LOCATION-DEPENDENT DIFFERENCES IN THE ^{14}C CONTENT OF WOOD

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ABSTRACT. The long ^{14}C chronologies currently used as calibration curves combine results from wood that grew in the western United States, the British Isles and Germany. Although these results show few significant differences in the ^{14}C content of contemporaneous wood when averaged over the length of the chronology (*i.e.*, the means of overlapping sections of chronology are the same), closer examination shows considerable variability. Separating the sections of chronology according to the provenance of the wood used for calibration reveals patterns that suggest small but finite differences in the ^{14}C content of wood from different locations. We conclude that there is some evidence that German and American wood give dates older by between 20 and 40 yr from those of Irish oak for some periods. Additionally we suggest that the shift of the Belfast 1986 calibration data by *ca.* 18 yr toward older dates may not be valid and that the resultant offset between the Belfast 1986 and Seattle 1993 data shows a small but real difference in the ^{14}C content of contemporaneous American, German and Irish wood. Intralaboratory measurements made in Belfast on contemporaneous German and Irish oak, and bristlecone pine and Irish oak, give offsets of 39 and 41 yr, respectively, with the Irish oak dating younger. Previous studies, in which sample pairs of American and English and French wood were processed in the same laboratory, also showed American wood to be slightly depleted in ^{14}C. None of the findings of this study would significantly alter calibrated ^{14}C dates.

INTRODUCTION

The ^{14}C contents of contemporaneous organic material of the same type from different locations are not always identical. The largest discrepancies are found in the oceans where shells can have apparent age differences of 500 yr or more from contemporary terrestrial material (Stuiver, Pearson and Braziunas 1986). This difference is caused by the mixing of old deep ocean water with surface water from which the shell carbonate derives. Variations in the ^{14}C content of contemporaneous terrestrial material have also been observed. Lerman, Mook and Vogel (1970) and Vogel *et al.* (1993) showed that trees from the southern hemisphere were older by *ca.* 30 and 41 ± 5 ^{14}C yr, respectively, compared to similar-aged northern hemisphere trees. Cain and Suess (1976) attributed the difference to the influence of the larger expanse of ocean in the southern hemisphere and the slightly greater wind agitation there. On a smaller scale, Bruns *et al.* (1980) showed that local depletions of ^{14}C can occur in the vicinity of active volcanoes, where the out-gassing of ^{14}C-deficient CO_2 causes nearby trees to date older than contemporaneous trees that are located well away from the volcano. The offset in such cases can be as much as 1600 yr. Levin *et al.* (1985) showed how the ^{14}C level varies from rural ground-level sites in Central Europe to higher-elevation mountain regions, illustrating anthropogenically induced ^{14}C distribution variations. Damon *et al.* (1992) described a regional effect when they measured ^{14}C differences of 2.6‰ in wood from the Olympic Peninsula and the Mackenzie delta area. Thus, at present, and in the past, ^{14}C has not been uniformly distributed throughout the troposphere, but has exhibited both local- and hemispheric-scale variations as observed in terrestrial organic material.

Prior to the industrial revolution, intrahemispheric variations in ^{14}C are thought to have been negligible, as apparently confirmed in the extensive calibration data published by Stuiver and Pearson

Proceedings of the 15th International ^{14}C Conference, edited by G. T. Cook, D. D. Harkness, B. F. Miller and E. M. Scott. RADIOCARBON, *Vol. 37, No. 2, 1995, P. 395–407*

(1986) and Pearson and Stuiver (1986), where high-precision measurements of Irish oak wood and contemporaneous Douglas-fir, sequoia and German oak show no significant offsets for periods of overlapping data. Despite the apparent uniformity exhibited in the extensive Pearson and Stuiver (1986) and Stuiver and Pearson (1986) data sets, close examination (*cf.* Fig. 1), shows short periods during which they diverge by as much as 50 yr. Stuiver and Pearson (1986) showed this by plotting the distribution of the differences between the Belfast and Seattle data, which they determined had a standard deviation of 25.6 yr. Given that the laboratory uncertainties had predicted a standard deviation of 22.9 yr between measurements and that the average difference over the length of the chronology was <1 yr, the observed differences would seem to be explained.

