, and propensity for entering the food chain. In a global context, the collective dose from world-wide discharges of ^{14}C, integrated over 10,000 yr, comprises about 80% of the collective dose from the complete nuclear fuel cycle (UNSCEAR 2001; Isogai et al. 2002). Data concerning environmental concentrations of ^{14}C are, however, limited, and it is important that ^{14}C be taken into account in estimating the future collective dose commitment to the Irish population, given the proximity of the Sellafield reprocessing plant in the UK.

Sellafield

The Sellafield complex is located on the Cumbrian coast of northwest England, about 120 km from the northeast coast of Ireland, and is operated by British Nuclear Fuels plc. The complex has been identified by UNSCEAR (1993) as the largest single contributor of ^{14}C to the global environment and the vast majority of the ^{14}C discharges from the site result from nuclear fuel reprocessing operations.

Low-level liquid radioactive waste containing ^{14}C has been discharged from Sellafield into the northeastern Irish Sea since operations began in the early 1950s. A decision in the early 1990s to divert ^{14}C waste from the aerial to the liquid effluent stream resulted in an increase in ^{14}C discharges to the marine environment, which is particularly evident in the peak levels of 12.4 and 13 TBq discharged in 1995 and 2002, respectively (BNFL 1995; MAFF 1996; BNFL 2001; FSA/SEPA 2003). Discharges are mainly in inorganic form and are released as aqueous effluent. In this paper, we examine the relationship between the historical record of ^{14}C in liquid discharges and the ^{14}C content of *Fucus* spp. samples from the Irish coastline.

Seaweed as a Bioindicator

Seaweeds of the genus *Fucus* spp. have proved to be extremely useful and practical as bioindicators for monitoring radioactive discharges from nuclear power plants. The advantages of using *Fucus*

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spp. are discussed by O'Donnell (1997) and can be summarized as follows: a) sensitivity is greatly increased because of the higher concentrations of radionuclides in the algae relative to the surrounding waters; b) *Fucus* spp. is found in great masses in shallow waters along most marine and brackish water coasts at the latitudes we are investigating; and c) seaweeds are much easier to collect, treat, and store for long periods in comparison to water samples.

A large percentage of the ^{14}C content of aqueous discharge effluent from Sellafield is in the form of carbonate/bicarbonate, and so is immediately incorporated into the dissolved inorganic carbon (DIC) fraction of the water column. Seaweeds, as primary producers, utilize the DIC during photosynthesis and, thus, record a time-integrated ^{14}C value (Cook et al. 1998).

The relevance of using *Fucus* spp. as a bioindicator for monitoring ^{14}C discharges from Sellafield is clearly demonstrated by comparing the measured ^{14}C concentration in seaweed from the Sellafield site with the level of ^{14}C in annual liquid discharges (Figure 1). The superimposed trends are clearly correlated and indicate that the concentrations of ^{14}C in *Fucus* spp. reflect the levels of ^{14}C discharged to the Irish Sea (with a short time lag).

