[RADIOCARBON, VOL 28, NO. 2A, 1986, P 681–688]

APPLICATIONS OF THE USE OF HAWTHORN BERRIES IN MONITORING ^{14}C EMISSIONS FROM A UK NUCLEAR ESTABLISHMENT OVER AN EXTENDED PERIOD

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ABSTRACT. The paper describes a study undertaken to examine the dispersion pattern of gaseous emissions, as indicated by ^{14}C uptake in natural materials, around the nuclear reprocessing plant at Sellafield, Cumbria, UK. The extent and directional dispersion of the released ^{14}C is established and its variability over an extended period assessed. Results of measurements taken during three sampling seasons (1981, 1982, 1983) of the ^{14}C activity in hawthorn berries collected over a wide area are presented and the dispersion pattern contours constructed from them are examined. Only limited agreement of the results with the theoretical relationship 1/r is found and both meteorology and topography are seen to be important in determining the shape of the observed patterns.

INTRODUCTION

Measurement of ^{14}C in natural materials to monitor emissions, in the form of $^{14}CO_2$, from nuclear establishments is a well-established technique (Levin, Münnich & Weiss, 1980; Fischer & Muller, 1982; Otlet, Walker & Longley, 1983; Segl *et al,* 1983). The method is extremely sensitive because it was originally developed for measurement of activities below natural, as in dating applications, and is, in relative terms, very precise (±1%). In our earlier paper preliminary measurements were presented of the uptake of ^{14}C by some naturally growing or derived materials, eg, potatoes, milk, tree rings, and hawthorn berries, all collected from the vicinity of the BNFL reprocessing plant at Sellafield, Cumbria, UK. The feasibility of relating the observed values in the materials to gaseous dispersion patterns was considered and some trial dispersion transects from hawthorn berries growing in 1981 were given. It was in order to examine further the dispersion pattern side of the work that measurements were continued covering two more growing seasons.

Hawthorn berries were chosen for the continuation, in preference to leaves, shoots or grass, because it had been suggested that only with fruits could it be guaranteed that the incorporated carbon originated solely from CO_2 assimilated from the atmosphere during the growing period. The behavior of the hawthorn berry in assimilating carbon has now been separately studied. Random samples were plucked weekly (grab samples taken in darkness), from a tree in one of the authors' garden for ca 4 months, from the 2nd week of June 1983, the time the fruit set, to the 3rd week in October, when it began to wither. In each case the whole sample collected was combusted quantitatively to CO_2 and the average carbon content per berry calculates from the number in the sample (usually ca 50). The results are given in Figure 1. Carbon was assimilated for at least 3 months following an approximately exponential build-up curve with a half build-up time of ca 3 weeks.

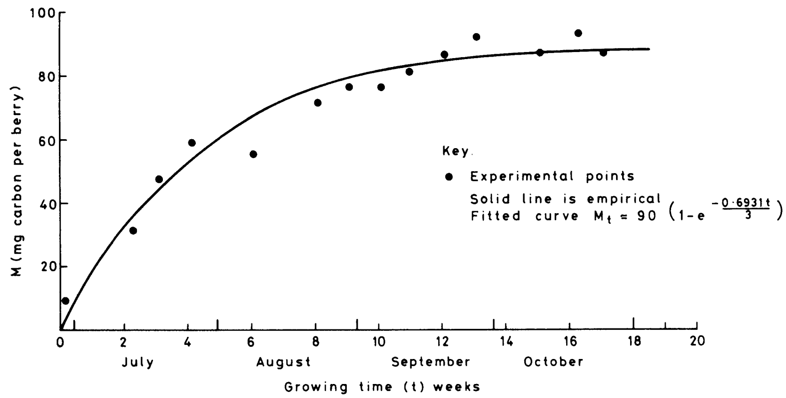

Fig 1. Uptake of carbon by Hawthorn berries over the growing period (Oxfordshire, 1983)

EXPERIMENTAL PROGRAM AND INTERPRETATION

The overall aim of this further experimental program was to consolidate the results of the initial experiment and to examine the variability in the directional distribution and the extent over which the emitted ^{14}C could be detected.

