SURVEY OF THE DISPERSION OF ^{14}C IN THE VICINITY OF THE UK REPROCESSING SITE AT SELLAFIELD[1]

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ABSTRACT. We have been measuring ^{14}C in natural, biological materials growing in the vicinity of Sellafield, Cumbria as a continuing project with BNFL to understand the dispersion of releases from the site over several seasons. We have measured locally grown foodstuffs for monitoring purposes, individual tree rings to establish a chronology of releases, hawthorn berries for spatial investigations and are now carrying out controlled plot uptake experiments. We have been attempting to validate a current dispersion model (Clarke 1979) over a six-year period, and although we observe general agreement in most years, certain anomalies, which probably relate to topographical features, are leading to more detailed correlation with local meteorological data.

INTRODUCTION

The nuclear establishment at Sellafield, Cumbria, in northern England (54° 25′ 30″ N, 3°, 30′ W) was set up in the early 1950s and has, with continuing development, remained a major center for reactor research and development, power production and fuel reprocessing. The initial installations were two air-cooled graphite reactors specifically built to produce fissile material for the UK weapons program. In 1952, the first UK fuel reprocessing plant began work, in 1956 the Calder Hall power reactor—the first power reactor in the UK—came on line and in 1962 the first UK advanced gas-cooled reactor began operation.

Of these four nuclear installations, only the fuel reprocessing and the Calder Hall reactors are relevant to present-day emissions, and of these, the only radioisotope considered in these studies is ^{14}C. The mechanisms by which ^{14}C may be produced in reactor operations is given in Table 1, taken from Davis (1979). In practice, the dominant reaction in all the different types of reactor is ^{14}N(n,p)^{14}C. The main source of the target material, nitrogen, is from impurities in the fuel and its cladding, the latter largely containing the ^{14}C during its time in the reactor. The main release of ^{14}C occurs during fuel reprocessing at Sellafield after which it is discharged either as liquid effluent to the sea or gaseous emissions to the atmosphere. Evidence of these ^{14}C emissions was first observed in this laboratory (Harwell) in 1977 through measurements on various foodstuffs, milk, grain and potatoes collected from farms in the vicinity of the Sellafield site (Otlet, Walker & Longley 1983). Finding localized values higher than the normal atmospheric levels prompted subsequent investigations with initial studies focusing on establishing a chronology of ^{14}C emissions over the earlier years of operation.

CHRONOLOGY OF ^{14}C EMISSIONS

Using the technique of measuring ^{14}C levels in individual tree rings to establish annual values, we began work on a 100-yr-old sycamore tree, found in Knocking Wood, 1.6km NE of the site. Maximum and minimum diameters of the trunk were 1m and 0.5m, respectively, and ring widths were 1–10mm. Wood of each ring was stripped off using a carpenter's mallet and chisel providing ca 15–25g for measurement.

The results for the Knocking Wood tree are shown in Figure 1 compared to the average Northern Hemisphere atmospheric levels for the same period. For 1952–1963, the levels are slightly above but generally follow the atmospheric line for the Northern Hemisphere. The small peak in 1957 may represent the Windscale incident when one of the graphite reactors caught fire. It occurred in October, when leaves would still have been on the tree, but how much photosynthesis would have still been taking place is unclear.

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[1] This paper was presented at the 13th International Radiocarbon Conference, June 20–25, 1988, in Dubrovnik, Yugoslavia.

TABLE 1
Activation cross-sections and reactions for ^{14}C production in nuclear reactions (after Davis 1979)

Cross-section (barns)

Reaction	2200 m/s neutrons	LWR neutron spectrum	Fast reactor neutron spectrum
$^{13}C(n,\gamma)^{14}C$	0.9×10^{-3}	1×10^{-3}	0.5×10^{-6}
$^{14}N(n,p)^{14}C$	1.81	1.48	12.6×10^{-3}
$^{15}N(n,d)^{14}C$	0	0	1×10^{-3}
$^{16}O(n,^3He)^{14}C$	0	0	0.03×10^{-6}
$^{17}O(n,\alpha)^{14}C$	0.235	0.183	0.12×10^{-3}

After 1963, as fuel reprocessing grew, the line shows major departures from the atmospheric level with clear peaks in 1973 and 1976 and minima in 1968–1969, 1974 and 1978. We first thought that these features might show a direct relationship to the plants' operations. Figure 2 compares some recently estimated ^{14}C discharge values (based on operational data) with the tree ring values. The estimated discharges and the tree-ring values are plotted on different scales chosen to demonstrate their similarities to best advantage.
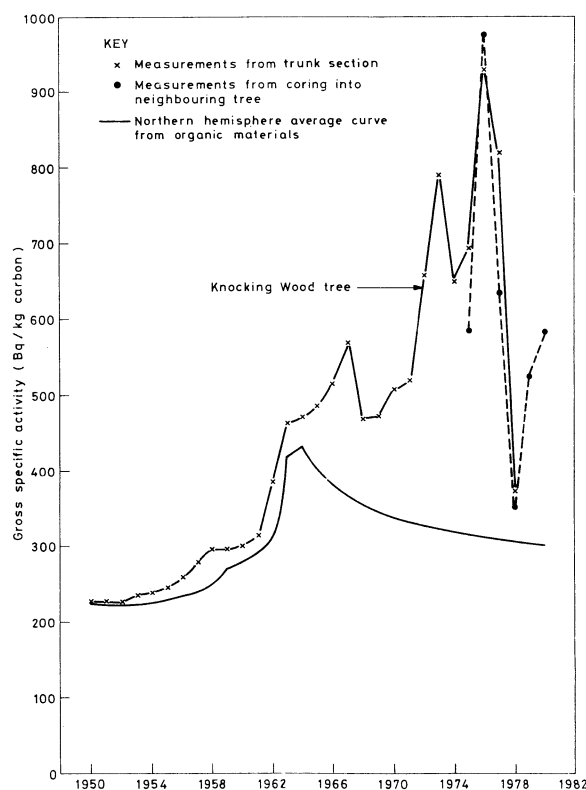

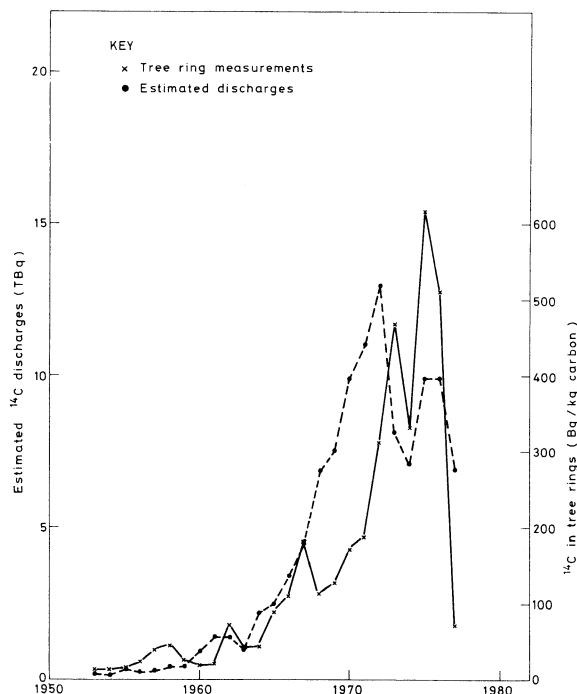Fig 1. ^{14}C in tree rings from Knocking Wood sycamore, 1950–1980, compared with Northern Hemisphere levels

Fig 2. Comparison of the tree-ring ^{14}C levels and estimated discharges of ^{14}C to atmosphere, 1953–1977

Although some agreement is visible, there are also some striking discrepancies, particularly the 1968–1969 trough in the tree-ring curve, matched with a strong increase in estimated discharges throughout, rising to a peak in 1972, one year ahead of the tree-ring profile. It is important, however, to realize the limitations of such a comparison. A tree only assimilates carbon during the daylight hours of the growing season and, possibly of more relevance, at only one location. Later work, described below clearly demonstrates that the average growing season concentrations in some directions are subject to large variations year to year due to the predominant wind fraction direction of that season.

An interesting spinoff result relevant to the techniques commonly used for wood pretreatment is the difference observed between samples pretreated using the standard AAA method (30 min boiling in 3M HCl + 30 min boiling in 1M NaOH + wash in 3M HCl) and those for cellulose extraction in clear verification of Olsson's (1980) earlier observations. Table 2 shows 6 samples measured by both techniques. The comparison was made after levels higher than the global average emerged from measurements made by the AAA technique for pre-1952 samples, *ie*, before the plants started working. In all cases, cellulose extraction samples gave lower results than those treated by the AAA technique, on average 2–3% lower, except one which was 8% lower. The greater efficiency of the cellulose technique is clearly demonstrated.