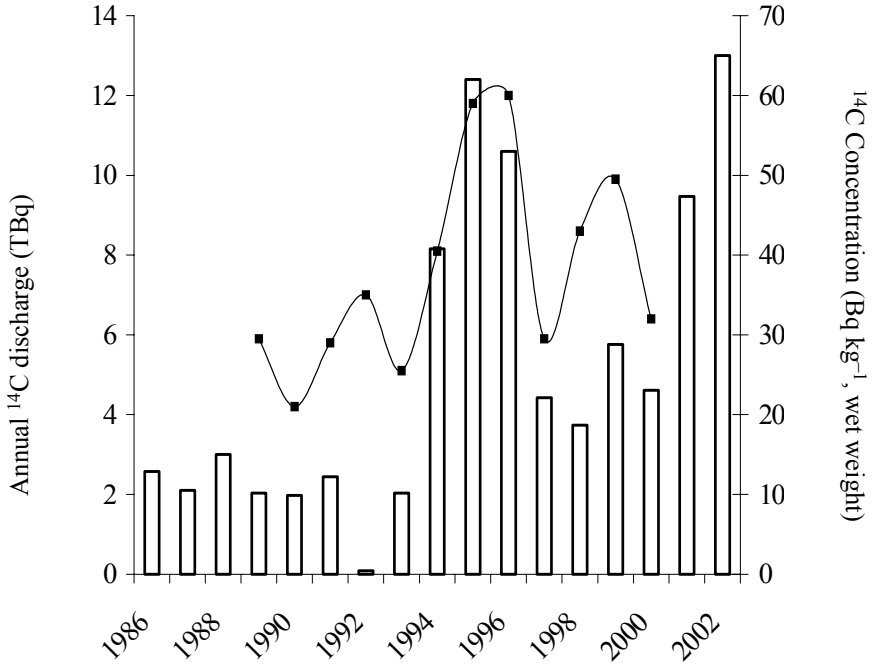


Figure 1 Bar chart showing liquid ^{14}C discharges from Sellafield (Source: FSA/SEPA 2003) and curve showing concentrations of ^{14}C in *Fucus* spp. from the Sellafield shoreline (Source: DEFRA 2002).

METHODS AND MATERIALS

Sample Collection

Seaweeds were sampled from locations selected to provide a representative geographical distribution on all aspects of the Irish coast. The sites were chosen after consulting with the Radiological Protection Institute of Ireland (RPII) in order to coincide with regular monitoring stations for other radionuclides such as ^{137}Cs, ^{99}Tc, and ^{131}I. The locations of these sites are shown in Figure 2.

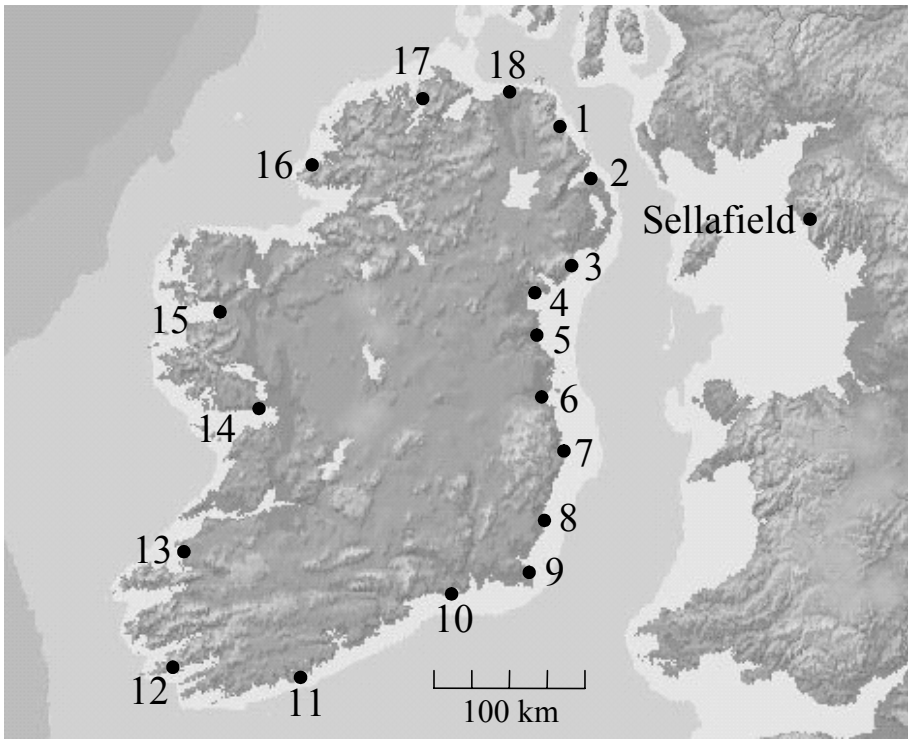


Figure 2 Map of Ireland showing the locations of *Fucus* spp. sampling sites relative to the Sellafield nuclear reprocessing complex in Cumbria, UK.

Seaweed was collected from the intertidal area of each sampling site between November 2002 and August 2003. Archived samples collected between November 1985 and March 1986, and between March 1994 and November 1995 during the course of previous research projects, were sourced from the RPII and the Radiation Physics Research Laboratory of University College Dublin. For convenience, the 3 sets of seaweed samples will hereafter be referred to as relating to 1985, 1994, and 2003, respectively.

Three *Fucus* spp. samples collected near Arosa, Cabo Prior, and Muros (Galician coast, northwest Spain) during 1984 were recovered from our archives. These samples were previously analyzed for plutonium isotopes and showed the ^{238}Pu, ^{239}Pu, and ^{240}Pu concentrations in *Fucus* spp. from this region to be in very good agreement with predicted and reported fallout ratios in mid-latitudes of the Northern Hemisphere (Sanchez-Cabeza 1989).

Fucus spp. samples from both the Irish and Galician coastlines were pretreated and analyzed for ^{14}C as described below.