Hawthorn Berry Measurements

The most striking characteristics of the results from the initial 29 samples, collected in 1981 along five transects radiating from the center of the sites had been the significantly lower values seen at the coastal boundaries and the slower rate of fall-off of inland compared with the coastal transects. The transects had, for reasons of convenience, been chosen along valley routes where roads and tracks could provide easy access for sampling and, with hindsight, some doubts were expressed as to whether the landform had modified the dispersion patterns, in particular, as to whether funneling of the air mass into the valley regions had occurred.

The primary objective of the 1982 program was, thus, to investigate the variability of the distribution around a continuous arc extending from coast to coast on a constant radius. Twelve sampling points were selected, all lying on an arc 4km from the site. This distance was chosen for two reasons, 1) it was far enough to get into the hills, the highest sampling point lay ca 170m asl, where it should be possible to see clearly any contrast between high ground and valley readings, and 2) the values previously obtained at this distance were sufficiently above the background level to allow unambiguous interpretation of possible contrasting values.

The results together with those from the 1981 season which, coincidentally, lay on the same arc, plus the selected points of the 1983 program are shown in Figure 2 plotted around the points of the compass from the northern to southern coastal boundaries and a smooth curve of similar pattern for each year is obtained. The curves highlight the repeating pattern of lower values of ^{14}C towards the coastal boundaries and consistent maxima

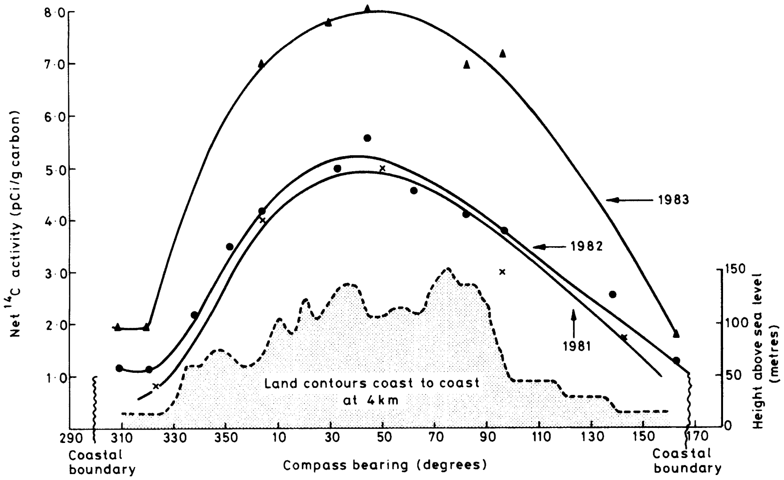

Fig 2. Results taken along a 4km arc in comparison with coastal boundary and topography

in the northeast quarter. 1983 is seen to be of higher overall amplitude throughout. A representation of the land height at the 4km arc is shown at the base of the figure and its shape is in rough agreement with the experimental curve showing higher values were the land is higher and *vice versa.*

An analysis was also made on the wind direction data available for the Sellafield site to investigate whether the pattern seen in 1981 was due to prevailing on-shore winds. Results of the 1982 4km arc are shown (Fig 3) in comparison with a histogram of the frequency of wind direction. The histogram of the plume fractions represents the percentage of time that the

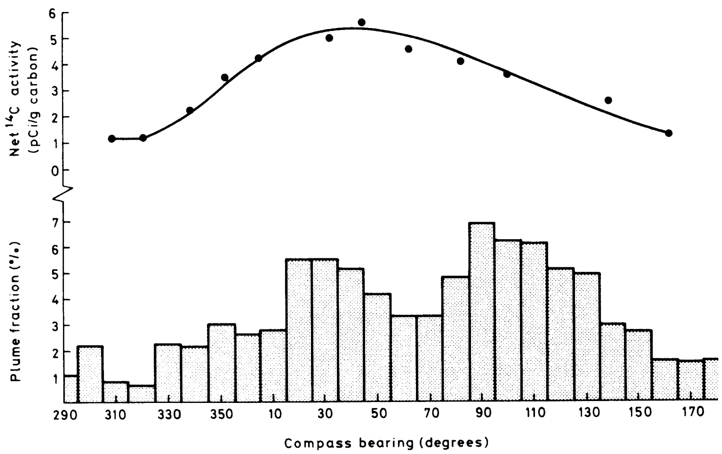

Fig 3. Comparison of 1982 4km arc with wind fractions weighted to include daylight hours and growing season effects (see main text)

