A RECENT HISTORY OF ^{14}C, ^{137}Cs, ^{210}Pb, AND ^{241}Am ACCUMULATION AT TWO IRISH PEAT BOG SITES: AN EAST VERSUS WEST COAST COMPARISON

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ABSTRACT. Data on radiocarbon (^{14}C), ^{137}Cs, ^{210}Pb, and ^{241}Am levels in an ombrotrophic peat sequence from a montane site on the east coast of Ireland are compared with data from a similar sequence at an Atlantic peatland site on the west coast. The ^{14}C profiles from the west and east coasts show a broadly similar pattern. Levels increase from 100 pMC or less in the deepest horizons examined, to peak values at the west and east coast sites of 117 ± 0.6 pMC and 132 ± 0.7 pMC, respectively (corresponding to maximal fallout from nuclear weapons testing around 1964), thereafter diminishing to levels of 110–113 pMC near the surface. Significantly, peak levels at the east coast site are considerably higher than corresponding levels at the west coast site, though both are lower than reported peak values for continental regions. The possibility of significant ^{14}C enrichment at the east coast site due to past discharges from nuclear installations in the UK seems unlikely. The $^{210}Pb_{ex}$ inventory at the east coast site (6500 Bq m^{-2}) is significantly higher than at the west coast (5300 Bq m^{-2}) and is consistent with the difference in rainfall at the two sites. Finally, ^{137}Cs and ^{241}Am inventories at the east coast site also exceed those at the west coast site by similar proportions (east:west ratio of approximately 1:1.2).

INTRODUCTION

On 10 October 1957, a serious reactor accident (classified as level 5 on the international nuclear event scale) occurred at Windscale (now referred to as Sellafield[2]) in Cumbria (UK), which resulted in the accidental discharge of a large quantity of radioactivity into the atmosphere in the 24-hour period following the incident (Cmnd 302, 1957; Arnold 1992). Important radionuclides released included ^{131}I, ^{132}Te, ^{137}Cs, and ^{210}Po, though some radiocarbon (^{14}C) was also discharged. By the early 1980s, renewed concern as to the radiological impact of this accident (called the Windscale Fire) on exposed workers and on the populations of the UK and northern Europe prompted a complete reassessment of consequential mortality and cancer morbidity in these populations (Crick and Linsley 1982; 1983; McGeoghegan and Binks 2000).

Our laboratory is currently completing a retrospective assessment of the impact of the Windscale Fire on Ireland. As part of this study, selected environmental archives have been examined for evidence of radionuclide input following the incident. The chronologies of the peat and lake sediment sequences analyzed were established using a combination of fallout markers and ^{210}Pb dating. Here, we report preliminary findings based on the analysis of ^{14}C, ^{137}Cs, ^{210}Pb, and ^{241}Am levels in peat monoliths from two ombrogenous peatland sites, the geographical locations of which are shown in Figure 1. One of the sites is on the Carlingford Peninsula, facing the Windscale/Sellafield complex 170 km distant across the Irish Sea, while the second is near Louisburgh on the western Atlantic coast, remote from this complex. The two sites have similar ecology, being dominated in each case by a 3–6 cm deep layer of live *Sphagnum cuspidatum*, with a gradual transition from moss to humified peat down each sequence.

Retrospective identification of any Windscale Fire fingerprint is unavoidably complicated by the impact of fallout from nuclear weapons testing, which peaked in the late 1950s and early 1960s, as well as by fallout from the Chernobyl reactor accident in 1986. On the local scale, radioactive effluents which have been discharged routinely to the atmosphere from the Sellafield complex since com-

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[2]Sellafield incorporates the Windscale site and is the location of a spent nuclear fuel reprocessing plant, operational since the early 1950s.

https://doi.org/10.1017/S0033822200041175 Published online by Cambridge University Press