Sample Analysis

The seaweed was washed thoroughly to remove extraneous material, then oven-dried at 80 °C for 24 hr, ground, and homogenized. Approximately 50 g of each sample was boiled in 1 M HCl for 8 hr to hydrolyze any carbonate and remove surface organic debris. Samples were then rinsed in distilled water and oven-dried again before conversion to benzene via carbon dioxide and acetylene (O'Donnell 1997). The ^{14}C activity of the benzene was measured using low-background liq-

uid scintillation counting (Tri–Carb 2770 TR/SL). The UCD Radiocarbon Age Calculation Program (Gallagher et al. 2002) was used to calculate the ^{14}C concentrations of the seaweeds.

Quality Control

Measurement of the C3 cellulose intercomparison material (IAEA 1998) gave a concentration of 129.8 ± 1.5 (2 σ) percent modern carbon (pMC), which compares very well with the consensus value of 129.41 ± 0.12 (2 σ) pMC and demonstrates the measurement capability of our laboratory. In addition, 8 seaweed samples from the Irish coastline were measured in duplicate to test analytical reproducibility; the resulting data are given in Table 1.

Table 1 Quality assurance data showing values for replicate analyses (#1 and #2) carried out on samples of *Fucus* spp. from selected locations. Quoted uncertainties are ± 2 σ.

		Replicates (pMC)	
Site number	Year	# 1	# 2
3	1985	121.3 ± 1.4	121.7 ± 1.4
3	1994	128.9 ± 1.5	130.4 ± 1.6
4	1994	127.5 ± 1.6	126.6 ± 1.4
4	1985	124.5 ± 1.4	125.8 ± 1.6
5	1985	121.0 ± 1.5	121.4 ± 1.4
12	1994	109.4 ± 1.4	110.6 ± 1.3
15	1985	115.8 ± 1.3	114.2 ± 1.3
16	1985	117.2 ± 1.5	117.2 ± 1.3

RESULTS AND DISCUSSION

Anthropogenic ^{14}C in the Marine Environment

Results are presented as pMC in Table 2. Figure 3a–c reveals enhancements in the pMC value of seaweed taken from the northeast coast of Ireland in 1985, 1994, and 2003 with respect to the pMC value of seaweed taken from the western coast, and indicate that ^{14}C discharged from Sellafield has a measurable impact on Irish marine biota from the north and east coasts. The enhancement along the northeastern coastline appears to reflect the predominantly south to north direction of residual currents of the Irish Sea (Jefferies et al. 1982). These findings concur with previous measurements of ^{14}C enrichments in seawater and biota which revealed a pattern of clockwise water circulation around mainland Britain (Cook et al. 1998).

There is a clear correspondence between the recorded level of ^{14}C in liquid discharges from Sellafield (Figure 4) and the measured concentration of ^{14}C in seaweed from the northeast coast (Figure 3). Peak values of approximately 130 pMC are found in seaweed taken from sites 2 and 3 in 1994 and site 1 in 2003, reflecting the significantly increased discharges from Sellafield in those years relative to 1985.

Ambient Background Levels of ^{14}C

Seaweed from the west coast showed the highest mean concentration of approximately 116 pMC in 1985 with a decrease to 112 pMC in 1994 and a further decrease to 107 pMC in 2003 (Table 3). These measurements concur closely with a previous measurement of ^{14}C activity in seaweed from the west of Ireland in 1995, namely 109.7 pMC (Cook et al. 1998), and show that there has been a decline in ^{14}C activity in North Atlantic surface waters since the mid-1980s.