wind blew towards the compass-bearing sector shown during the daylight hours of the hawthorn berry growing season. As the rate of hawthorn berry growth is non-linear (Fig 1) an additional weighting has been given to take into account the build-up rate of carbon in the berries during the growing period. Thus, greatest weight is given to the wind directions occurring during the most rapid growing time, *ie*, the first three weeks. As with Figure 2 the overall shape of the two distributions is similar with greater frequencies of wind occurring in the directions where the activity peak lies and it seems that both wind and topography are influencing the distribution.

Although 1983 provided data for the 4km arc survey above, the primary objective of this program was to examine the full geographic extent over which the emitted ^{14}C could be detected and to consider the results in comparison with theoretical relationships. A wider-reaching survey was, therefore, planned, aimed at covering distances up to 36km from the site. Accordingly, 5 arcs were chosen at ca 4, 8, 16, 24, and 36km from the site and 60 samples collected along them.

Figure 4 shows a map of the study area with the 1983 sampling points marked. Plotted around them are isopleths, *ie*, contours of equal activity, selected from those produced using a computer package developed at Harwell by D Newton. The package generates smoothed curves from sets of values at known coordinates, which are randomly distributed. Both the size of the map and the density of the contours can be specified, thus, in this case, it was possible to specify a scale of 1:250,000 and isopleths at 1pCi/g carbon intervals.

The survey clearly showed the extent of the dispersion with measurable levels of ^{14}C found at over 30km in the northeast quarter. An interesting facet of the year's results was the similarity of the activity isopleths to the land contours after ca 5km, eg, the way the isopleths follow the line of the deep valley (Wasdale) to the east of the Sellafield site. The shaping of the contours suggests a marked topographic effect, the valleys offering preferred routes, which becomes more marked at greater distances. Measurable values were obtained on high ground and also in valleys beyond hill ranges. One sampling point, eg, lay 28km to the southeast in the shadow of hills rising to >800m; no valleys cut through in this direction but a ^{14}C value, measurable above the laboratory's minimum detectable level, was found. It would, thus, appear that the behavior of the plume on meeting hilly ground is complex and further experimental results would probably be needed to describe the situation in more detail.

It was thought useful, however, to compare the patterns observed from the 1983 measurements for agreement with what might be expected from a basic Gaussian plume model (Miller, 1984). The results were normalized to the 4km arc of Figure 2 to account for both meterologic and topographic effects and a suitable method of presentation sought. A first attempt simply plotted results as activity against the reciprocal of the distance, (1/r). Although of value for a limited range, the more distant points were too compressed for clear interpretation. A clearer picture is presented in Figure 5, where activity and distance from the source are plotted on a log scale. In this the 1/r relationship is approximately true up to 15km,