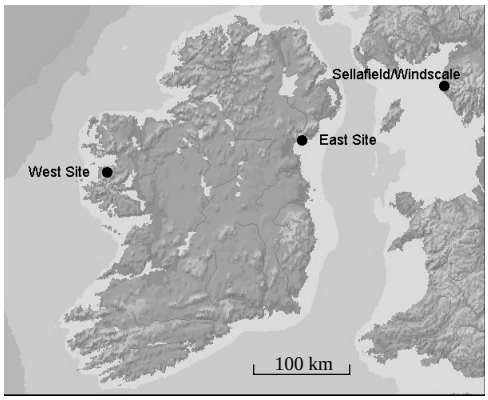

Figure 1 Locations of the sampling sites on the west and east coasts of Ireland, and the Windscale/Sellafield complex on the west coast of England

mencement of operations in 1951 may also complicate interpretation. Historically, discharges from Sellafield have greatly exceeded those from any other nuclear complex in western Europe and have been reviewed in detail elsewhere (Gray et al. 1995; Chamberlain 1996; Jones et al. 1996; McNamara and McCartney 1998). It is sufficient to note that annual discharges of ^{14}C to the atmosphere from Sellafield in the period 1951–1999, inclusive, averaged 7.0 TBq, peaking at 32 TBq in 1976 (Gray et al. 1995). The cumulative ^{14}C release from the UK's nuclear industry as a whole is estimated to have been about 1 PBq in the same period (McNamara et al. 1998). By comparison the cumulative release from atmospheric nuclear testing is estimated to have been 220 PBq (UNSCEAR 1993).

METHODS

Three peat monoliths were extracted from within a 10 m^2 area at both sites. Upon transfer to the laboratory, they were sectioned at a resolution of one centimeter and the outer rind of each discarded. Following drying, corresponding sections from the monoliths taken at each site were pooled and homogenized to produce a single composite sequence of samples. This approach is designed to reduce "front-end" variability, a feature often observed in studies of this nature (McGee et al. 1995). The bulk density of each section was also measured and recorded.

Equal volume aliquots of sample material were placed in low background, polyethylene counting vials and the activities of ^{210}Pb, ^{137}Cs, ^{241}Am, ^{214}Pb, ^{214}Bi, and ^{7}Be determined by low-energy photon spectrometry. The detector system was calibrated for energy and efficiency using standards supplied by NIST (US).

Normally, when ^{14}C dating, careful pretreatment of the sample is considered essential (Williams 1989; Cook et al. 1998). Here, however, the object of our study was to measure the total ^{14}C content of each section. Samples were, accordingly, converted to benzene without pretreatment. Measurement of the ^{14}C activity was carried out using an actively shielded, low-level, liquid scintillation spectrometer and was expressed as a percentage of modern carbon activity (pMC) according to convention (Stuiver and Polach 1977).

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RESULTS AND DISCUSSION

Radionuclide Profiles

The Western (Louisburgh) Site

The results of the analyses carried out on the west coast sequence are depicted in Figures 2 and 3. The ^{137}Cs deposition profile (Figure 2) shows a single, unresolved peak, comprising the relatively sharp Chernobyl accident pulse of May 1986 and the more protracted weapons fallout pulse which peaked in 1963–64. For this reason it is difficult to assign a precise horizon to the Chernobyl event, though there are grounds for assuming that it corresponds to a depth of between 4 and 6 cm. Similarly, the horizon associated with the peak in weapons fallout appears to lie between 7 and 10 cm depth. Evidence in support of this assignment is provided by the detection of weapons fallout ^{241}Am, which peaks within the same depth interval.

^{137}Cs has been reported to show some mobility in peat (McGee et al. 1995). Our data are consistent with this observation in that limited down-core movement of ^{137}Cs is apparent, with measurable concentrations present at levels well below the first detected occurrence of ^{241}Am. On the other hand, it is recognized that ^{241}Am is less mobile in peat than ^{137}Cs (McGarry 1991; Mitchell et al. 1992). Thus, it is reasonable to regard the ^{241}Am peak as a reliable horizon marker, approximating to the year 1964, when maximal fallout from weapons testing occurred (Testa et al. 1999).

The data for ^{7}Be deposition in the sequence are also given in Figure 2. Input of this short-lived, naturally occurring radioisotope ($T_{1/2}$ = 53.3 days) is via wet deposition. Given the half-life, detection was restricted, as expected, to the top few centimeters of the sequence within the zone of live vegetation where soil bulk density is least (Figure 3).

In the west coast sequence, there is good stratigraphic correlation between the ^{241}Am peak, referred to above, and the ^{14}C peak (Figure 2). It is logical that excess concentrations of ^{14}C in the terrestrial environment should reflect the occurrence of ^{241}Am since, in the main, they share the same source, namely, atmospheric nuclear weapons tests prior to the limited test ban treaty of 1963. The ^{14}C profile shows enhanced concentrations down to a depth of approximately 16 cm, with subsequent relaxation to concentrations slightly in excess of 100 pMC at greater depths. The profile follows a smooth progression with the exception of a lower than expected pMC value for the 6–7 cm interval. This interval coincides with a sharp density fluctuation, mirrored by a marked $^{210}Pb_{ex}$ anomaly (Figure 3). The latter may explain the low pMC value, since they suggest an exceptionally dry period during which there may have been some redeposition of carbon from older reservoirs onto the peat surface via wind blow.