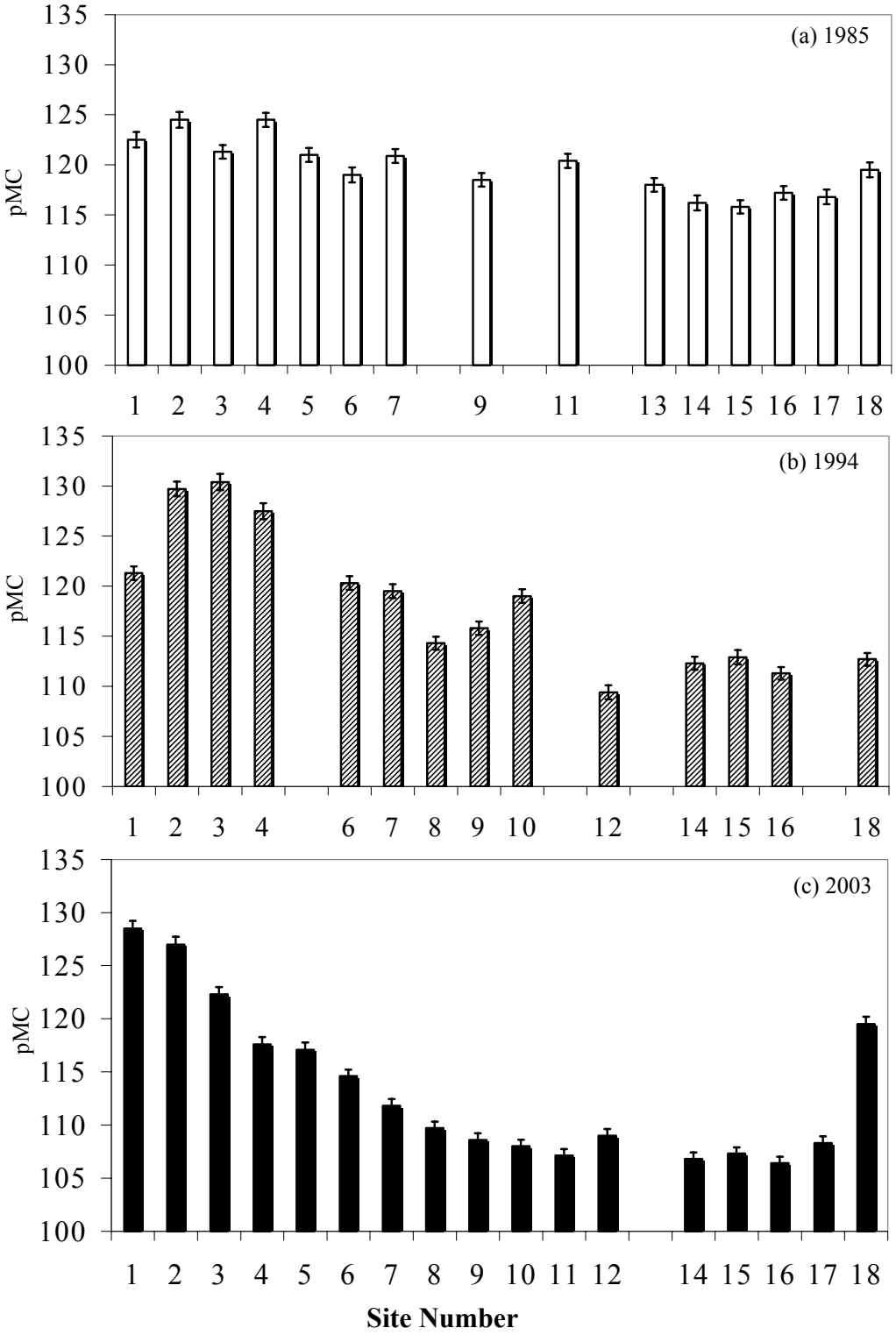


Figure 3 ^{14}C concentrations in *Fucus* spp. from Irish coastal sites taken in (a) 1985, (b) 1994, and (c) 2003. Refer to Figure 2 for locations of site numbers (indicated uncertainties are ±2 σ).

Table 2 ^{14}C concentrations (pMC[a]) in seaweed sampled around the Irish coastline in 1985, 1994, and 2003. Quoted uncertainties are ±2 σ

Site nr	Location	1985	1994	2003
1	Cushendall	122.5 ± 1.5	121.3 ± 1.4	128.5 ± 1.5
2	Bangor	124.5 ± 1.6	129.7 ± 1.5	127.0 ± 1.4
3	Newcastle	121.3 ± 1.4	130.4 ± 1.6	122.3 ± 1.4
4	Carlingford	124.5 ± 1.4	127.5 ± 1.6	117.6 ± 1.3
5	Clogherhead	121.0 ± 1.4	—	117.1 ± 1.3
6	Dublin Bay	119.0 ± 1.5	120.3 ± 1.4	114.6 ± 1.2
7	Wicklow	120.9 ± 1.4	119.5 ± 1.4	111.8 ± 1.3
8	Cahore	—	114.3 ± 1.3	109.7 ± 1.3
9	Rosslare	118.5 ± 1.4	115.8 ± 1.3	108.6 ± 1.2
10	Dunmore East	—	119.0 ± 1.4	108.0 ± 1.2
11	Kinsale	120.4 ± 1.4	—	107.1 ± 1.2
12	Castletownbere	—	109.4 ± 1.4	109.0 ± 1.2
13	Ballyheigue	118.0 ± 1.3	—	—
14	Galway Bay	116.2 ± 1.5	112.3 ± 1.3	106.8 ± 1.2
15	Clew Bay	115.8 ± 1.3	112.9 ± 1.4	107.3 ± 1.2
16	Glen Columcille	117.2 ± 1.3	111.3 ± 1.3	106.4 ± 1.2
17	Lough Swilly	116.8 ± 1.5	—	108.3 ± 1.2
18	Giants Causeway	119.5 ± 1.5	112.7 ± 1.3	119.5 ± 1.4

[a]Note: To convert from pMC to Bq kg^{-1}(carbon) dry weight, multiply by a factor of 2.26.

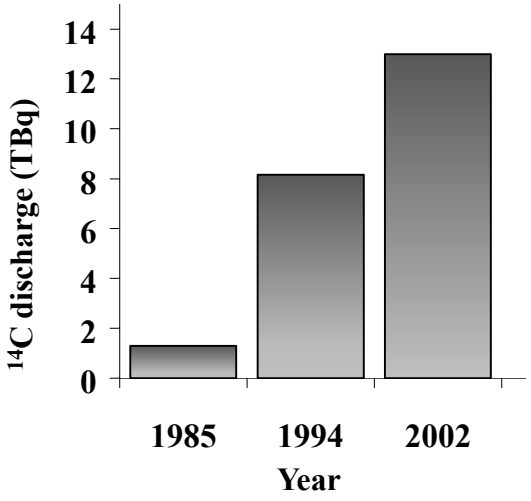


Figure 4 ^{14}C discharged from Sellafield into the Irish Sea during 1985, 1994, and 2002 (UNSCEAR 2001; FSA/SEPA 2003)

It was found that Galician seaweed sampled in 1984 had ^{14}C concentrations that closely correspond with those in *Fucus* spp. sampled along the west coast of Ireland in 1985 (Table 3). This indicates that the ^{14}C activities on the west coast of Ireland are due to a combination of bomb ^{14}C and natural production (ambient background), and accordingly demonstrates that Sellafield discharges have had little impact on the sites west of Ireland.

Table 3 ^{14}C in *Fucus* spp. from the west coast of Ireland and from Galicia. Uncertainties quoted at ±2 σ.

Year	Location (site number)	pMC
1985	West of Ireland (14)	116.2 ± 1.5
	West of Ireland (15)	115.8 ± 1.3
	West of Ireland (16)	117.2 ± 1.3
1994	West of Ireland (14)	112.3 ± 1.3
	West of Ireland (15)	112.9 ± 1.4
	West of Ireland (16)	111.3 ± 1.3
1995	West of Ireland (Burtonport)	109.7 ± 1.4[a]
2003	West of Ireland (14)	106.8 ± 1.2
	West of Ireland (15)	107.3 ± 1.2
	West of Ireland (16)	106.4 ± 1.2
1984	Galicia (Arosa)	116.9 ± 1.3
	Galicia (Cabo Prior)	113.6 ± 1.3
	Galicia (Muros)	115.5 ± 1.3

[a]Converted from 248 ± 2 Bq kg^{-1}(carbon), dry weight (Cook et al. 1998)

CONCLUSIONS

It is clear that levels of ^{14}C found in *Fucus* spp. from the Irish coast are influenced to different degrees by ^{14}C liquid discharges from Sellafield, depending on location, with clear spatial differences found between the ^{14}C content of seaweeds from east and west coasts. Samples from the most westerly locations were found to be unaffected by Sellafield discharges, corresponding closely with global fallout levels, and showing a progressive decrease in concentration from 1985 to 2003.

All data sets show a progressive increase in concentration from the south and west towards the east and north coasts, with maximum values recorded in seaweeds collected along northeastern shorelines. In that region, the lowest ^{14}C concentrations in seaweed were found in samples from 1985, with highest values recorded in 1994 and 2003 (peaking in tandem with Sellafield releases). The spatial patterns observed are attributed to a combination of Sellafield discharges into the northeastern Irish Sea, and the predominantly south to north movement of water through the Irish Sea.

All measurements concur well with data on ^{14}C concentrations in seawater and seaweeds available from other studies (Cook et al. 1998; DEFRA 2002; FSA/SEPA 2003).

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