More trees were sought to observe spatial variations in the measured ^{14}C activities both at a distance and around more points of the compass. As there were not many that could be felled in the area, cores (ca 12mm in diameter) were taken from living trees; these were then split into yearly sections (ca 100mg wood per ring) and measured using small sample (mini-gas counter) measuring techniques (Otlet, Walker & Longley 1983). We were able to obtain results from a transect southward along the coast from Sellafield to ca 17km at Bootle. We later realized that this choice of line was unfortunate since later dispersion studies showed levels along the coastal boundaries are much lower than along inland transects and considerably more susceptible to perturbations caused by the meteorology of specific years.

TABLE 2

Comparison of ^{14}C results of AAA pretreated (whole wood) and extracted cellulose

Ring no.	Year	Gross activity (Bq/kg carbon)		Difference (%)
		Cellulose	Whole wood	
4	1975	674.5	694.5	2.9
10	1969	434.0	471.7	8.0
15	1964	458.4	472.9	3.1
19	1960	291.9	299.3	2.4
24	1955	239.4	246.8	3.0
30	1949	221.6	226.1	2.0

Thus, Figure 3 shows a general consistency but not an exact image in the main features of the Knocking Wood profile. Two groups of results that agree quite well within themselves appear to emerge (Fig 3), one for the sets of Knocking Wood, the Windscale AGR and Lady Wood (vii, iv, vi) and another for the distant sites of the coastal transect, Ravenglass and Bootle (ii, i) with those of Seascale (v) and Drigg (iii) showing less agreement to either in the more recent years.
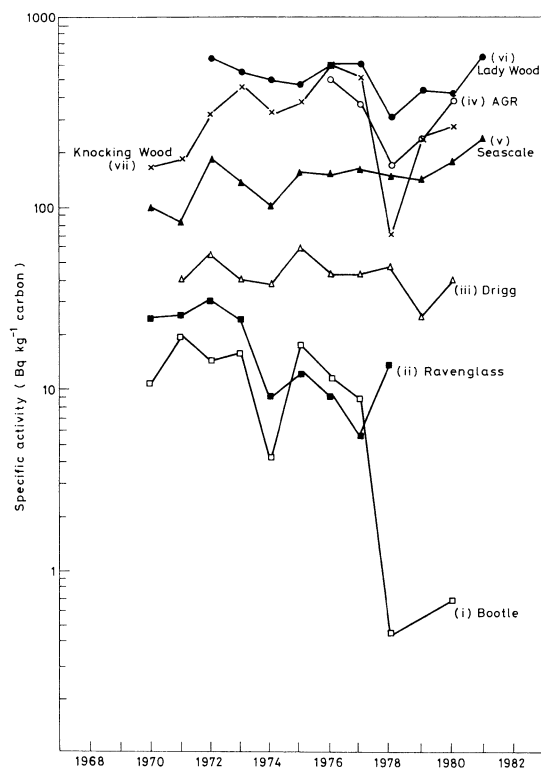

Fig 3. Tree-ring ^{14}C profiles from trees taken along the coastal transect

The suggested controlling factor of these groupings is the distance inland. Thus, the Lady Wood and Knocking Hill results, which are both approximately NE to E of the site, agree well with each other and with the few values obtained from the AGR tree. The AGR results, however, were noteworthy in that they do not appear to have the highest values of the series which, in view of their proximity to the Sellafield Works (the tree closest to the Works on which any measurements were made), might have been expected.

The less obvious, or in some cases, apparent lack of synchrony in the profiles, of the more distant sites is explained by the stronger effects of the sea breezes during the growing season at these coastal sites and highlights the point that this transect was not the best for dispersion study.

DISPERSION OF ^{14}C

Although the tree-ring technique yielded good averages over past years, the sparsity of trees in certain areas made it difficult to obtain data in a current year by year study over a wide area. Thus, we decided to choose a sampling material with guaranteed single-year growth, available over the widest possible area. Although grass and leaves, if sampled carefully would be candidates, a fruit was considered more reliable. Apple is known to accumulate CO_2 from neighboring leaves but, since apple is not common in the area, we chose hawthorn, *Crataegus*, a member of the apple family, which is prolific here. It grows from June to September, accumulating carbon, as determined experimentally both in southern England and in Cumbria, approximately exponentially with a halving time of about three weeks. Carbon uptake is similar to buildup in southern England (Walker, Otlet & Longley 1986) but about one week later in starting (this is relevant to the modeling assumptions mentioned below).

Initial studies on hawthorn looked at the variation with distance in samples collected along five transects radiating from the site (Fig 4). Although good agreement was found between the southward coastal transect and the tree-ring results, marked differences were observed between inland and coastal transects and these differences are the subject of continuing studies.

