# Radiocarbon

1982

# A HIGH-PRECISION CALIBRATION OF THE AD RADIOCARBON TIME SCALE

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ABSTRACT. A high-precision calibration curve, derived from the radiocarbon age determinations of 195 decade samples spanning the AD 1 to 1950 interval, is presented.

Though derived for the Pacific Northwest and California, the curve can be used for a large part of the northern hemisphere. This is proven by the radiocarbon ages of contemporaneous sample pairs which are, in most instances, identical within the quoted precision. Two sets of single-year data reveal no evidence for an 11-year cycle with an amplitude beyond the 12-year measuring precision. This indicates that the calibration curve is also applicable for single-year <sup>14</sup>C samples.

Analysis of the Seattle data sets and comparison with those published by the Belfast, La Jolla, and Heidelberg laboratories show that the total variability in a radiocarbon age determination is often larger than that predicted from the quoted errors. Upper limits for the error multiplier (*ie*, the factor with which the quoted error has to be multiplied to obtain the overall laboratory variability) are estimated at 1.5 for Seattle and Belfast, 1.1 to 1.4 for La Jolla, and 2.0 for Heidelberg.

The comparisons with Belfast, La Jolla, and Heidelberg also reveal offsets with the Seattle calibration curve of, respectively, 4, 27 to 55, and 58 years. These offsets are most likely due to laboratory bias. An improvement of the present calibration curve by combining data sets from other laboratories will only be possible when offsets and error multipliers are precisely known through interlaboratory calibration.

# INTRODUCTION

The basic information contained in a sample submitted for radiocarbon dating is the remaining present-day <sup>14</sup>C activity. A conventional radiocarbon date (Stuiver and Polach, 1977) is derived from this information by comparing the present-day sample activity with an atmospheric <sup>14</sup>C level which is assumed to have been constant in the past. Past atmospheric <sup>14</sup>C levels have fluctuated, however, and as a result, a radiocarbon age is only an approximation of the historical age expressed in calendar years.

Past atmospheric <sup>14</sup>C levels are recorded in trees because carbon, derived from atmospheric carbon dioxide through photosynthesis, is incorporated into their cells. The cell-wall cellulose formed each year during the growing season has a <sup>14</sup>C content that reflects the atmospheric <sup>14</sup>C content of that year. After fractionation of the isotopes has been taken into account through normalization procedures (Stuiver and Polach, 1977), the original atmospheric <sup>14</sup>C level can be calculated from the measured <sup>14</sup>C tree-ring activity. This procedure is the reverse of <sup>14</sup>C dating because the age of the material is known through dendrochronological means and tree-ring counting, thus enabling the researcher to correct for the <sup>14</sup>C decay that took place after the tree ring was formed.

Radiocarbon ages were determined for dendrochronologically dated wood of each decade of the AD 1 to 1950 interval (where the first "decade"

covers 9 years (AD 1 to 9)). For perfectly dendrochronologically dated wood, the number of "dendroyears" is equal to the number of calendar years. When constructing a calibration curve, the years are given as dendroyears in order to indicate the possibility of errors in the tree-ring count. Such errors in the dendro-age are probably no more than a couple of years.

#### DENDROCHRONOLOGY

The trees used for the radiocarbon age determinations were either Douglas Fir (*Pseudotsuga menziesii*) from the Pacific Northwest, or Sequoia (*Sequoiadendron giganteum*) from California. These trees, with the corresponding dendro-age intervals used for construction of the time-scale calibration curves, are listed in table 1.

The Sequoia trees were cross-dated by H Garfinkel of the University of Washington with the Sequoiadendron master chronology (Douglas, 1919). The Douglas Fir from Vancouver Island (used for the AD 730 to 1320 interval) was collected and cross-dated by M L Parker, L Joza, and P Bramhall of the Western Products Forestry Laboratory in Vancouver, British Columbia. The ages of the rings of the post AD 1320 Douglas Firs were all determined by tree-ring counting. The well-developed rings of these trees were easily counted, and errors in this part of the chronology are unlikely (pers commun, H C Fritts, after inspection of the trees).

The determination of radiocarbon ages of tree sections that overlap in dendro-age provides a check on the chronology. For instance, the four-decade sample pairs of common dendro-ages of the two Sequoia trees (AD 230 to 270 interval) differ in radiocarbon age by  $12\pm22$ ,  $18\pm18$ ,  $22\pm17$ , and  $-47\pm22$  years. The mean weighted age difference for the four samples is a negligible 5 radiocarbon years. An even better test is provided by the 14 contemporaneous samples of the youngest portion of the Sequoia chronology, and the oldest part of the Douglas Fir chronology (AD 730 to 940 interval). The decadal samples of the Sequoia chronology yield radiocarbon age  $R_s$  (x), where x is the dendro-age. The 14 Douglas Fir radiocarbon ages  $R_f$  are compared to the  $R_s$  (x) curve by taking the square root of the average quadratic deviation

$$\sqrt{\Sigma(R_{\rm f}(x+\Delta x)-R_{\rm s}(x))^2/N}$$

where N is the number of samples being compared. We calculated this radiocarbon age "difference" by assuming the dendro-ages of the Douglas Fir to be accurate ( $\Delta x = 0$ ), or to be offset by systematic age differences

TABLE 1

Tree species	Lab code	Location	Dendro-age used		
Douglas Fir	С	47°46′N, 124°06′W	AD 1910 - 1950		
Douglas Fir	Α	47°46′N, 124°06′W	AD 1820 - 1910		
Douglas Fir	$\mathbf{F}$	43°07′N, 123°40′W	AD 1510 - 1820		
Douglas Fir	R	~47°N,~122°W	AD 1320 - 1510		
Douglas Fir	S	48°40′N, 123°40′W	AD 730 - 1320		
Sequoia	RC	~36.5°N, 118.5°W	AD 230 - 940		
Sequoia	SR	~36.5°N, 118.5°W	AD 1 - 270		

of 1, 2, etc years ( $\Delta x = \pm 1, \pm 2...$ ). The best fit for the Douglas Fir chronology is obtained for an offset of 4 years only (see fig 1).

The quadratic term (standard deviation of the  $R_f$  radiocarbon ages around curve  $R_s$  (x)) is 22 years minimally (fig 1). This age is, as expected, of the same magnitude as the average quoted precision of 15 years for the radiocarbon ages. For a  $2\sigma$  (30 years) cut-off, the matching of the Sequoia and Douglas Fir dendro-ages is fixed within -8 and +6 years (see fig 1).

# **TECHNIQUE**

A new underground <sup>14</sup>C facility was constructed in 1972, as part of the Quaternary Research Center at the University of Washington. The laboratory uses the code QL (Quaternary Isotope Laboratory) for the reporting of radiocarbon dates. High-precision <sup>14</sup>C tree-ring measurements started in December 1973 with a 4.5-liter copper CO<sub>2</sub> gas counter. The background counting rate of this counter is 1.6 cpm, and oxalic acid count rate is 90 cpm (Stuiver, Robinson, and Yang, 1979). Three slightly larger counters (oxalic acid counting rates up to 100 cpm) were added in 1976 and 1977. The data reported here were obtained during several years of measurements with these four counters. A typical precision for a four-day measurement is 15 radiocarbon years.

Additional details of the analytical procedures were described previously (Stuiver, 1978; Stuiver and Quay, 1980; 1981). Cellulose was prepared for 20th century samples, whereas for the older samples, the De Vries wood preparation method was utilized. The error introduced

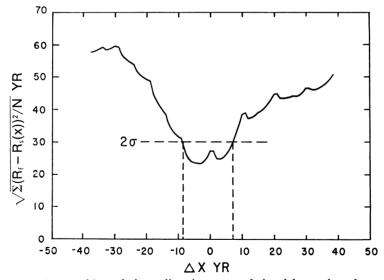


Fig 1. The matching of the radiocarbon ages of decadal samples of two trees (Sequoiadendron giganteum and Pseudotsuga menziesii). The ordinate gives the square root of the average quadratic deviation in radiocarbon ages. The deviation was calculated by assuming no error in the dendrochronology ( $\Delta x = 0$ ), or a shift of  $\Delta x$  years between both chronologies. The best fit is obtained for a shift of 4 years.

in the radiocarbon age by using the De Vries method amounts to approximately three radiocarbon years maximally (Stuiver, 1978; Stuiver and Quay, 1980; 1981).

#### GLOBAL VALIDITY OF CALIBRATION CURVES

A time-scale calibration curve is universally acceptable only if regional (latitudinal and longitudinal) differences in atmospheric <sup>14</sup>C levels can be neglected. Any regional difference in <sup>14</sup>C level, if constant, would introduce systematic age differences between contemporaneous sample pairs. Such postulated age differences can be evaluated by comparing the radiocarbon ages of wood samples of the same dendro-age. A complicating factor in such an investigation is possible biases in the laboratory measurements which also introduce offsets between data sets (Scott, Baxter, and Aitchison, in press).

The age differences of sample pairs measured in the same laboratory will not be influenced by laboratory bias if such bias is constant with time. Most of the comparisons made in this section are for measurements made in a single laboratory (Quaternary Isotope Laboratory) during a seven-year interval. The radiocarbon ages obtained by Pearson of the University of Belfast are also suitable for comparison with the Seattle data because international calibration efforts show the absence of systematic radiocarbon age differences between the Seattle and Belfast laboratories (Scott, Baxter, and Aitchison, in press; Mann, pers commun, 1980).

A comparison of Irish Oak (Pearson, 1980; pers commun, 1981) and Pacific Northwest Douglas Fir radiocarbon ages of 53 sample pairs with dendro-ages between AD 955 and 1840 yields a mean age difference of  $2\pm3$  radiocarbon years. The standard deviation in the age difference is based on the quoted errors in the radiocarbon age measurements. Similarly, the mean radiocarbon age difference of the previously discussed 14 sample pairs of identical dendro-age of one of our Sequoias (36.5°N, 118.5°W), and the oldest Douglas Fir (48°40'N, 123°40' W), is  $9\pm5$  years.

Additional sample pairs measured in the Quaternary Isotope Laboratory, listed in Stuiver and Quay (1981) yield a mean age difference of  $16 \pm 9$  years for 3 sample pairs between 1829 and 1844 of English Oak (51°48′N, 2°37′W) and Douglas Fir (47°46′N, 124°06′W), of  $0 \pm 12$  years for two samples of Bristlecone Pine (36°N, 118°W) and Douglas Fir (47°N, 122°W), and of  $7 \pm 15$  years for a single pair of Douglas Fir samples from 47°46′N, 124°06′W, and 32°23′N, 110°41′W. For none of these samples do we find differences beyond twice our quoted measuring precision.

We do find a systematic difference of  $23 \pm 6$  radiocarbon years between 12 sample pairs of German Oak and Douglas Fir wood. However, as discussed in the section on interlaboratory time-scale comparisons, we may have been biased in our sample selection towards the largest possible age discrepancies. Further work is needed for a confirmation of the suspected systematic difference (which, if real, also could be caused by errors in the dendrochronology).

The above-cited radiocarbon age differences of contemporaneous sample pairs from different regions are not statistically significant (except for the latter case), proving that regional differences in atmospheric <sup>14</sup>C concentration, if they exist at all, are small. Thus, the time-scale calibration curves can be used at least for a large part of the northern hemisphere. Systematic radiocarbon age differences for southern hemispheric samples still have to be tested in more detail. An upper limit appears to be the 32-year radiocarbon age difference given by Lerman for South American wood (Lerman, Mook, and Vogel, 1970).

#### TIME-SCALE CALIBRATION

The <sup>14</sup>C determinations of 195 decades were used for the construction of the calibration curve (fig 2). Calibration data can be plotted in various ways. A normal orthogonal orientation is given in figures 2 and 3 where dendroyears are directly compared to radiocarbon years. Figure 4 plots the age anomalies versus radiocarbon age. This gives the appearance of a "tilting" of the calibration curve.

For conventional radiocarbon dating, the zero year BP is AD 1950. The dendro-age, T, corresponding with the radiocarbon age, R, is also given in years BP, and is equal to the number of calendar years before AD 1950 if the tree-ring determination is without error. The correction to be applied to the radiocarbon age is  $\Delta t$  years, as listed in the vertical axis of figure 4. Thus, dendro-age BP (calendar years before AD 1950 for perfect tree-ring counts) = radiocarbon age plus  $\Delta t$ , or  $T = R + \Delta t$ .

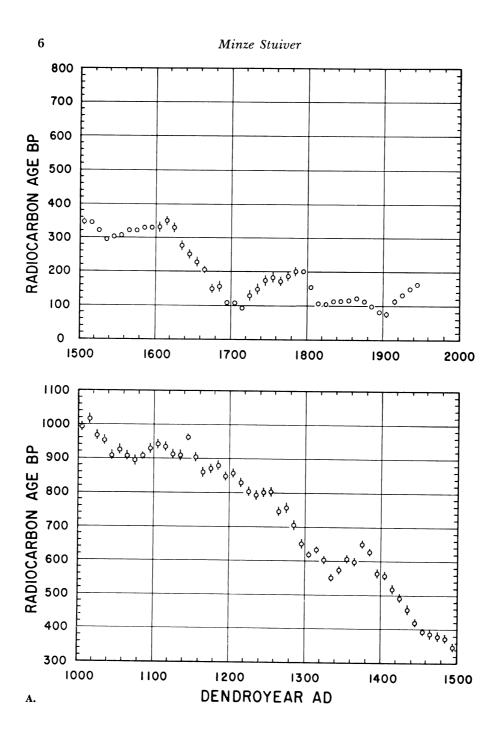
Compared to dendro-ages, the radiocarbon ages are too young for positive  $\Delta t$ , and too old for negative  $\Delta t$ . Thus, the radiocarbon ages between 700 and 2000 yr BP are mostly too old, whereas many of the 0 to 700-year BP dates are too young. Multiple intersections of the horizontal (fig 2) or vertical (fig 4) line drawn through a given radiocarbon age result in multiple dendroyear ages.

The standard deviation of each <sup>14</sup>C activity determination is 16 years or less. For parts of the curve, single years were dated and averaged for the decade. Here, the standard deviation in the radiocarbon age approximates four radiocarbon years.

The errors (one standard deviation) given in the figures were derived from the Poisson counting statistics of the sample and standard activities. The effect of additional errors is to enlarge the long-term overall laboratory error. In the following sections, the possibility of a larger error is discussed. It will be shown that the errors in the figures, although probably somewhat underestimated, are a fairly realistic estimate of the overall variability in the measurement.

#### AGE CONVERSION

Because the past atmospheric <sup>14</sup>C levels are variable, it is possible for samples with differing dendro-ages to have identical present-day <sup>14</sup>C activities, and thus, identical conventional radiocarbon ages. This case is illustrated in figure 5 where the <sup>14</sup>C decay of a sample formed in 1690 is followed. The exponential decay of <sup>14</sup>C is, as a first approximation,



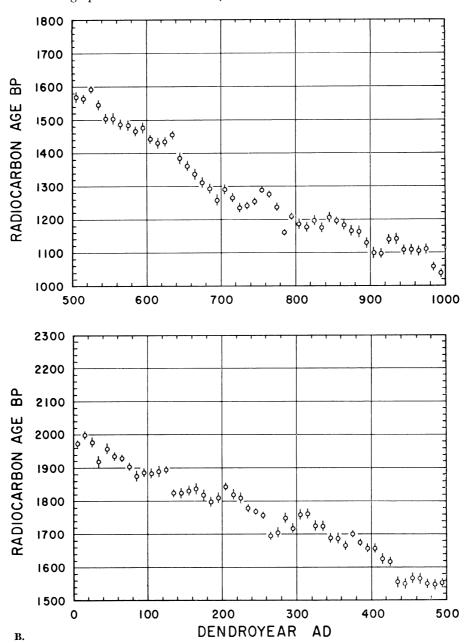
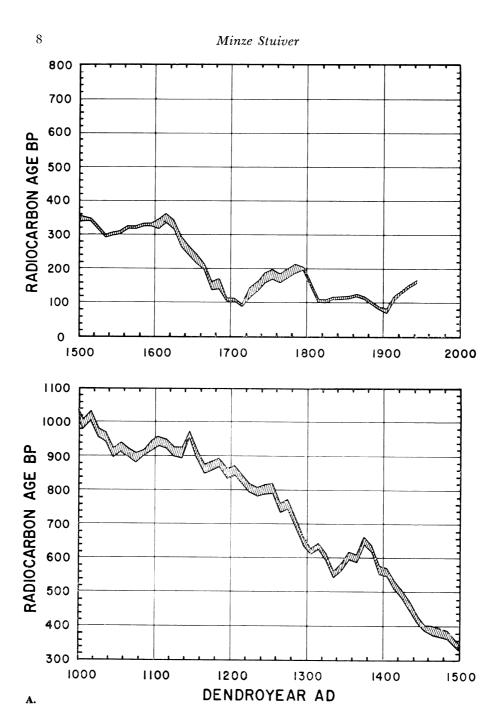


Fig 2A, B. Radiocarbon age versus dendro-age of decadal samples of Pacific Northwest Douglas Fir and California Sequoia. The number of dendroyears was dendrochronologically derived. Dendro-ages are equal to calendar years AD for perfect ring counts. The ordinate gives the conventional radiocarbon age in years BP (before 1950). The vertical error bars are one standard deviation in the measurement, based on counting statistics only. The standard deviation is about equal to the radius of the open circles when error bars are missing. The overall laboratory variability may be up to 1.5 times the given standard deviation (see text).



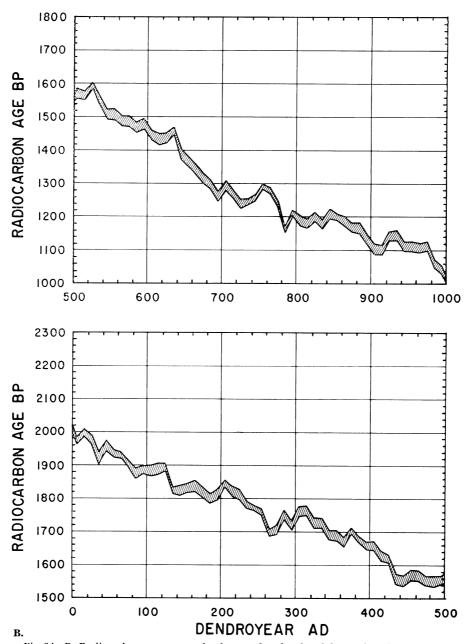


Fig 3A, B. Radiocarbon age versus dendro-age for the decadal samples plotted in figure 2. The shaded area represents one standard deviation (derived from counting statistics only) on each side of the measured radiocarbon ages.

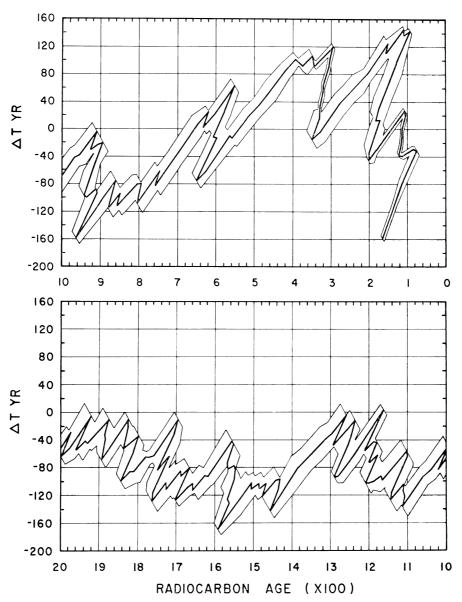


Fig 4. The correction  $\Delta t$  (years) to be applied to a conventional radiocarbon date in order to obtain a dendro-age in years before AD 1950. The center line was constructed from the data in figure 2. The width of the "open" area is twice the standard deviation in the radiocarbon age (one sigma on each side). The standard deviation was derived from counting statistics only.

linear over short time intervals (solid "straight" line in figure 5). The  $\Delta^{14}$ C values given in the figure are the relative deviations of the measured <sup>14</sup>C activities, after correction for age and isotopic fractionation, from the National Bureau of Standards (NBS) oxalic acid 14C activity. The  $\Delta^{14}$ C contents of samples formed in the years 1729, 1809, and 1927 fall on the 14C decay line as well. Thus, all four samples, each formed in equilibrium with atmospheric  $\Delta^{14}$ C levels prevalent during their time of formation, will end up with the same <sup>14</sup>C deficiency of 16 per mil in the year 1950. This  $\Delta^{14}$ C level (measured in later years but age-corrected back to the year 1950) is the only available information we have from the <sup>14</sup>C counts, and results in a radiocarbon age of 130 years ("AD 1820") for all four samples. The calculation of a conventional radiocarbon age is based on the 5568-year half-life, which is different from the 5730-year half-life used for the actual decay; hence, the different decay line (dashed) that yields the radiocarbon age when intersecting the assumed constant  $\Delta^{14}$ C level (the horizontal zero axis in figure 5).

When converting from a radiocarbon age to a dendro-age, the error in the radiocarbon measurement, as illustrated in figure 5, has to be considered also. Whereas the error in the radiocarbon age is normally symmetrical around the age, such is not true for the derived dendro-age. Thus, the  $130 \pm 32$  years BP radiocarbon age is equivalent with dendroages of  $260 \pm 20$ , 221 + 10 - 14, 141 + 9 - 72, and 23 + 15 - 23 years before 1950.

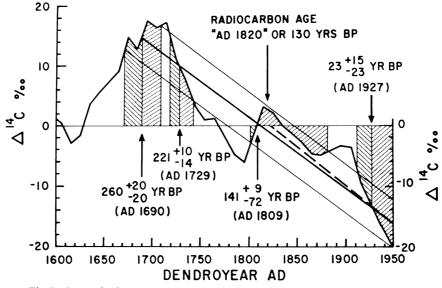


Fig 5. Atmospheric  $\Delta^{14}C$  levels, expressed as per mil deviation from NBS oxalic acid standard <sup>14</sup>C activity, of the AD 1600 to 1900 period. The heavy line follows the radioactive decay of a sample formed in the year AD 1690. All four samples listed in the figure have, after decay, a 16 per mil <sup>14</sup>C deficiency by AD 1950, and thus, the same radiocarbon age. The error (4 per mil, or 32 years for the radiocarbon age) is also illustrated for each sample. The radiocarbon age is the intercept of the  $\Delta^{14}C=0$  axis with the dashed line (see text).

When the <sup>14</sup>C levels decrease parallel to the decay line, the errors are increased, whereas a smaller dendro-age error occurs when the <sup>14</sup>C increases or the change is fast. It is, therefore, possible that a radiocarbon age error decreases when converted into a dendro-age error.

When calculating the dendro-age errors, the uncertainty in the calibration curve should be considered. The following steps are recommended:

- 1) Estimate the width, in radiocarbon years, of the calibration curve in figure 3 for the age range to be considered. Divide this number by two to obtain the average standard deviation of the calibration curve ( $\sigma_{\rm cal}$ ).
- 2) Use figure 2 to convert to a dendro-age, with the sample-age error equal to  $\sqrt{\sigma^2_{\text{sample}} + \sigma^2_{\text{cal}}}$ .

For a typical routine radiocarbon age determination with an age error of about 50 years or more, the increase in error due to the uncertainty of the calibration curve is negligible. For instance, for  $\sigma_{\text{sample}} = 60$  years and  $\sigma_{\text{cal}} = 15$  years, the square root term is 62 years. Of course, the influence of the uncertainty in calibration becomes more important when the quoted radiocarbon age error approaches the average error of the calibration curve.

### SINGLE-YEAR AGES VERSUS DECADE AGES

The calibration curves are based on radiocarbon ages of decadal samples. Radiocarbon ages of samples grown in single years (such as seeds,

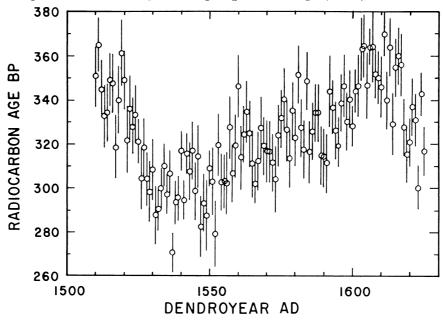


Fig 6. Radiocarbon ages of single-year Douglas Fir samples between AD 1510 and 1625. Vertical bars denote one standard deviation. The variability of the single-year radiocarbon ages around the decadal means does not exceed the quoted precision (see text).

leaves, etc) could deviate from the calibration curves. Such anomalies could possibly be introduced by the 11-year modulation of the cosmic ray flux, and upper atmospheric <sup>14</sup>C production. Some recent studies of the modulation of 19th and 20th century atmospheric <sup>14</sup>C levels indicate 11-year variability with either about 8 radiocarbon years or 33 radiocarbon years amplitude (Stuiver and Quay, 1981; Burchuladze, Pagava, and Povinec, 1980).

Two sets of single-year data were measured. They span the AD 1510 to 1625 and AD 1820 to 1952 intervals. A few points of the latter interval comprise 2- or 3-year samples (see table in Stuiver and Quay, 1981). The radiocarbon ages of the single-year measurements are given in figures 6 and 7.

As will be shown in the following section, the scatter of the single-year data around the decade average trend is entirely compatible with the scatter expected solely from the quoted errors in the single-year measurements. The curves in figures 2 to 4 can, therefore, also be used for radiocarbon ages of single-year samples. The resulting dendro-ages are equally accurate as those obtained for samples covering an entire decade.

Spectral analysis of the data given in figures 6 and 7 does not yield any evidence for an 11-year cycle with an amplitude beyond the 12-year measuring precision.

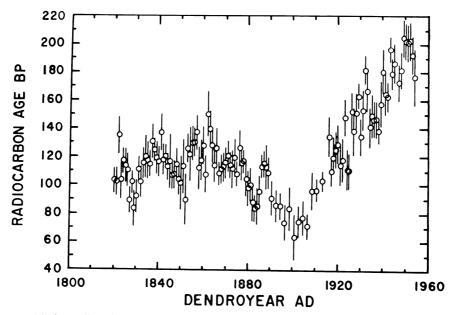


Fig 7. Radiocarbon ages of single-year Douglas Fir samples of the AD 1820 to 1954 interval. Vertical bars denote one standard deviation (derived from counting statistics only). The variability of the single-year radiocarbon ages around the decadal means does not exceed the quoted precision (see text).

#### INTERLABORATORY TIME-SCALE COMPARISONS

To establish the curves in figures 2 to 4 as an absolute radiocarbon age-correction curve, the possibility of a systematic difference in the measured radiocarbon ages has to be explored. Likewise, the contributions of factors other than counting statistics to the overall age variability has to be assessed to estimate the uncertainty in the calibration curves. These two goals can be achieved by interlaboratory comparisons combined with careful analysis of the Seattle data base.

G W Pearson (1980) recently compared the radiocarbon ages of 23 samples of Irish Oak with the previously published AD 1450 to 1950 portion (Stuiver, 1978) of the calibration curve of figure 2. The differences between the Irish and North American results were small, and Pearson concludes that the results

show remarkable agreement, especially when considering that the two techniques — liquid scintillation versus gas counting, and two tree species — oak versus pine were used, and the samples were from Ireland and America, respectively (Pearson, 1980).

The complete set of published Belfast determinations (AD 1400 to 1950) and the Seattle results are given in figure 8.

Plots of radiocarbon ages of Seattle Douglas Fir and Sequoia versus La Jolla Bristlecone Pine (Suess, 1978) are given in figures 9A and B. Suess' measured AD samples span the AD 1 to 500 and AD 700 to 1300 intervals. For both intervals there is good agreement between the basic trends, but the Bristlecone Pine radiocarbon ages have a tendency towards

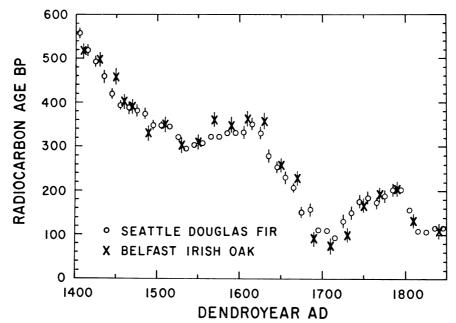


Fig 8. A comparison of Seattle Douglas Fir  $(\bigcirc)$  and Belfast Oak  $(\times)$  radiocarbon ages. Vertical bars represent the quoted standard deviation.

younger ages. This is also true for the La Jolla German Oak results (Suess, 1978) of the AD 1100 to 1300 interval (fig 9C).

For the AD 250 to 750 interval, Heidelberg German Oak determinations are available (Bruns, Münnich, and Becker, 1980). Again we note agreement in trends between the Seattle and Heidelberg data (fig 9D). There is, however, a clear offset in the Heidelberg data towards younger radiocarbon ages. This has been corrected for in figure 9D by adding 58 years to all Heidelberg ages.

The wood used for the radiocarbon age determinations of the Seattle laboratory always covers one decade (for instance, AD 1720 to 1729). Samples from the comparison laboratories, however, may cover a time span of a few years to two decades. For the following statistical analysis, we neglected the difference in time span of each individual sample, and only considered the midpoint in dendro-age.

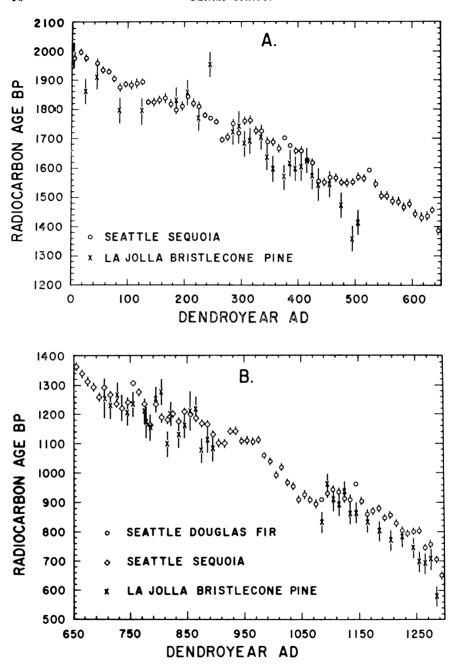
The Seattle data are evenly spaced with dendro-age midpoints in the middle of each decade. The samples to be compared with the Seattle data set (comparison samples) are treated as follows: If the midpoint dendro-age of the comparison sample differs less than 2.5 years from a Seattle midpoint, the difference,  $x_i$ , in radiocarbon age is calculated. If the age difference is more than 2.5 years, the radiocarbon age of the comparison sample is evaluated against a Seattle radiocarbon age obtained by averaging the radiocarbon ages of the two relevant consecutive decade samples. Therefore, the midpoints of the dendro-ages of the Seattle and comparison samples never differ by more than 2.5 years.

With N being the number of comparisons made, each yielding a radiocarbon age difference of  $x_i \pm \sigma_i$  years, the mean difference,  $\tilde{x}$ , in radiocarbon ages is given by  $\tilde{x} = \sum x_i W_i / \sum W_i$  with  $W_i = \frac{1}{\sigma_i^2}$ . The calculated mean radiocarbon age differences are given in table 2.

A very small radiocarbon age difference (4.4 years) is found for the Seattle and published Belfast data set. These laboratories also participated in the calibration of the new NBS oxalic acid standard (Mann, pers commun), and in the Glasgow Calibration Project (Scott, Baxter, and Aitchison, in press). These efforts showed the absence of systematic differences in the Belfast and Seattle radiocarbon ages. Evidently, the Irish Oak and Pacific Northwest Douglas Fir have nearly identical <sup>14</sup>C levels (and radiocarbon ages).

Additional Belfast results, back to AD 955, were recently made available for comparison by G W Pearson of the University of Belfast. Again, excellent agreement is obtained, with a mean age difference of  $2 \pm 3$  radiocarbon years between the complete data sets of 53 sample pairs (AD 955 to 1840).

The radiocarbon ages of the Seattle Douglas Fir are, on average, 55 years older than the La Jolla Bristlecone Pine ages. The radiocarbon ages of two Bristlecone Pine samples (AD 1080 to 1090 and AD 1480 to 1490), determined in the Seattle laboratory (Stuiver and Quay, 1981), differ in age from the corresponding Douglas Fir samples by  $+10 \pm 14$  and  $-10 \pm 14$ 



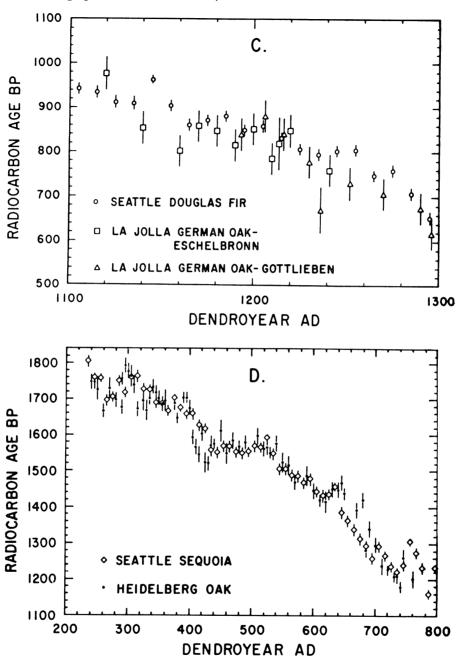


Fig 9. A) Seattle Sequoia (○) and La Jolla Bristlecone Pine (×) radiocarbon ages for the AD 1 to 650 interval. Vertical bars represent the quoted standard deviations in the age determinations. B) Seattle Sequoia (◇), Seattle Douglas Fir (○) and La Jolla Bristlecone Pine (×) radiocarbon ages for the AD 650 to 1250 interval. C) Seattle Douglas Fir (○), and La Jolla German Oak (□ and △) radiocarbon ages between AD 1100 and 1300. D) Seattle Sequoia (◇) and Heidelberg Oak (\*) radiocarbon ages between AD 200 and 800. All Heidelberg radiocarbon ages were increased by 58 years.

18 years. These intralaboratory results support the idea that systematic age differences between Bristlecone Pine and Douglas Fir are less than the 55 years calculated from the La Jolla-Seattle data sets. The difference is, therefore, more likely to result from a difference in laboratory calibration. A complicating factor in evaluating this difference is the absolute calibration of the La Jolla data. Suess (1978) mentions the possibility of uncertainties in the <sup>14</sup>C contents of the wood standards used prior to 1973. Different portions of this standard, wood from the second half of the 19th century, may have varied in <sup>14</sup>C content by some 7 per mil (Stuiver and Quay, 1981). Such variability could possibly introduce calibration inaccuracies in the La Jolla data set of up to 58 radiocarbon years.

The Seattle Douglas Fir radiocarbon ages average 27 years older than the La Jolla German Oak dates. Similarly, Seattle Sequoia dates average 37 more radiocarbon years than the La Jolla Bristlecone Pine dates. These small differences likewise indicate the probability of laboratory bias.

# Table 2

 $\bar{x}$  (in years) is the weighted mean of the radiocarbon age differences of contemporaneous sample pairs from two data sets.  $\sigma_n$  is the standard deviation in this mean, calculated from the quoted errors in the radiocarbon ages. The second column gives the average error (in years) in the age differences of sample pairs, calculated from the quoted errors. The actual standard deviation  $\sigma_{12}$  in the age differences is given in years in the third column. The error multiplier,  $k_{12}$ , is  $\sigma_{12}$  divided by average,  $\sigma$ . The number of sample pairs used for the calculations is given in the last column.

Tree species	$ ilde{x} \pm \sigma_n$	Quoted average $\sigma$ in difference	Actual $\sigma_{\scriptscriptstyle 12}$	$K_{12}$	Ab interval	Number of comparisons
Seattle Douglas Fir Belfast Irish Oak	$4.4\pm3.9$	18.6	27.4	1.47	1410-1840	23
Seattle Douglas Fir La Jolla German Oak	$-27.2 \pm 8.7$	39.0	43.1	1.11	1120-1296	20
Seattle Douglas Fir La Jolla Bristlecone Pine	$-55.3 \pm 9.7$	38.8	43.3	1.12	1085-1285	16
Seattle Sequoia La Jolla Bristlecone Pine	$-37.4 \pm 7.6$	47.6	65.0	1.37	245-895	39
Seattle Sequoia Heidelberg German Oak	$-57.9 \pm 2.8$	23.0	44.4	1.93	240-760	66
Seattle Sequoia Seattle German Oak	$-22.9 \pm 5.6$	19.4	25.1	1.30	585-765	12
Seattle A Seattle B		18.1	27.7	1.53	-	30

A substantial systematic difference exists between the Seattle Sequoia and Heidelberg German Oak radiocarbon ages. In order to investigate the reality of a 58-year offset, we determined the radiocarbon ages of 12 oak samples provided by Bernd Becker of the University of Hohenheim. These samples were selected from parts of the radiocarbon record where the age discrepancies were largest. An age difference of nearly  $23 \pm 6$  radiocarbon years was found between Sequoia and German Oak, both measured in the Seattle laboratory. However, because we were biased in our sample selection towards the largest discrepancies, it is possible that the actual differences between complete series is less than 23 years.

Currently, the Heidelberg laboratory is remeasuring their secondary Heidelberg standard to investigate the possibility of a laboratory offset (Bruns, pers commun, 1981).

The excellent agreement between the Seattle and Belfast results, obtained on different tree species from different longitudes using different counting techniques, confirms the accuracy of the calibration curves. The offsets between the Seattle and La Jolla, and the Seattle and Heidelberg data sets, may be partly, and perhaps completely, due to laboratory bias. When measured in a single laboratory, the only systematic radiocarbon age difference is found between California Sequoia and German Oak, where the age difference amounts to 23 years maximally.

The laboratory error, quoted for a radiocarbon age, should ideally include the individual error resulting from any factor affecting the accuracy of the measurement. The error (one standard deviation) quoted for the Seattle radiocarbon determinations is based on the Poisson counting statistics of the sample and standard activities. The effect of additional errors that are unaccounted for in the quoted precision is to enlarge the overall laboratory error.

Systematic differences ("bias"), as well as increased variability, contribute to the larger error. The increased variability can be expressed relative to the quoted error  $\sigma$ . The actual laboratory error  $\sigma_L$  associated with the variability (effective laboratory variability) can be expressed as  $\sigma_L = k\sigma$ , where k, the "error multiplier", is a constant.

The Seattle Quaternary Isotope Laboratory recently participated in two calibration projects that provide information on the magnitude of the Seattle error multiplier  $k_s$  and bias, during the time these intercalibration samples were measured. Six samples were measured as part of the Glasgow intercalibration project of 20 radiocarbon laboratories (Scott, Baxter, and Aitchison, in press). Three different estimates of base lines were made for this study. For the Quaternary Isotope Laboratory, the average bias was 16.7 years (range of 7.4 to 25.1 years) towards older radiocarbon dates. As part of the calibration of new oxalic acid (Mann, pers commun, 1980) we determined the activity ratio of the new oxalic acid versus old oxalic acid. When expressing this ratio as an age, the bias of the Quaternary Isotope Laboratory, relative to the mean for nine laboratories, was less than two years.

For the Glasgow study, the Seattle error multiplier  $k_s$  was estimated at 1.7 in two different baseline interpretations (Scott, Baxter, and Aitchison, in press). When combining these results with the two Seattle ratio measurements of the oxalic acid project, we arrive for eight measurements at a  $k_s$  value of 1.6. These measurements would indicate an overall laboratory variability 1.6 times larger than the mean quoted error. However, these results were obtained for a small number of samples, and  $k_s=1$  cannot be excluded.

The above intercalibration projects provide valuable information on the order of magnitude of the effective laboratory error. The inherent weakness of these efforts is that they cover only the laboratory conditions encountered during a small fraction of its operating time. The following comparison of time-scale calibration data sets provides information on long-term variability.

When comparing two data sets, the standard deviation in the age difference of contemporaneous samples of identical <sup>14</sup>C content would be  $\sqrt{\sigma_1^2 + \sigma_2^2}$  when the mean quoted laboratory errors  $\sigma_i$  and  $\sigma_2$  are valid. This error can be compared to the standard deviation  $\sigma_{12}$  of the dif-

ferences of the two data sets [variance  $\sigma_{_{12}{}^2}=\frac{1}{N\text{-}1}\;\Sigma(x_i\text{-}\bar{x})^2]$  where N is

the number of comparisons,  $x_i$  the difference in radiocarbon ages of wood samples of the same dendro-age, and  $\bar{x}$  the mean age difference between the radiocarbon ages of contemporaneous samples. Ideally, the standard deviation around the mean age difference should be of the same order of magnitude as the mean quoted laboratory error in the difference. The distributions of age differences, around the mean radiocarbon age difference, are given in figure 10. A gaussian distribution (solid line) was fitted to the data using the calculated  $\sigma_{12}$ . The dashed line gives the calculated distribution based on the quoted laboratory errors. In all instances we find a broadening of the sample distribution. Table 2 gives the measure of broadening through the parameter,  $k_{12}$ , which is defined through  $\sigma_{12} = k_{12} \sqrt{\sigma_1^2 + \sigma_2^2}$ .

The larger-than- expected variances in the age differences may be due partly to actual differences in  $^{14}$ C content of the wood. In the following discussion, we first neglect the contribution to the variance caused by these differences, and ascribe the complete variability to an underestimation of the laboratory error. Through this approach, it is possible to obtain an upper limit for the effective laboratory variability,  $k\sigma$ .

If the entire increase in variance is ascribed to laboratory variability, the standard deviation of the age differences  $\sigma_{12}$  equals  $\sqrt{k_1^2\sigma_{12}^2 + k_2^2\sigma_{22}^2}$ . Substitution of the mean quoted errors,  $\sigma_1$  and  $\sigma_2$ , yields a relationship between the error multipliers,  $k_1$  and  $k_2$ . This relationship is plotted in figure 11 for the various data sets.

The horizontal  $k_1$  axis in figure 11 refers to the Seattle data whereas the vertical axis represents the possible k values of the comparison laboratories. For a set of 30 comparisons of pairs on contemporaneous samples of different trees, all measured in the Seattle laboratory, we

obtain a  $k_s$  value of 1.53 ( $\sigma_{11} = k_s \sigma_s \sqrt{2}$ , table 2, last row and figure 10). Acceptance of the 1.53 error multiplier for the Seattle laboratory (which is close to the 1.6 derived from the international calibration projects) leads to error multipliers of about 2.0 for Heidelberg, 1.5 for Belfast, and 1.1 to 1.4 for La Jolla. A slightly larger upper limit for the k values of these laboratories is given by the intercepts with the  $k_2$  axis (figure 11). Calculated in radiocarbon years, the mean quoted error associated with variability (not bias) should be increased from 12 to 18 years for Seattle, from 17 to 25 years for Belfast, from 21 to about 42 years for Heidelberg, and from 41 to 48 years for La Jolla. Expressed as additive sources of variances, these increases are, for the respective laboratories, equivalent with an additional variance of 13<sup>2</sup>, 18<sup>2</sup>, 36<sup>2</sup>, and 25<sup>2</sup> radiocarbon years<sup>2</sup>.

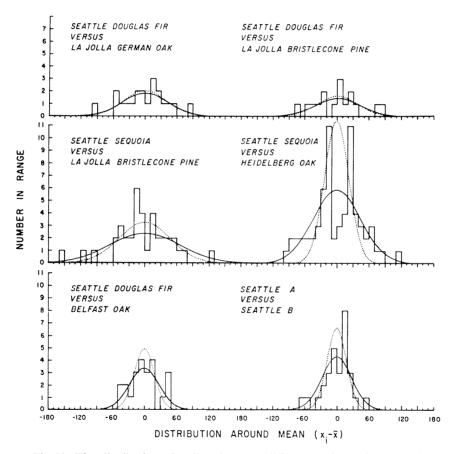


Fig 10. The distribution of radiocarbon age differences of sample pairs of the same dendro-age. The dashed curve gives the Gaussian distribution derived from the quoted standard deviations in the radiocarbon ages; the solid line gives the actual distribution. Narrowest distributions are derived for data sets with the highest precision. The frequency distributions were derived from data given in figures 8 and 9, except for Seattle A versus B which is an intra-lab comparison.

There are also factors outside the realm of the radiocarbon laboratory that may account for differences in reported radiocarbon ages of wood of the "same" dendro-age. Errors in the dendrochronologic evaluation of the tree rings, yielding an incorrect AD age, invalidate the premise of wood of equal age. Such errors probably contribute mostly to offsets between data sets. Once the correct AD age has been obtained, the wood cellulose <sup>14</sup>C content of different trees could possibly differ, and thus, provide different radiocarbon ages. Such differences could be due to variable regional differences in atmospheric <sup>14</sup>CO<sub>2</sub> levels, or perhaps to a very limited extent of variable root <sup>14</sup>CO<sub>2</sub> uptake in regions with a limestone soil CO<sub>2</sub> component. Variable levels of recycled CO<sub>2</sub> of biospheric origin, different in <sup>14</sup>C content from the atmospheric level, may also introduce anomalies.

In the previous discussion, we arrived at a maximum estimate of the error multiplier by neglecting the  $^{14}$ C content variability of wood series of the same dendro-age (but not necessarily the same AD age). By taking the  $^{14}$ C differences (standard deviation  $\sigma_{\rm tr}$ ) of the wood analyzed by different laboratories into account, as well as the effective laboratory errors, the following expression is obtained:

$$\sigma_{12}^2 = k_1^2 \sigma_1^2 + k_2^2 \sigma_2^2 + \sigma_{tr}^2$$

For the comparison of age series of samples of different trees of the same dendro-age within a single laboratory we obtain:

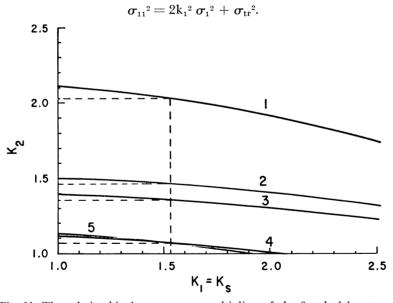


Fig. 11. The relationship between error multipliers of the Seattle laboratory ( $k_s$ ) and comparison laboratories ( $k_2$ ). