[Radiocarbon, Vol 25, No. 2, 1983, P 593-602]

THE USE OF ^{14}C IN NATURAL MATERIALS TO ESTABLISH THE AVERAGE GASEOUS DISPERSION PATTERNS OF RELEASES FROM NUCLEAR INSTALLATIONS

R L OTLET*, A J WALKER*, and H LONGLEY**

ABSTRACT. The Harwell Low Level measurements Laboratory has been measuring a variety of natural materials from locations close to United Kingdom nuclear installations since 1977. The measurements are made first, to establish the past effects of releases to the atmosphere of ^{14}C as observed from individual tree rings and second, to establish dispersion contours averaged over extended periods. The main study area has been in Cumbria, around the BNFL nuclear installation at Sellafield. ^{14}C, which can be measured to good precision even at values close to the normal natural levels, provides a powerful technique for the provision of practical experimental values much wanted for theoretical dispersion models.

INTRODUCTION

The many and varied applications of low-level ^{14}C measurements play an important role in a large number of research disciplines. The measurement technique has been developed to a very high standard to meet the demands of its original and major application, ^{14}C dating. For ^{14}C dates to be of value to an archaeologist, they must be made to a precision of at least ± 1% and this on levels well below those occurring naturally, i e below 6 pCi/g of carbon. With this precision, it is clear that levels only a few picocuries above natural can be easily detected and measured. We present here some new applications of particular relevance to the nuclear industry and, on a wider scale, the protection of the environment. Investigations were made by Harwell in the vicinity of the BNFL nuclear installation at Sellafield in collaboration with BNFL, the operators of the plant, and the Department of the Environment. The goal was to obtain data on gaseous dispersion patterns and the effect of releases of ^{14}C to the atmosphere.

*Low Level Measurements Laboratory, AERE, Harwell, Oxfordshire,
OX11 ORA, UK
**Environmental Section, British Nuclear Fuels Ltd, Sellafield,
Cumbria, CA20 1PF, UK

BACKGROUND AND EARLY WORK

The natural level of ^{14}C has been extensively studied in recent years (Vogel and Marais, 1971, Levin, Münnich, and Weiss, 1980; Barrette et al 1980) etc. Research began in Britain with Baxter, Ergin, and Walton (1969) and was continued by Harwell from 1971 onwards (fig 1).

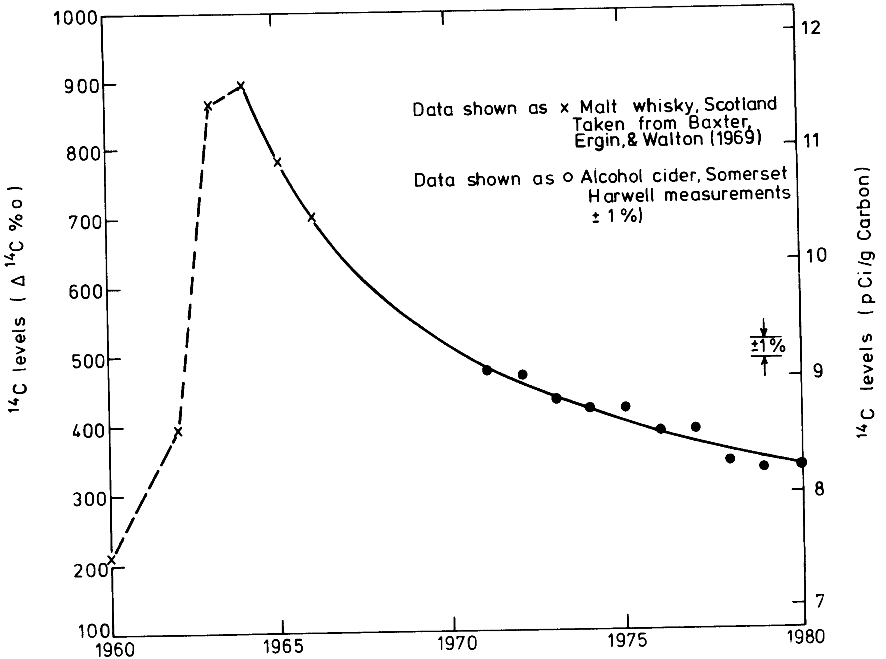

FIG 1. ^{14}C LEVELS IN ALCOHOL SINCE 1960 (UK)