The data are also interesting in that the concentration of ^{14}C never exceeds 117 pMC; a value which is considerably lower than previously reported for organic rich sediments in the northern hemisphere. In similar work, McGeehin (1998) found peak ^{14}C levels of over 150 pMC in a deltaic marsh sequence in Louisiana coinciding with the year 1964, and recorded pre-bomb levels of approximately 97 pMC, while Schell (personal communication) recorded peak 1964 levels of approximately 145 pMC in an organic soil sequence from Imnavait Creek in Alaska, with values falling to 100 pMC in 1950. Harkness et al. (1986) recorded levels of up to 155 pMC in the leaf litter of a forest soil sampled in northwest England in autumn 1972, while Jungner et al. (1995) recorded peak levels of 156 pMC in Sphagnum from a Finnish peat bog.However, peak ^{14}C content in atmospheric CO_2 and in tree rings considerably exceed all of the above values, with maximum concentrations in the region of 180 pMC (Goodsite et al. 2001).

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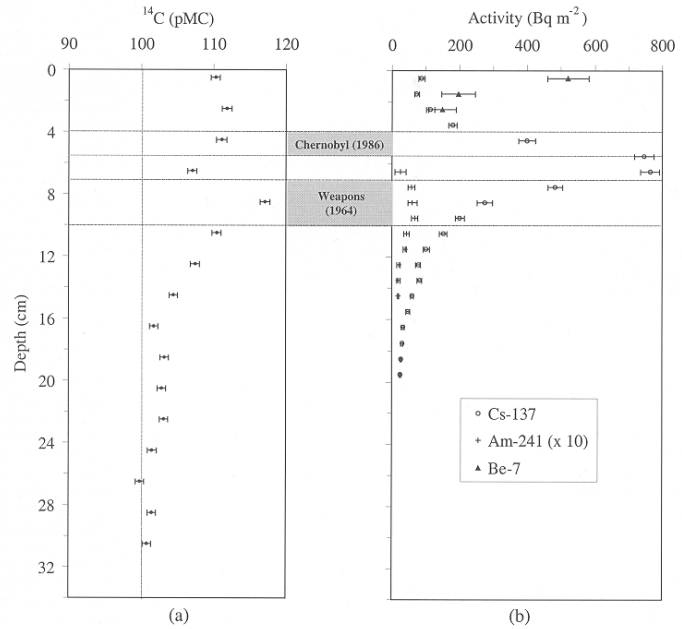

Figure 2 Radionuclide analyses of a composite peat sequence from the west coast site (Louisburgh) showing (a) ^{14}C and (b) ^{7}Be, ^{137}Cs and ^{241}Am profiles. Strata approximating to 1986 (Chernobyl event) and 1964 (weapons fallout peak) are also indicated on the diagram. Note, ^{241}Am values are scaled up by a factor of 10 for ease of display. Error bars represent ± 1 S.D.

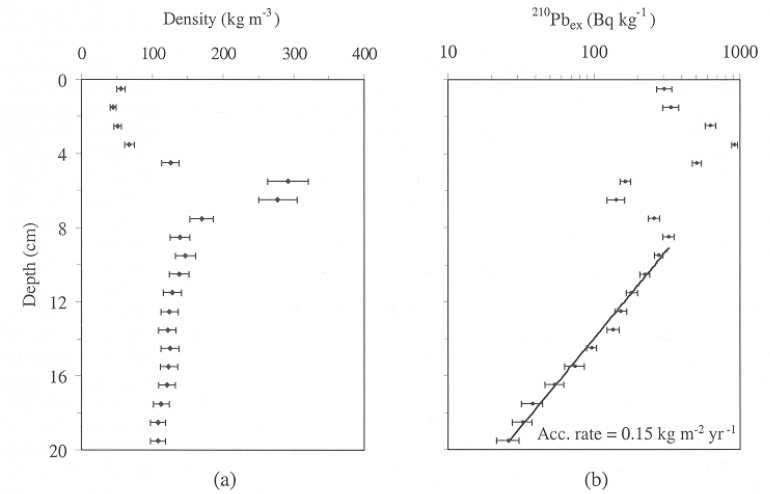

Figure 3 Bulk density and ^{210}Pb$_{ex}$ profiles for a composite peat sequence from the west coast site (Louisburgh). Error bars represent ± 1 S.D. The accumulation rate shown was determined using the CIC model.

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Analysis of ^{214}Bi and ^{214}Pb levels (not presented) in this sequence confirm that the ^{210}Pb content is almost entirely unsupported. However, the upper sections (i.e. 0–8 cm), besides showing lower than expected $^{210}Pb_{ex}$ concentrations, also exhibit considerable variability and, in isolation, cannot be used to construct a reliable chronology for this part of the sequence. The highest sections (i.e. 0–4 cm) comprise live vegetation and the lower than expected $^{210}Pb_{ex}$ concentrations observed can plausibly be attributed to growth dilution. Anomalous values at 6 and 7 cm depth coincide with greater ^{214}Bi and ^{214}Pb activities, indicating increased supported ^{210}Pb content. Below 8 cm, the $^{210}Pb_{ex}$ profile represents an excellent radioactive decay sequence ($r^2 = 0.99$). The peat accumulation rate, calculated using the constant initial concentration (CIC) model of Appleby and Oldfield (1992), was found to be 0.15 kg m^{-2} yr^{-1} (equivalent to a growth rate of 0.12 cm yr^{-1}) for this part of the sequence. This value is within previously observed ranges for peat accumulation at these latitudes (Clymo 1991; Mitchell et al. 1992; Tolonen et al. 1992).

The Eastern (Carlingford Peninsula) Site

The results of the analyses performed on the east coast sequence are given in Figures 4 and 5. In this case, the ^{137}Cs deposition profile (Figure 4) shows a broad and distinctly asymmetric peak which, again, can only be interpreted as an unresolved composite of Chernobyl and weapons fallout inputs to the sequence. The ^{241}Am profile displays a similar asymmetry; and both appear to be related to a sharp reduction in peat accumulation at the 13 cm depth horizon following a period of very rapid growth. The position of the ^{241}Am peak, in conjunction with the near identical position of the ^{14}C peak (Figure 4), clearly constrains the 1964 weapons fallout horizon within the 8–12 cm depth interval. Again, the data illustrate the comparative mobility of ^{137}Cs relative to ^{241}Am in peat systems. The presence of ^{7}Be in the first few sections of the sequence mirrors the pattern observed at the west coast site.

Levels of ^{14}C rise progressively to a maximum of 132 ± 0.7 pMC at a depth of approximately 9 cm, in good agreement with the ^{241}Am data. Peak ^{14}C levels in this sequence are significantly higher than the corresponding levels in the west coast sequence but, again, considerably lower than peak levels reported for other regions. It is acknowledged that there can be marked regional differences in ^{14}C concentrations (Levin et al. 1985; McCormac et al. 1995), and this may be a case in point.

It is significant that the ^{14}C content in strata below 21 cm falls to lower than expected levels, pMC values (circa 90 pMC or less) indicating a conventional ^{14}C age of approximately 800 years. The most likely explanation for these low values is a hiatus in the sequence at this depth. An abrupt change in the $^{210}Pb_{ex}$ and bulk density gradients at this stratum (Figure 5) supports this contention. However, the absence of detectable ^{241}Am below 19 cm suggests that any break in the sequence predates the onset of nuclear weapons testing (effectively, the early 1950s). It is noteworthy that we could not have identified a break in the sequence solely on the basis of ^{210}Pb, ^{241}Am and ^{137}Cs data, thus illustrating one of the benefits of ^{14}C analysis.

Along the east coast of Ireland peat soils occur only on high ground and are a rarity in that region relative to the western seaboard where they frequently occur at sea level (due to the oceanic rainfall pattern). Finding ideal sampling locations on the east coast was difficult and we sampled in the knowledge that the site chosen had been cut for fuel within living memory, but "probably not within the last 50 years". Our data appear to validate this information.

Again, low ^{214}Bi and ^{214}Pb concentrations in this sequence indicate that the ^{210}Pb content is almost entirely unsupported. The $^{210}Pb_{ex}$ data (Figure 5) indicate distinct phases of peat accumulation, with the CIC model giving accumulation rates in the range 0.09 to 0.33 kg m^{-2} yr^{-1} (equivalent to growth

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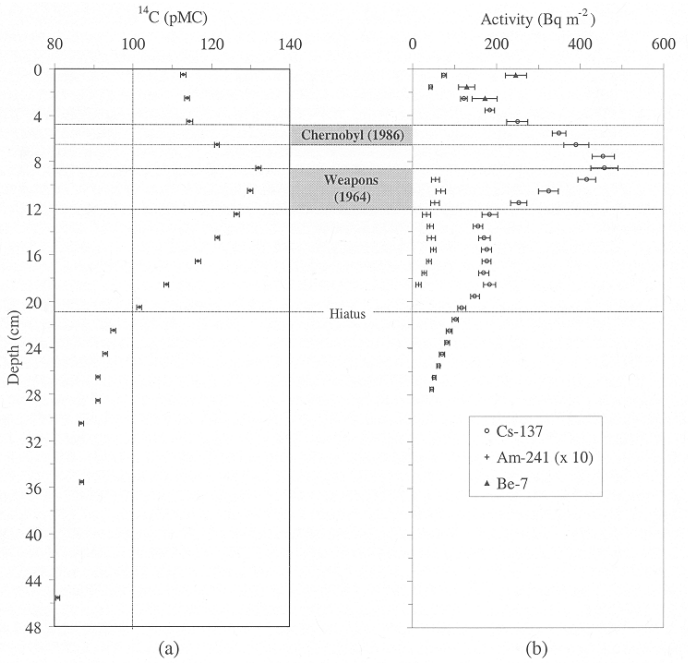

Figure 4 Radionuclide analyses of a composite peat sequence from the east coast site (Carlingford Peninsula) showing a) ^{14}C and b) ^{7}Be, ^{137}Cs, and ^{241}Am profiles. Strata approximating to 1986 (Chernobyl event) and 1964 (weapons fallout peak) are also indicated on the diagram. Note, ^{241}Am values are scaled up by a factor of 10 for ease of display. Error bars represent ± 1 S.D.

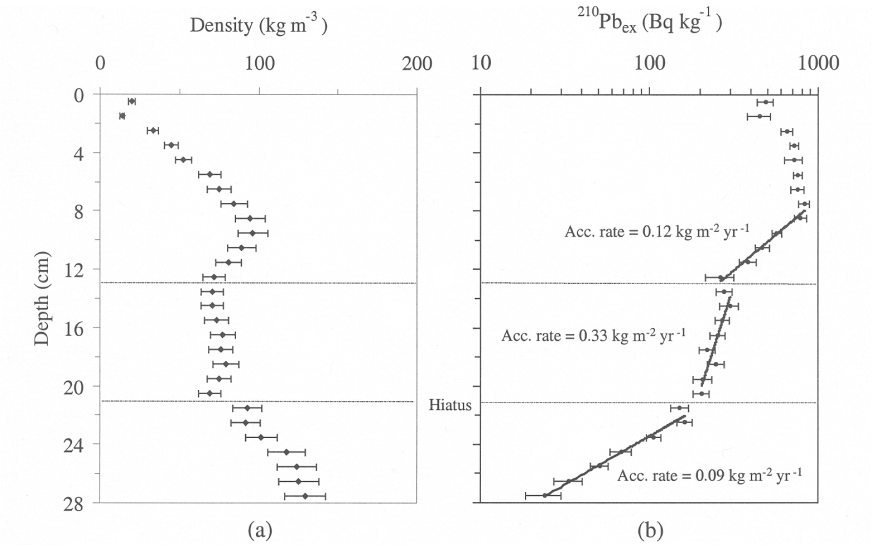

Figure 5 Bulk density and ^{210}Pb$_{ex}$ profiles for a composite peat sequence from the east coast site (Carlingford Peninsula). Error bars represent ± 1 σ. The accumulation rate quoted was determined using the CIC model. Horizontal lines indicate changes in trends of bulk density and ^{210}Pb$_{ex}$ data.

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rates of 0.08 to 0.45 cm yr^{-1}). Bulk density values increase progressively down the sequence to almost 100 kg m^{-3} at 9 cm depth. This value is similar to previously published data for Sphagnum bogs (Clymo 1991). Below this depth, abrupt changes in the bulk density gradient are apparent, which are matched by changes in the peat accumulation rate. Both of these features are probably indicative of changes in the composition of the plant communities that colonized the bog.