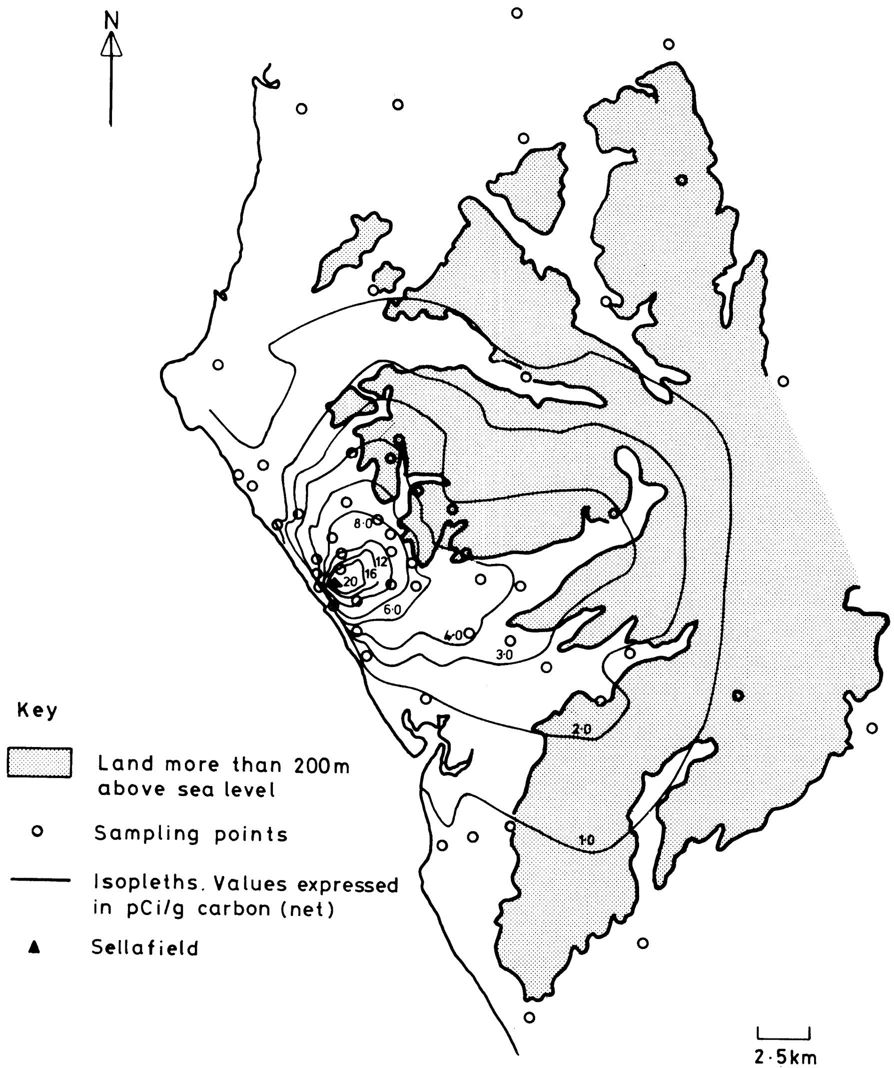

Fig 4. Isopleth representation of 1983 hawthorn berry survey

(indicated by the 45° line around which the experimental results cluster reasonably well), but after that agreement breaks down, the decrease in activity is much more rapid. The results provide data which will be used in a more sophisticated model by an associated group at Harwell.

Air Sample Measurements

As we realized that natural materials had limitations in providing the true picture of dispersion from the establishment, which works 24 hours a