At first we thought that the differences were due to funneling, *ie*, concentration of air masses along valley lines, which, for convenience, were chosen for the transects. This idea was dispelled in 1982 when a sequence of points taken along a 4km arc to look for fine structure showed no particular correlation with the topography. We looked for an alternative explanation in annual meteorology, bearing in mind that, on the coast, pronounced sea
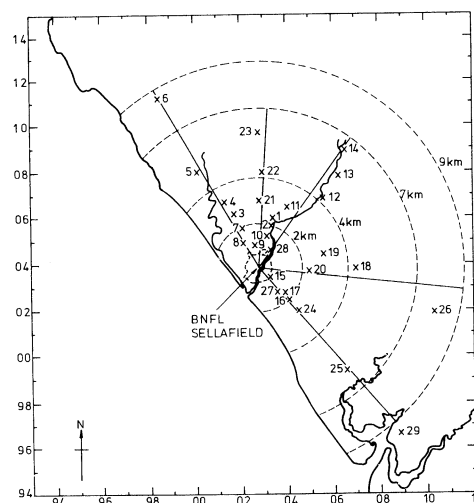

Fig 4. Hawthorn berry sampling sites along 5 transects, October 1981

breezes tend to occur during the daylight CO_2 uptake period. Thus, we must examine the directional wind frequency for the specific uptake period since this may be very different from the annual average. We have collected hawthorn berries from the same 4km arc and collection points over the last 6 years. The predicted distribution for each year was calculated from the meteorological data, using the NRPB dispersion model R-91 (Clarke 1979). We average the values deduced from the dispersion categories recorded at intervals during daylight and weight them according to the wind fraction of the sector and the relevant uptake rate of carbon from the carbon uptake curve. Figure 5 shows the results of the 6-yr analyses compared to the model predictions.

For years 1982, 1985, 1986, the predicted distribution generally agrees with the hawthorn results, although the positions of the predicted peaks are not precisely the same as those measured. Agreement is especially good in 1984, both curves showing a pronounced peak at 120° which can be explained by wind pattern. During the growing period there was a high frequency of winds blowing towards the 120° sector, *ie*, compared with the whole year, the growing period in 1984 had predominantly on-shore winds. Figure 6 illustrates the wind rose analysis.

In 1981 and 1983 (Fig 5), a quite large difference of ca 50° occurs between the measured and predicted peaks. The wind patterns for 1981 and 1983 were similar to 1984
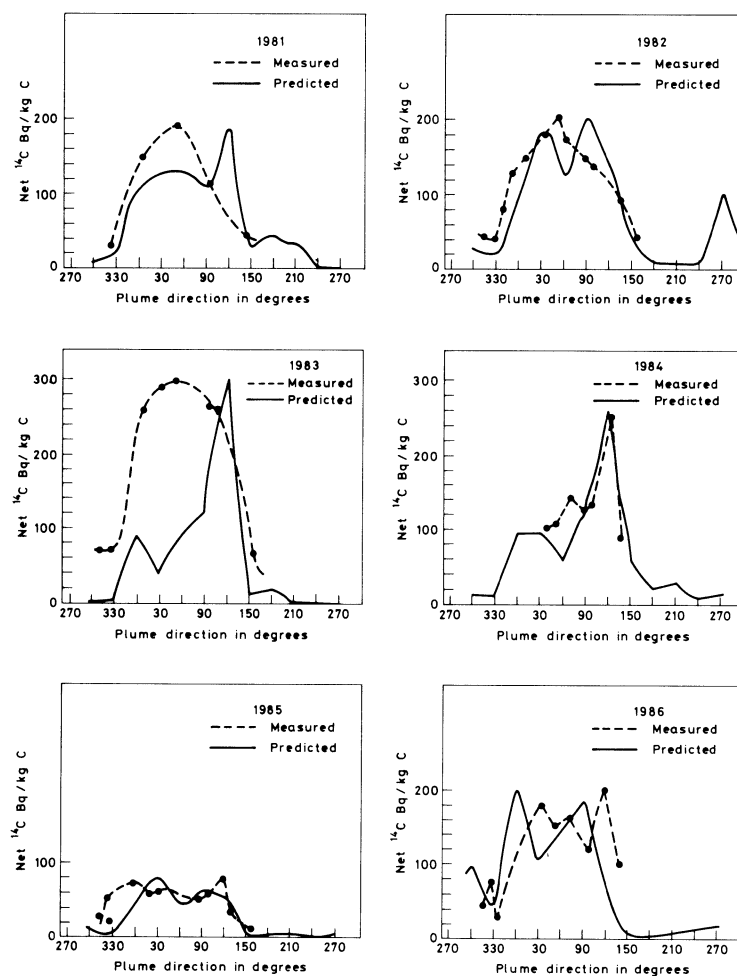

Fig 5. Net ^{14}C in hawthorn berries at 4km compared with predicted angular distribution, 1981–1986
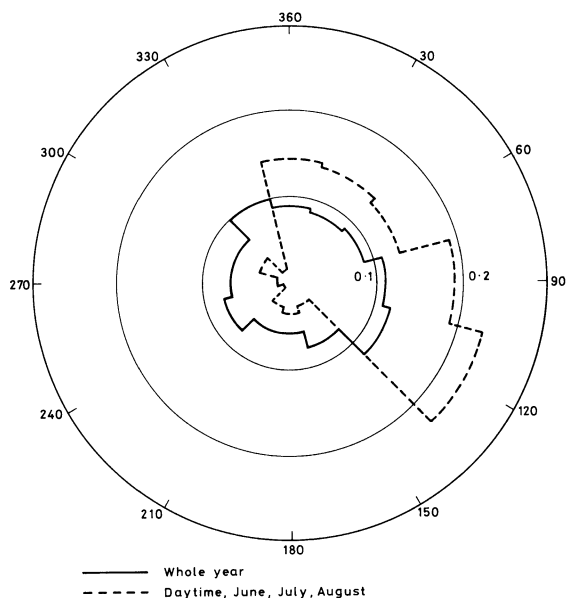
Fig 6. Frequency of wind blowing into different sectors, 1984

with predominantly on-shore winds blowing into the 120° sector and, although initially considered a possibility, the discrepancy cannot be explained by a difference in growing period for the hawthorns in these 2 years compared with the other 4. It is therefore clear that the experimental results cannot be explained in all cases simply in terms of wind fraction and dispersion category. A possible hypothesis is that of the particular topography creating variable plume curvature. In some directions, the land rises to 170m asl at 3.5km from Sellafield. Should this be causing plume curvature, the plume direction derived from the anemometer close to the source may not be adequate for predicting its position at 4km. (Plume curvature has been observed in SF_6 experiments carried out in similar wind conditions.) This year meteorological data are being collected at three points on the 4km arc as well as the Sellafield coast in the hope that a logical explanation for the discrepancies can be found.

We are also paying closer attention to on-site air-sampler measurements, operating one sampler during daylight hours in comparison with a second one operating full time. Other crops (leaf and root vegetables) are also being grown at one of the collection points to provide comparative uptake data.

CONCLUSION

Radiologically, the impact of ^{14}C from the Sellafield operations to the local environment is not significant and its presence can be detected only because of the sophistication of the ^{14}C measurement techniques. In the assessment of collective dose, however, with the big reductions of the earlier main contributors, eg ^{137}Cs, Webb *et al* (1986) predicted that ^{14}C would become the dominant isotope of the inventory. Studies directed to understanding the dispersion of ^{14}C and pathways to foodstuffs are, thus, likely to be of increasing importance in validating these prediction.

REFERENCES

Clarke, RH 1979 *A model for short and medium range dispersion of radionuclides released to the atmosphere.* NRPB-R91, Natl Radiological Protection Bd, Oxfordshire.
Davis, W 1979 Carbon-14 production in nuclear reactors. *In* Carter, MW, Moghissi, AA and Kahn, B, eds, *Management of low-level waste.* Oxford, Pergamon Press 1: 151–191.

Olsson, IU 1980 ^{14}C in extractives from wood. *In* Stuiver, M and Kra, RS, eds, Internatl ^{14}C conf, 11th, Proc. *Radiocarbon* 22 (2): 515–524.

Otlet, RL, Walker, AJ and Longley, H 1983 The use of ^{14}C in natural materials to establish the average gaseous dispersion patterns of releases from nuclear installations. *In* Stuiver, M and Kra, RS, eds, Internatl ^{14}C conf, 11th, Proc. *Radiocarbon* 25 (2): 593–602.

Walker, AJ, Otlet, RL and Longley, H 1986 Application of the use of hawthorn berries in monitoring ^{14}C emissions from a UK nuclear establishment over an extended period. *In* Stuiver M and Kra, RS, eds, Internatl ^{14}C conf, 12th, Proc. *Radiocarbon* 28 (2A): 681–690.

Webb, GAM, Cooper, JR, Dionian, J, Haywood, SM and Jones, JA 1986 The radiological impact on the public of nuclear power in the UK, 1970–1990. *In* Foratom IX (ENC 4/ ENC 86), *Nuclear energy of today and tomorrow.* Internatl conf, Proc. European Nuclear Soc, Geneva, June 1986.