Inventories

Measured inventories for ^{14}C, ^{137}Cs, ^{210}Pb, and ^{241}Am at the two sites are given in Table 1. In determining these inventories we used as our reference horizon the depth at which ^{14}C enrichment values relaxed to pre-1950 levels, i.e. around 100 pMC.

The $^{210}Pb_{ex}$ inventory at the east coast site (6500 ± 300 Bq m^{-2}) exceeds that at the west coast site (5300 ± 400 Bq m^{-2}) by approximately 20%. A similar difference is evident in the case of ^{137}Cs and ^{241}Am, and can be attributed in all three cases to the difference in rainfall between the two sites (Table 2), which was also about 20% in the relevant period.

Bulk density measurements reveal greater organic matter accumulation at the western site (22.5 kg m^{-2}) relative to the eastern site (14.2 kg m^{-2}), where both values represent accumulation above the 100-pMC horizon. Hence, although we observe higher ^{14}C peak levels at the latter, the ^{14}C inventory for this site, at 1790 ± 20 Bq m^{-2}, is considerably smaller than the corresponding inventory for the west coast site (2100 ± 20 Bq m^{-2}). On the other hand, the post-1950 enrichment at the east coast site, although very small at 300 ± 10 Bq m^{-2}, is almost twice that found at the west coast site (170 ± 10 Bq m^{-2}). Whether any of this difference can be attributed to the greater oceanic influence at the latter site is unclear. However, the possibility of significant east coast enhancement due to past ^{14}C discharges from nuclear installations in the UK seems unlikely given the distance of our study site from these installations. It is relevant that Otlet et al. (1990), who measured enhanced ^{14}C concentrations in biota close to the Sellafield complex, reported a rapid fall off in concentrations with distance from the site.

Table 1 ^{14}C, ^{137}Cs, $^{210}Pb_{ex}$ and ^{241}Am inventories (± 1 S.D.) in composite peat sequences from the east and west coasts of Ireland

Location/site	Inventory (Bq m^{-2})			
	^{14}C	^{137}Cs	$^{210}Pb_{ex}$	^{241}Am
East Coast /Carlingford	1790 ± 20	4800 ± 200	6500 ± 300	43 ± 3
West Coast /Louisburgh	2100 ± 20	3900 ± 200	5300 ± 400	35 ± 3
East:west ratio	0.85 ± 0.02	1.23 ± 0.08	1.23 ± 0.11	1.23 ± 0.14

Table 2 Sampling site location, altitude, and mean annual rainfall

Location/site	Grid reference	Altitude (m)	Rainfall[a] (mm yr^{-1})
East coast /Carlingford	J 154 131	390	1500
West coast /Louisburgh	L 886 741	110	1250

[a]Estimated from published annual rainfall data for 1961–90 (Fitzgerald and Forrestal 1996)

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CONCLUSIONS

The greater $^{210}Pb_{ex}$ input at the east coast site is consistent with higher wet deposition, and is reflected in similar differences in the ^{137}Cs and ^{241}Am inventories. Significantly, the data for ^{137}Cs and ^{241}Am show no evidence of enhanced deposition at either site from sources other than weapons testing and Chernobyl.

Peak ^{14}C concentrations are clearly higher at the eastern site than at the western site. The strong oceanic influence on the western seaboard may partially account for this finding. Both of our sampling sites show lower peak ^{14}C pMC values than previous studies at continental locations would lead us to expect. It is difficult to sample peat sequences at very fine resolution, and as a result it is likely that the sharp peak from weapons fallout may be partially flattened.

There is a good stratigraphic correlation between ^{241}Am and ^{14}C maxima in both cores, and these data are used as mutually supportive evidence for the depths at which peak weapons-test fallout occurs. It is worth stressing the value of contemporary ^{14}C data when assigning chronologies to recent stratigraphic sequences. Such data usefully complement other more commonly employed dating techniques.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the support provided for this project by Enterprise Ireland (Grant Code SC/98/721), and the National University of Ireland, Dublin (Grant Code RP137/98). We also wish to thank Bob Kalin and Neil Ogle of The Queen's University of Belfast, who were responsible for the stable isotope analysis, and our colleague Luis León Vintró who provided helpful advice and assistance. Finally, we thank Bart Finn, Dave Cooney, Tom McKenna, Peggy Curran and Marian Hanson for continued technical and administrative support.

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