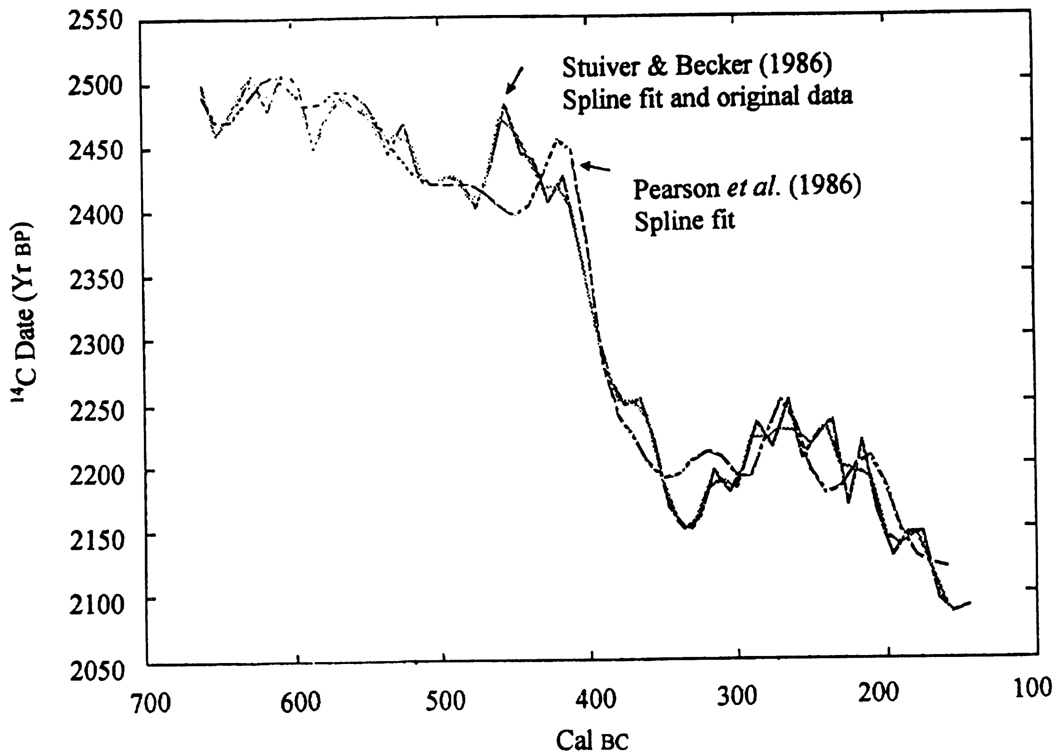

Fig. 1. Interpolated calibration curves from Stuiver and Becker (1986) and Pearson *et al.* (1986), illustrating that although the mean of the two data sets are very similar, considerable differences exist in the structure of the curves

After the publication of the 1986 calibration curve, the data were revised and extended, and a "new" calibration curve published (Stuiver and Pearson 1993; Pearson and Stuiver 1993). The section of this new curve that covers the same time period as the 1986 calibration data shows a shift toward older dates of about 18 yr for both Irish and American wood. Stuiver and Becker (1993) describe in detail the rationale behind the shift in the Seattle data set (*i.e.*, a radon correction was required). Pearson, Becker and Qua (1993) attribute the change in their data to an efficiency variation with time. We present evidence that suggests the Irish results published in 1986 are correct and the recent shift of 18 yr may not be valid. As a consequence, we suggest that an offset exists between the Irish wood and the American wood such that the American wood dates slightly older than wood from Ireland. This offset of American wood toward an older age is further supported by intralaboratory measurements of wood from the United States and Europe recorded in the literature and by quasi-simultaneous comparative measurements we have made on Irish oak and bristlecone pine.

The conclusions drawn from the excellent agreement between the Belfast and Seattle data in 1986 settled several issues in ^{14}C research. First, the accuracy of both laboratories using different techniques was established, implying that the true activity levels of the known-age wood had been determined. Second, the activity levels of wood from widely separated regions in the northern hemisphere were identical, apparently confirming hemispheric uniformity in ^{14}C. However, differences that have emerged in the revised 1993 calibration hint at a hitherto unsuspected level of complexity in the ^{14}C distribution both spatially and possibly temporally.

Replicate analysis 1986 to 1993

Pearson, Becker and Qua (1993) give the reason for moving the 1986 Belfast calibration data by *ca.* 18 yr toward older dates as a variation in measurement efficiency with time. This explanation is difficult to reconcile with the almost constant offset of the data throughout the length of the chronology (Fig. 2). Pearson, Becker and Qua (1993: Table 1) illustrated the consistency of measurements on replicate samples between 1977 and 1988. Their results show that for 6 sample pairs the mean difference between results obtained in 1977 and 1988 is 13.4 ± 10.0 yr, with the 1988 dates being younger. If the 1977 data had not been corrected for the efficiency variation with time the offset would have been −0.5 ± 10.0 yr. Pearson, Becker and Qua (1993) average the results in their Table 1 to obtain the new values for these intervals in the 1993 calibration. Interestingly, this group of points is not shifted significantly from its position in 1986 (Fig. 2A). Thus, the data set used to show the consistent reproducibility of measurements through time does not justify the shift of the rest of the dates.

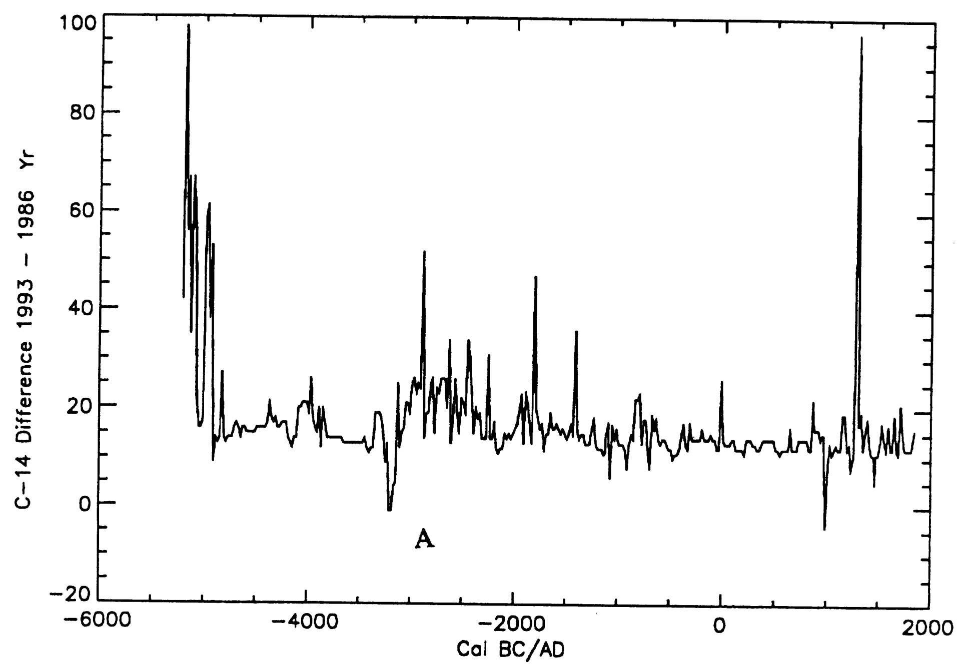

Fig. 2. Difference between the published ^{14}C dates of Pearson *et al.* (1986) and Pearson and Qua (1993). Positive values indicate 1993 dates are older. A = position of 1977–1988 replicates.

https://doi.org/10.1017/S0033822200030873 Published online by Cambridge University Press

TABLE 1. Intralaboratory Comparison of Dendrochronologically Dated German and Irish Oak Samples

Age (cal BC)	German oak (age in yr BP)	Irish oak (age in yr BP)	Difference (yr)
4910	6012 ± 18	6037 ± 20	+25
4930	6042 ± 18	5986 ± 11	−56
4950	6087 ± 18	6081 ± 15	−6
4970	6139 ± 25	6065 ± 15	−74
4990	6123 ± 22	6054 ± 15	−69
5090	6185 ± 20	6160 ± 18	−25
5110	6199 ± 20	6143 ± 17	−56
5130	6109 ± 20	6127 ± 17	+18
5150	6249 ± 18	6176 ± 18	−73
5170	6221 ± 18	6151 ± 17	−70

The Belfast pine sample TIRI core sample B (Gulliksen and Scott 1995) was a 40-yr block of dendrochronologically dated wood from Garry Bog, Northern Ireland (*i.e.*, the site of origin of the Irish oak samples that were used at that point in the original Belfast calibration curve (McCormac *et al.* 1995)). The pine comprised two bidecadal blocks of wood centered on 3210 and 3230 BC (*i.e.*, identical ages to the last two samples in Table 2 of Pearson, Becker and Qua 1993). Repeated measurement of this standard by the Belfast laboratory and other laboratories supports the original (uncorrected) Pearson *et al.* (1986) values and the repeat values obtained by Pearson, Becker and Qua (1993). The weighted mean from ten independent determinations gave a value of 4521 ± 7 yr BP. The new measurements by Pearson, Becker and Qua (1993) gave a value of 4525 ± 14 yr BP; the original Pearson *et al.* (1986) value was 4524 ± 9 yr BP. The corrected values for these points published in Pearson, Becker and Qua (1993) give a value of 4539 ± 9 yr BP. Ongoing quality control measurements in the Belfast laboratory using known-age wood also support the 1986 results. Thus, no new measurements that could justify a correction of the Irish oak data were made for the period 5210 BC to 1840 AD in the Belfast laboratory and measurements that have been made point to the 1986 data as being correct.

TABLE 2. Recent Belfast measurements on bristlecone pine and Irish oak. The original measurements of Pearson *et al.* (1986) are shown for comparison.

Age (cal BC)	1 Bristlecone pine (^{14}C yr BP)	2 Irish oak 1995 (^{14}C yr BP)	3 Pearson *et al.* (1986) (^{14}C yr BP)	Age 2−Age 3	Age 2−Age 1
495	2455 ± 23	2454 ± 16	2419 ± 18	35	−1
485	2501 ± 23	2430 ± 16	2422 ± 18	8	−71
465	2467 ± 20	2389 ± 17	2391 ± 18	−2	−78
425	2498 ± 20	2460 ± 19	2442 ± 16	18	−38
415	2444 ± 20	2432 ± 17	2440 ± 18	−8	−12
405	2407 ± 23	2389 ± 17	2404 ± 18	−15	−18
395	2353 ± 21	2317 ± 19	2321 ± 18	−4	−36
385	2375 ± 20	2318 ± 18	2266 ± 15	52	−57
325	2263 ± 21	2208 ± 19	2205 ± 18	3	−55

Further confirmation of the consistency of Belfast Irish oak measurements with time can be found in Pearson *et al.* (1977) (Table 2) where a floating chronology of dendrochronologically dated oak was published. The center date for the first block of wood in that floating chronology (tree ring age (0) as given in their table) was subsequently fixed at 2090 BC. Comparing the Pearson *et al.* (1977) measurements with Pearson *et al.* (1986), we find that the 1977 data, as published, are offset by 8.4 ± 4.1 yr toward younger dates, once again illustrating the long-term consistency of the results.

Differences in Irish, German and American Wood in the 1986 and 1993 Publications

Figures 3A and 3B show a comparison of the differences between Belfast and Seattle results from the 1986 and 1993 calibrations. The data from both chronologies have been fitted with cubic splines and interpolated at 10-yr intervals before subtraction (McCormac and Baillie 1993). Differences have been binned at 250-yr intervals. Although the overlapping periods from 1986 have differences that do not show significant systematic trends (Fig. 3A), the differences in the extended 1993 data sets (Fig. 3B) increase to a maximum near 1800 BC and thereafter decrease to 6000 BC. The period 5180–5500 BC in the 1993 data, we believe, is erroneous because some of the German wood supplied for calibration purposes may have been incorrectly tree-ring dated. This partially explains the unacceptably large offset of 54 yr for the interval 5180–5500 BC reported by Stuiver and Becker (1993). The systematic variation in the differences outside the 5180–5500 BC interval might be related to systematic measurement error. However, because the Belfast calibration measurements were not made in age order, systematic variations of the type observed are unlikely in these results. The radon correction described by Stuiver and Becker (1993) could induce a systematic time-dependent difference that would explain the older Seattle dates for the period when only European wood was used in the chronology (*i.e.*, 2495–145 BC) but it would not explain the trend toward younger German dates with respect to Irish between 2500 and 5000 BC.

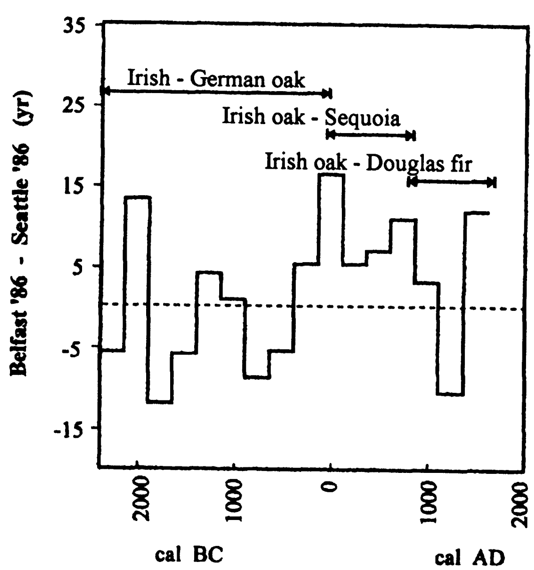

Fig. 3A. Difference between the Pearson *et al.* (1986) data set and the Stuiver and Becker (1986) data set with the differences binned and averaged every 250 yr. B. Difference between the Pearson and Qua (1993) and the Stuiver and Becker (1993) data sets with the differences binned and averaged every 250 yr. In both A and B, all data were fitted with cubic splines and interpolated at 10-yr intervals before subtraction.

Comparison of the data for the period *ca.* 2500 BC to 1820 AD in Figures 3A and 3B shows a significant change between 1986 and 1993. In 1986 the mean difference for the AD portion (essentially the Irish minus American ^{14}C ages) was 2.6 ± 2.3 yr and for the BC portion (Irish minus German) –3.4

https://doi.org/10.1017/S0033822200030873 Published online by Cambridge University Press

± 2.1 yr; the mean differences of the equivalent data in 1993 are −0.25 ± 2.2 yr for the AD period and −13.25 ± 2.1 yr for the BC period. If the 1986 Belfast data (5210 BC to 1840 AD) are correct (as we are suggesting) then the differences in Figure 3B should be increased by 17.75 yr, making the Irish wood date 17 yr younger than the American wood and 30 yr younger than the German oak back to 2500 BC (Fig. 4). Before 2500 BC the offset decreases, implying a time-varying difference between Irish and German wood. The large offset between Irish and German wood in the chronologies is unexpected. However, further support for this finding comes from intralaboratory measurements made in Belfast on German oak in 1988 to measure the overlap with the end of the Irish oak chronology (Table 1).

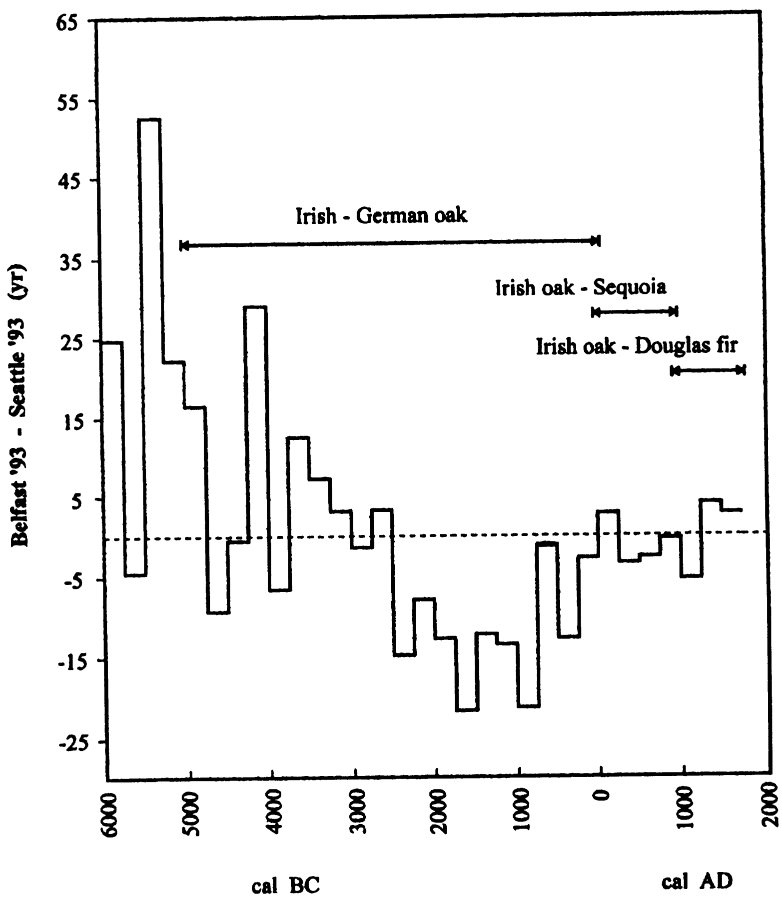

Fig. 3B. See Fig. 3A.

These results give an offset of −38.9 ± 8.0 yr between contemporaneous Irish and German oak, with the German dates being older (*i.e.*, a result consistent with the offset observed in the long chronologies). Thus, if the 1986 Belfast data and the corrected 1993 Seattle data are accepted (and all that follows assumes this), then the measured offset in the German samples given above is consistent with the calibration results between 2495 and 145 BC where an offset of *ca.* 30 yr exists. Assuming that the German dendrochronology is secure over this interval we are led to the conclusion that local effects must have resulted in ^{14}C differences between Irish and German oaks. The data for the inter-

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val 2500–5000 BC, which includes some of the above measurements, shows a progressive reduction in the offset between the German and Irish dates. To summarize, the differences in the 1993 Belfast and Seattle data sets are significantly greater than those for the equivalent time periods published in 1986. Justification for the shift of the Belfast dates in 1993 by 18 yr is lacking and hence we suggest an offset may exist between contemporaneous Irish and American wood. Additionally, 1986 Belfast data has an offset from the Seattle German oak measurements that is consistent with the offset measured in Belfast between contemporaneous German and Irish wood.

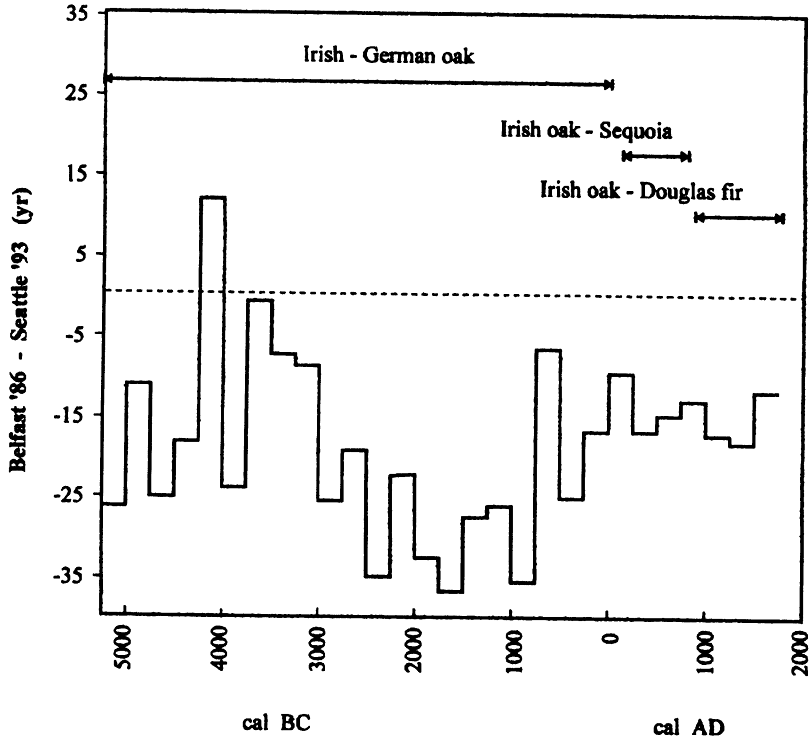

Fig. 4. Difference between the Pearson *et al.* (1986) and the Stuiver and Becker (1993) data sets with the differences binned and averaged every 250 yr

BRISTLECONE PINE AND IRISH OAK MEASUREMENTS

We obtained nine decadal samples of dendrochronologically dated bristlecone pine (500–320 BC) from the Laboratory of Tree-Ring Research at The University of Arizona. We isolated contemporaneous Irish oak samples and converted the carbon from the bristlecone pine and the Irish oak to benzene on our chemical rigs (Pearson 1984; McCormac, Kalin and Long 1993). We processed and counted the nine sample pairs quasi-simultaneously to determine the activity in each and applied $\delta^{13}C$ corrections to the samples. Table 2 shows the results of the 18 measurements. We observed a consistent offset of −41 ± 9.2 yr between the bristlecone pine and the Irish oak, with the bristlecone pine giving the older dates.

Table 2 also shows results from Pearson *et al.* (1986) for the same period. Samples 415–325 BC inclusive from Pearson *et al.* (1986) are interpolated from the original bidecadal measurements. The offset between the recent measurements on the Irish oak and interpolated values from Pearson *et al.*

(1986) is 10.6 ± 8.2. In addition to our own measurements, we located other studies in the literature where measurements of contemporaneous European oak and American wood were made in the same laboratory. A distinct advantage of these intralaboratory studies is that none are subject to laboratory offsets and they should provide results that should give correct relative ages. In all but one case, that of de Vries (1958), the American wood was depleted in ^{14}C with respect to the European wood (Table 3).

TABLE 3. Intralaboratory Comparisons of Wood from Different Regions in the Northern Hemisphere

Source	Difference	No. of sample pairs
de Vries (1958) fractionation corrected by Lerman *et al.* (1970)	German oak similar in activity to contemporaneous wood from Colorado, USA	2
Suess (1970)	French oak *ca.* 50 yr younger than bristlecone pine	12
Stuiver and Quay (1981)	English oak 16 ± 9 yr younger than Douglas-fir	3
Stuiver (1982)	German oak 23 ± 6 yr younger than sequoia	12
This paper	Irish oak 41 ± 9 yr younger than bristlecone pine	9

Stuiver (1982) compared calibration chronologies that were being developed in the high-precision laboratories. The results he obtained for the intralaboratory comparison of sequoia and German oak (Table 3) gave a value of −22.9 ± 5.6 yr. This value on 12 sample pairs differs from our findings, placing German oak younger than Irish oak (Fig. 5A). Possible explanations for this include: 1) regional differences may vary with time; 2) identical pretreatment methods may have resulted in a variable end product on the different wood species; or 3) dendrochronological problems are possible with one or both of the woods used.

Of these possibilities, the dendrochronology associated with the German oak samples used for this comparison is questionable. The relevant chronology of Becker and Delorme (1978) was linked to that of Hollstein at the time these samples were provided. Subsequently, the Hollstein chronology had to be moved by 26 yr toward the present (Hollstein 1980; Baillie 1995), meaning that calendar ages used by Stuiver (1982) for wood from this period may originally have been incorrectly dated. Correcting such a tree-ring error would have the effect of moving the calendar dates toward the present: for example, a sample accepted by Stuiver as dating to 585 AD might actually relate to 611 AD. The possibility of a dendrochronological error was mentioned by Stuiver (1982: 4) when he stated, "Further work is needed for a confirmation of the suspected systematic difference (which, if real, also could be caused by errors in the dendrochronology)". Thus, the ^{14}C age measured for 585 AD could be too young by *ca.* 38 yr (this value was calculated using the data in Stuiver and Becker 1986). Stuiver (1982) also mentions that because they biased sample selection toward those showing the largest difference in the Seattle/Heidelberg comparison, the 23-yr offset that they measured may be a maximum value. The Heidelberg German oak measurements given in Stuiver (1982) are likely to have had the same problem and, additionally, may have had a laboratory bias of *ca.* 35 yr toward younger dates.

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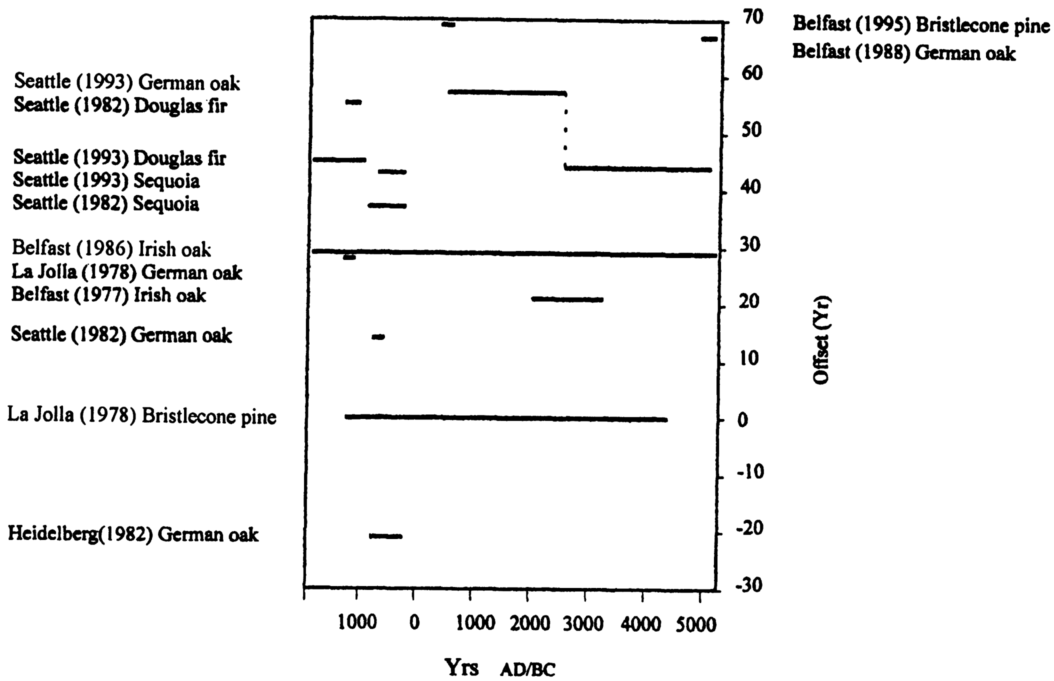

Fig. 5A. Schematic showing offsets from measured data (this paper) and previously reported data

Table 2 in Stuiver (1982) also shows a small offset (4.4 ± 3.9 yr) between 23 sample pairs from Belfast Irish oak and Seattle Douglas-fir. If the Pearson *et al.* (1986) data are correct, then the Seattle dates, which presumably require a radon correction, should move to a position *ca.* 15 yr older than the Belfast dates. The same table gives a value for the offset between Seattle Douglas-fir and La Jolla bristlecone pine (Suess 1978) of –55.3 ± 9.7 yr, with Seattle results being the older. Comparison of the Belfast 1986 data with the Suess (1978) data gives a value of –27 ± 9.1 yr, with Belfast results being older. There is thus a difference of 28 yr between the Belfast and Seattle comparisons with the bristlecone pine, which would be partially compensated for by a radon correction in the Seattle data. Further, Linick, Suess and Becker (1985) recalculated the Suess (1978) data using 95% of the activity of the original NBS oxalic acid standard as a reference, which resulted in a shift of the bristlecone pine dates by 48 yr toward older values. This places the bristlecone pine chronology 21 yr older than the Irish oak and within 7 yr of the Douglas-fir measurements of Stuiver (1982). The comparison of Seattle Douglas-fir with La Jolla bristlecone pine and La Jolla German oak indicates that the German oak dated in La Jolla was 28 yr older than the La Jolla bristlecone pine measurements. This places the German oak measured at La Jolla at 49 yr older than Irish oak.

Figure 5A summarizes the offsets described above. Using the bristlecone pine chronology of Suess (1978) as a zero reference, subsequent chronologies can be compared. Many of the papers published since 1978 describe the offset of their chronology compared to that of Suess (1978); in other cases, an offset can be calculated based on the measurements provided. Figure 5A uses measurements made by us and data listed in Pearson *et al.* (1977), Suess (1978), Stuiver (1982), Linick, Suess and Becker (1985), Pearson *et al.* (1986) and Stuiver and Becker (1993) and references therein to show the relative positions and periods of overlap of intralaboratory comparisons and chronologies. The

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dotted line joining the two segments of the Stuiver and Becker (1993) German oak data reflects the extension of the Stuiver and Becker (1986) from *ca.* 2500 to 6000 BC.

In Figure 5B, the bristlecone pine measurements (italicized) have been adjusted to account for the 48-yr adjustment given in Linick, Suess and Becker (1985). The consequences of the move are that the Seattle Douglas-fir (Stuiver 1982) and bristlecone pine have a relative offset of 7 yr and the La Jolla German oak measurements are 49 yr older than the Irish oak. The Seattle and Heidelberg German oak from 1982 measurements date younger than all others, but as we have explained, these may have a tree-ring error of 26 real years, which means that both need to be adjusted back in time by *ca.* 38 (^{14}C) yr (Fig. 5C, italicized). Figure 5C shows the relative positions of intralaboratory comparisons and chronologies with all known or suggested corrections applied. Ignoring the two extreme sets of values (*i.e.*, the La Jolla and early Heidelberg German oak measurements) means that the wood measurements, from within the northern hemisphere, lie within a 50-yr range, confirming that intrahemispheric and localized regional offsets are small. Nonetheless, the data appear to show a finite difference between Irish and German/American wood.

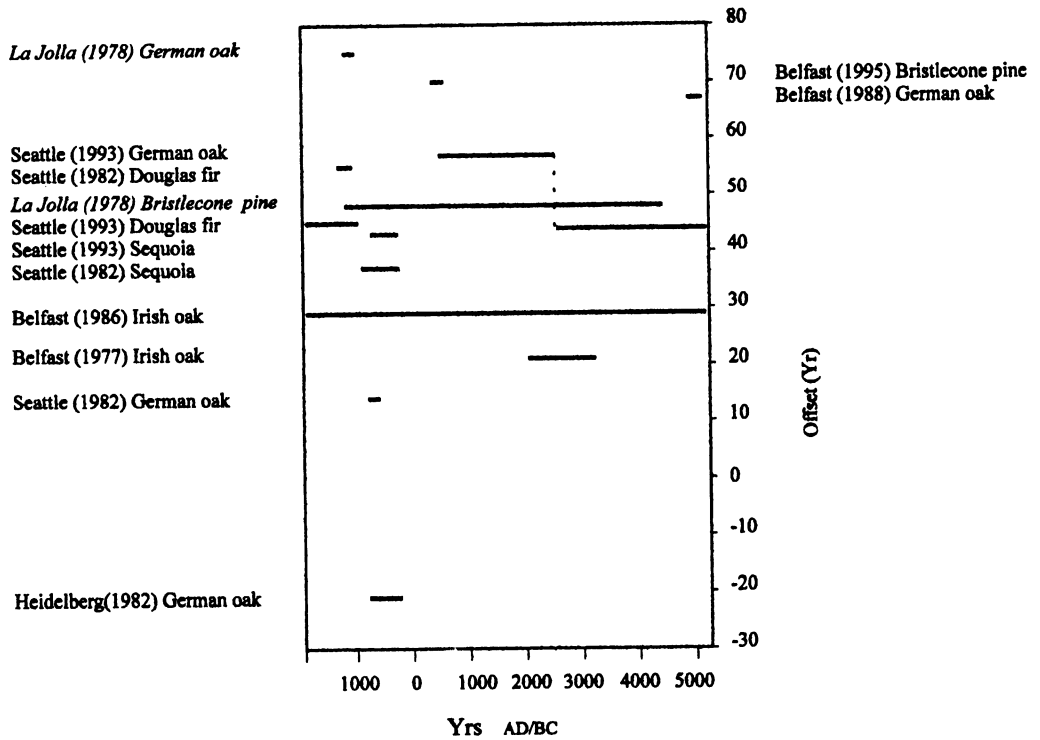

Fig. 5B. Data after adjustment of the La Jolla bristlecone pine and German oak measurements by 48 yr

Stuiver and Pearson (1993) reviewed systematic differences between laboratories for the 1993 calibration data. Interlaboratory comparisons using sections of the German oak chronology showed good agreement between participating laboratories, with the exception of Belfast and Seattle results for 5500–6000 BC, where Belfast was older by 15 ± 4 yr, and 5180–5500 BC, where Belfast dates were older by 54 ± 5 yr with respect to Seattle. Heidelberg dates for the intervals 4075–5265 BC and 5805–5995 BC were 41 ± 4 yr older than the Seattle dates. Stuiver and Pearson also compared German oak and bristlecone pine over the restricted interval 5680–5810 BC between several laborato-

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ries. The conclusion from this intercomparison was that bristlecone pine and German oak give the same ages over this interval.

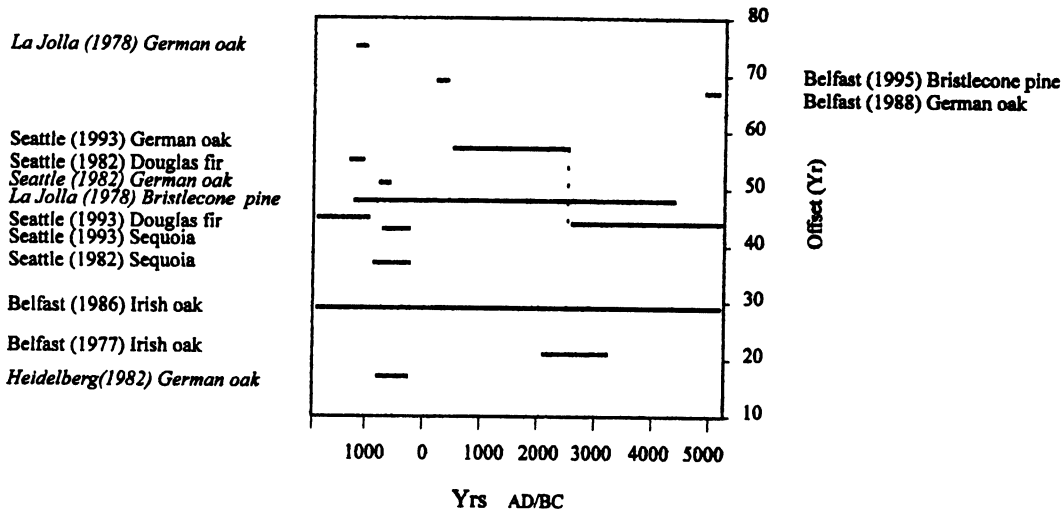

Fig. 5C. Data after adjustment of the La Jolla bristlecone pine measurements and adjustment of the German tree-ring dates forward in time by 26 yr

Conclusion

We have presented a set of results indicating that the ^{14}C activity of contemporaneous wood from different locations may not be the same at all times. The available intralaboratory measurements (for restricted periods) indicate that American wood yields dates that are older than their Irish equivalents. The recent shift by Stuiver and Becker (1993) of the Stuiver and Becker (1986) data to slightly older dates, and the lack of justification for the Pearson, Becker and Qua (1993) adjustment that would result in the realignment of the two data sets, supports the offset suggestion. Measurements made in Belfast, measurements from La Jolla (reported in Stuiver 1982) and differences observed in the calibration data published in Pearson *et al.* (1986) and Stuiver and Becker (1993) suggest that an offset exists between German oak and Irish oak of about 30 yr, with German oak giving the older dates. The extension of the German chronology by Stuiver and Becker (1993) suggests a time-varying offset between the two. German oak, bristlecone pine, Douglas-fir and sequoia all give ^{14}C dates that are older than Irish oak for the periods that have been measured.

We conclude that published data reveal evidence for small location-dependent ^{14}C differences. The cause of the variation between American (exclusively western) and Irish wood could be related to the influence of the oceans. Alternatively, local effects may dominate and bog oaks may differ in ^{14}C content from oaks growing near large river systems. If the indicated ^{14}C differences are real, hemispheric uniformity can no longer be assumed and location-dependent high-precision calibration curves would be necessary if not for routine archaeological dating then at least for high-precision wiggle matching. However, since the indicated magnitude of the activity difference is very small, calibrated dates would be altered by tens of years or less, thus making no significant difference to the archaeological interpretation of most if not all dates that have been calibrated. Given the complexity of the suggested ^{14}C variations in the northern hemisphere, it is imperative that future calibration efforts ensure intralaboratory measurements of the woods from different regions and, preferably, replication between laboratories. Only in this way will small regional differences be quantified.

Acknowledgments

The authors would like to thank personnel at the tree-ring laboratories in the University of Arizona and Queen's University, Belfast for making wood samples available.This work was supported by a research grant from the Royal Society to Dr. F. G. McCormac (Grant Number 403-010-0001).

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https://doi.org/10.1017/S0033822200030873 Published online by Cambridge University Press