The now familiar profile rises in 1963 to twice (ie $\sim$ 12 pCi/g carbon) the natural level which is close to the ^{14}C standard value (0.95 x NBS oxalic acid ^{14}C value of 6.16 pCi/g carbon equivalent to $\Delta^{14}C \pm 0^{o}/oo$). The 1963 peak provides an important time-marker for a number of applications. The measurements were made on alcohol (malt whisky by Baxter, cider by Harwell) but supporting measurements were also made at Harwell on grain and sugar from various locations. In 1976, we began work on a monitoring survey of samples of milk, grain, and potatoes from local farms close to BNFL, Sellafield. Some results of these measurements averaged between 1976 and 1979 are given in table 1. They show levels above the northern hemisphere of figure 1 from locations close to the site and to a lesser extent at distances up to and beyond 10km.

TABLE 1. ^{14}C in foods

Distance from Sellafield (km)	^{14}C in foods (pCi/g^{-1} C)* Milk	Grain	Potatoes
up to 1	2.90	3.23	0.88
1 to 2	6.28	0.68	4.42
2 to 4	2.40	3.29	3.62
5 to 7	1.64	1.44	0.59
ca 30	0.25	0.22	-

* Averages calculated from available data of 1976 to 1979, after subtraction of the contribution due to global levels.

Although discernible by ^{14}C measuring techniques, these levels are insignificant in terms of radiation exposure to the public, ie $< 0.2\%$ of the dose-equivalent limit recommended for critical groups by ICRP (ICRP, 1977). Their importance in providing much needed experimental dispersion parameters was, however, realized and promoted the investigations reported here.

DEMONSTRATION OF CHRONOLOGIC RECONSTRUCTION

The first of these, the establishment of a chronology of emissions of ^{14}C from the site, began with individual tree-ring measurements on a sycamore felled in April, 1979 ca 3km NE of the site. The tree was a large sycamore, felled for road widening. A slice was taken through the trunk, from the butt end of the tree. It was not particularly circular, with major and minor diameters of ca 0.90m, respectively. The pith was almost centrally placed but the ring widths varied considerably around the whole circumference. A sector was chosen in which the rings were even and visible and ring widths increased from ca 1mm near the bark to ca 10mm at ring 30.

Annual measurements were made on the last 30 years' growth of the tree. The samples ($\sim$ 15g dry wood from each ring) were pretreated using the standard 'AAA method' (acid-alkali-acid). Each sample was first boiled for 30 minutes in 3M $HC\ell$, backwashed to neutral, boiled for 30 minutes more in 1M NaOH and backwashed again. A final acid (3M $HC\ell$) wash removed any carbonates deposited in the sample from the tap water. The acid was washed out with distilled water. Supporting measurements were also made on cellulose but full details of these comparative results will be described elsewhere. Liquid scintillation counting techniques were employed using an optimum sample size of 6g C_6H_6. Samples were converted to C_6H_6 using the normal procedures of the laboratory and were processed in sequence starting with the bark and finishing with ring 30.

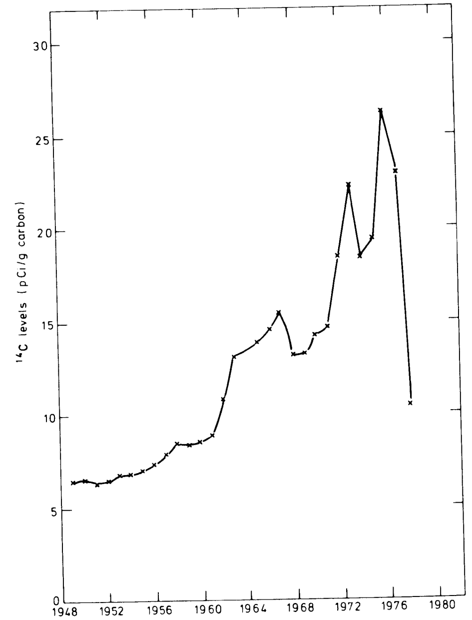

FIG 2 ^{14}C LEVELS FROM TREE RINGS OF A SYCAMORE TREE NEAR SELLAFIELD

Figure 2 presents absolute ^{14}C levels which include the contribution of the global average values. The initial rise between 1956 and 1963 is approximately coincident with the northern hemisphere average rise (fig 1) over the same period. The values from 1976 to 1978 are also broadly consistent with the values of ^{14}C in milk, potatoes and grain (table 1). The detailed structure of the profile probably relates to the nuclear fuel reprocessing programmes at BNFL, Sellafield. It is of interest to note that no significant variation can be attributed to the 1957 Windscale incident.

Following these preliminary measurements, we repeated the experiment using five trees from selected locations at increasing distances and different directions from the site. The results from annual measurements on these generally support the initial results but showed attenuation of the ^{14}C levels at greater distances.

The introduction of micro-sample (micro-counter) measurement facilities (Sayre et al, 1981) should greatly facilitate the tree ring measurement research. The two counter sizes require only 60mg and 10mg of carbon, respectively. The 60mg sample can be measured to ± 1% (counting statistics) in ca 10 days, and with simultaneous operations of a group of such counters, sample throughput need not be a problem.

The advantage of using miniature samples is that they can be obtained by boring and extracting a complete core from healthy living trees. Coring can be done without damage to the tree which facilitates the collection of suitable samples at required locations. Preliminary experiments using a 12mm core borer on a variety of tree species shows that the size of core from a healthy tree provides well-defined ring sections which are not difficult to isolate and of ample weight for the larger counter size (fig 3).

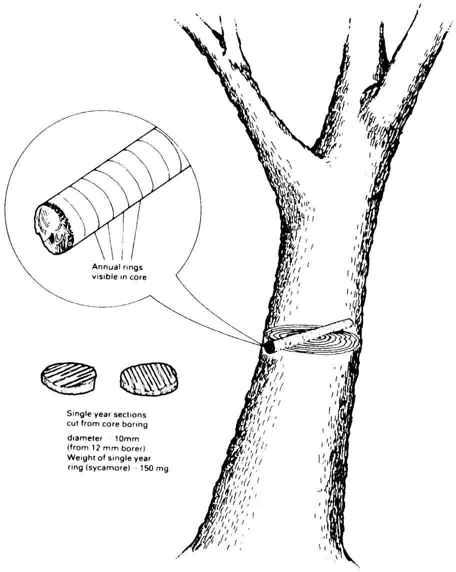

FIG 3 A CORE EXTRACTED FROM A BORING ACROSS A FULL DIAMETER AND ITS BREAK-UP INTO ANNUAL SECTIONS

These tree-ring methods can provide simultaneous chronologic and dispersion information. However, at the study site, the problem of obtaining trees even suitable for the micro-counter severely limited the possibility of finding full spatial dispersion data quickly. Thus, we sought a different type of environmental material for this purpose.

DEMONSTRATION OF GASEOUS DISPERSION PATTERNS

Advice was sought from the Department of Forestry, Oxford on the most suitable material for this study. A number of criteria had to be satisfied: 1) the material should be truly representative of a single years growth; 2) it should be readily available throughout the area; 3) it should grow for a period long enough to provide an average of wind and weather patterns, and 4) it should occur naturally rather than being planted and tended for the experiment. We were advised that fruit would be better than grass, fresh shoots or leaves. The growth period of grass is short and difficult to prove even at all sampling points. New shoots are difficult to identify later in the year and are known to be produced partly from carbohydrate stored in the tree from the previous year (Kramer and Kozlowski, 1960). Leaves of a tree species found widespread in the area seemed more suitable and have been used successfully for this type of application. However, as with shoots, the first leaves of the season are made from the previous years' carbohydrate and only when photosynthesis has been active for some time can the leaves be certainly attributed to that year. Thus, we decided to use a fruit since it fits all the criteria and appeared to have no intrinsic problems.

Extensive research on the apple has shown (E Reynolds, (pers comm) that it is produced solely from photo-synthesis in the surrounding leaves. In the area under study, the hawthorn (Crataegus Monogyna) which belongs to the apple family is ubiquitous and was therefore selected.

The first samples of hawthorn berries were collected in October, 1981 at 32 sites, 31 in the study area (fig 4) and 1 in Yorkshire, 110km away, as a control.

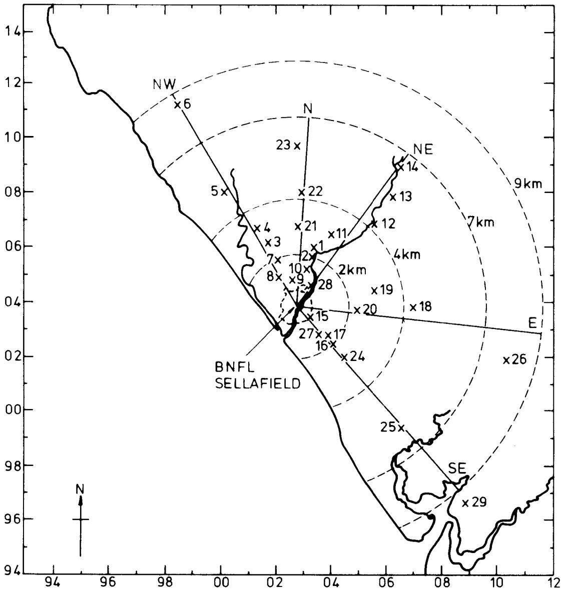

FIG 4 HAWTHORN BERRY SAMPLING SITES, OCTOBER 1981

The sites were located along 5 transects radiating inland from the site along different compass directions. The three major transects, along each of which 6 to 8 samples were collected, lay at approximately NE, NW and SE'ly directions. The two minor transects, lying N and E provided 4 more samples each. For each sample, ca 200g of ripe hawthorn berries were collected. Pretreatment consisted of removing stems and leaves, washing, boiling in acid, washing again in distilled water and drying in a hot cupboard. The process produced 40 to 50g of dry weight; 17g of this produced ∿ 12 STP 1 of CO_2 which provided 6g of benzene for liquid scintillation counting.

Figure 5 shows preliminary results of the first sampling, in which the ^{14}C levels, expressed as picocuries ^{14}C/g of carbon minus the expected global levels, are plotted as a function of distance from Sellafield. The individual transects are identified by lines hand-drawn through the relevant experimental points. These lines indicate the dilution along each of the transects, from which the two-dimensional pattern of dispersion can be derived. Thus, these measurements provide an opportunity to study the actual dispersion of ^{14}C around Sellafield, to compare the observed concentrations with those predicted from theoretical dispersion models (eg Clarke, 1979), and to consider the effects of local topography and meteorology. These further studies are now in hand.

We have observed that all the lines show a reduction of ^{14}C concentration with distance from Sellafield, but the concentration gradient varies for different directions and the spatial dispersion of ^{14}C appears to be non-uniform.

Similar dispersion patterns are observed for the N, NE and E transects. However, the lines are considerably different for the NW and SE transects which show lower concentrations of ^{14}C and very different concentration gradients. The highest concentrations are seen on the NE transect, and are ca 5 times those of the NW transect. Figure 5 shows that the NW and SE transects are approximately parallel to the coast and 1 to 2km inland; in general, the sampling points are at lower elevations than those on the N, NE and E transects. The ^{14}C concentrations on the NW transects are about half those on the SE transect. Thus, it seems that both topography and proximity to the sea appear to be influencing the dispersion pattern. Directional dispersion is affected by the average wind vectors during the growing season, but this does not account for the differences observed.

CONCLUSIONS

Both experiments provide basic data for more detailed analysis and interpretation. They indicate the importance of low-level ^{14}C measurements on natural materials in providing

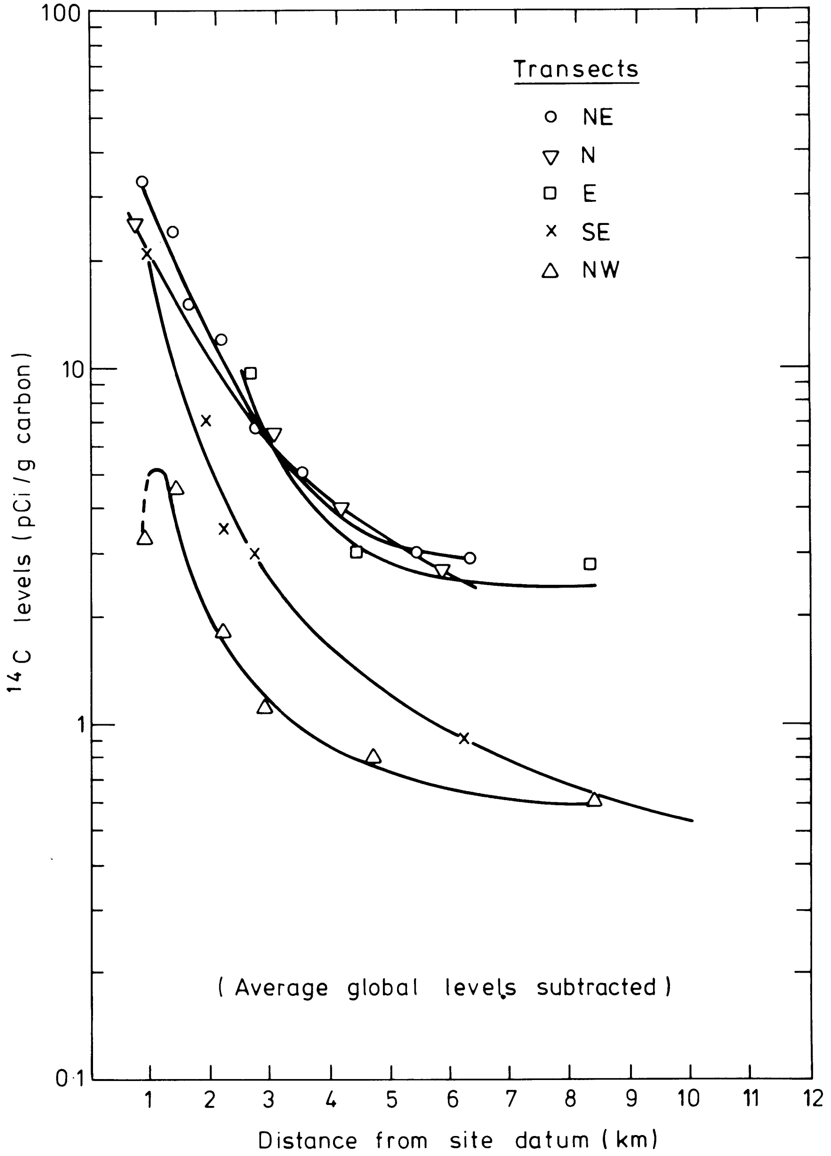

FIG 5. PRELIMINARY DISPERSION PATTERN DATA FOR 5 TRANSECTS OF THE 1981 HAWTHORN BERRY SURVEY

data that would be difficult to collect by physical instrumentation. The difficulties of setting up and tending instruments at 31 sites for several months are considerable.

Since not enough is known of the rate of carbon uptake of natural materials during growth, we plan to investigate this problem. Direct air sample measurements are also being made to determine the relation of air concentration to plant uptake. With the data on the dispersion parameters for specific sites, this relationship will correlate the ^{14}C tree-ring data to chronologic discharges from Sellafield.

Further, these ^{14}C measurements will help to assess the past and present environmental impact of ^{14}C discharges on the radiation exposure of the public, since ^{14}C levels in locally grown foods will be similar to those recorded in tree-rings and hawthorn berries. The studies will also refine our knowledge of the effects of ^{14}C emissions even though observed levels are low and represent only a small fraction of the ICRP dose limit for the public. Although much more research is needed, these initial measurements make a valuable contribution to the nuclear power industry in providing the first experimentally determined dispersion data for ^{14}C.

ACKNOWLEDGMENTS

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

REFERENCES

Barrette, L, Lasalle, P, Martel, Y, and Samson, C, 1980, Variations of ^{14}C in oats grown from 1957 to 1978 in Quebec, in Stuiver, M and Kra, RS, eds, Internatl ^{14}C Conf, 10th, Proc: Radiocarbon, v22, no. 2, p 410-414.

Baxter, MS, Ergin, M, and Walton, A, 1969, Glasgow university radiocarbon measurements I: Radiocarbon, v 11, p 43-52.

Clarke, RH, 1979, A model for short and medium range dispersion of radionuclides released to the atmosphere: NRPB-R91, London HMSO, Nat Radiol Protection Bd

ICRP Pub 26, 1977, Internatl Comm on Radiol Protection, Annals of the ICRP, v 1, no. 3.

Kramer, PJ and Kozlowski, TT, 1960, Physiology of trees: New York, McGraw-Hill Book Co, p 150-159.

Levin, I, Münnich, KO, and Weiss, W, 1980, The effect of anthropogenic CO_2 and ^{14}C sources on the distribution of ^{14}C in the atmosphere, in Stuiver, M and Kra, RS, eds, Internatl ^{14}C Conf, 10th, Proc: Radiocarbon, v 22, no. 2, p 379-391.

Sayre, EV, Harbottle, G, Stoenner, RW, Otlet, RL, and Evans, GV, 1981, The use of small gas proportional counters for the carbon-14 measurement of very small samples, in Methods of low level counting and spectrometry: Vienna, IAEA, p 393-408.

Vogel, JC and Marais, M, 1971, Pretoria radiocarbon dates I: Radiocarbon, v 13, p 378-399.