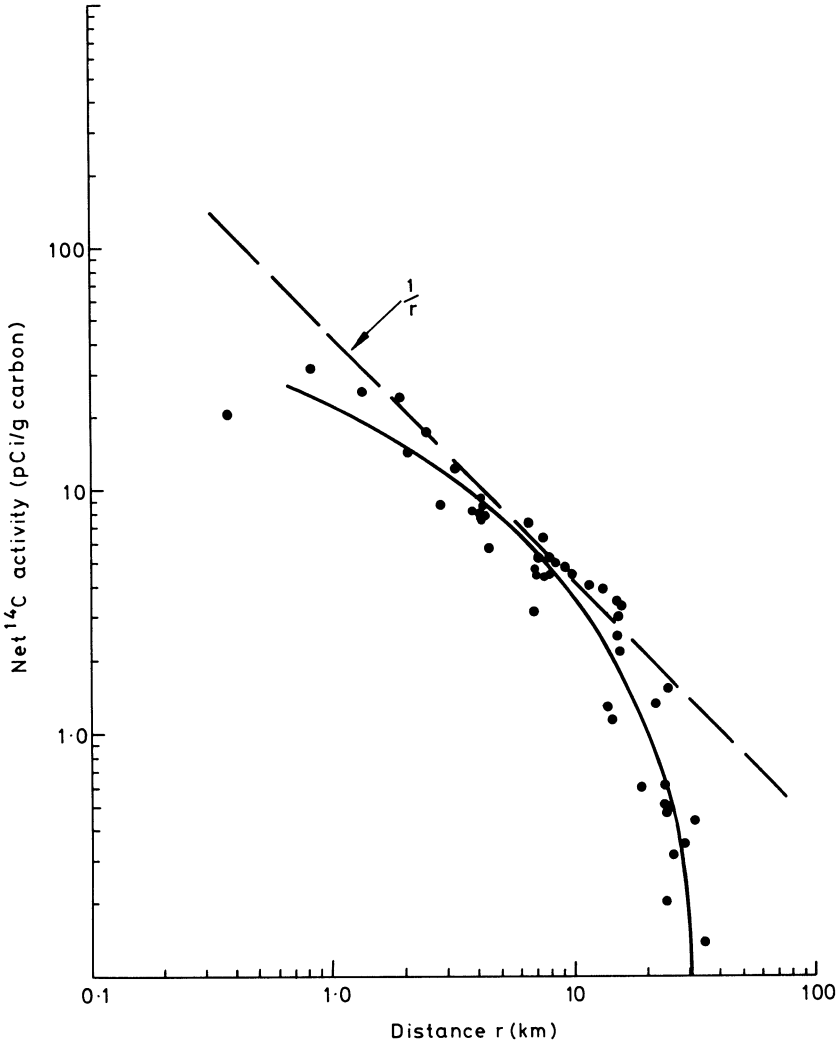

Fig 5. Uptake in comparison with distance

day all year, three air sampler stations were set up to provide continuous measurements to compare with the hawthorn berries. Two of the stations were very close to the site, just beside the perimeter fence and one, Brow Top, lay ca 3km north. All three sites are close to hawthorn berry collection points. The sampling apparatus consisted simply of a small aerator type pump (1 L^{-1}), a rotameter, and a vessel containing 10L 0.25M NaOH solution.

Table 1 shows the activities recorded at the three stations for each month from March to September 1983. Monthly variations are seen with highest values in June and July. In Table 2 a comparison is made of the air

TABLE 1
Air samples during growing period, March to September 1983

Distance from site	Net ^{14}C activity (pCi/g carbon) Brow Top 2.5km N	Calder Gate 1.0km E	North Group 0.8km NE
Month			
March/April	1.68	9.27	8.64
May	4.77	11.33	7.02
June	10.77	13.06	(25.16)
July	(10.28)*	12.47	19.73
Aug/Sept	7.78	(13.47)	9.45

* Results in parentheses are interpolated values

sampler measurements and the results for those locations derived from the isopleths. The air sampler measurements are means for the months of the growing period only and are weighted to account for the carbon build-up rate of the berries. An acceptable agreement was obtained between the two sets during this period.

CONCLUSIONS

From the results of this survey we conclude that measurement of ^{14}C in natural materials can provide a valuable indicator of local gaseous dispersion patterns around nuclear establishments as well as straight monitoring of the effects of emitted activity. To provide a reliable picture, however, a number of factors must be considered, eg, period of growth, rate of carbon uptake,topography, and meteorology. The limited agreement found in this study with the simplified Gaussian plume model theory was interesting and provides a basis for further research in determining the effect of topography on dispersion patterns in hilly or mountainous terrain.

ACKNOWLEDGMENTS

Thanks are due colleagues at Harwell who have helped in the preparation of this report and, in particular, to I G Hutchinson of Instrumentation and Applied Physics Division, for supplying the computer isopleth contours

TABLE 2
Comparison of air samples and isopleths

Sampling site	Net ^{14}C activity (pCi/g carbon) Air sample (mean for hawthorn growing period)	Isopleth value
Brow Top (2.5km N of site)	9.61	9.1
Calder Gate (1km E of site)	13.00	16.2
North Group (0.8km NE of site)	18.11	16.0

of Figure 5. The financial support of the Department of the Environment, British Nuclear Fuels and the Atomic Energy Research Establishment, Harwell is gratefully acknowledged. This research is conducted by agreement with these agencies and is carried out as part of a comprehensive study of radioactive materials in the Cumbrian environment.

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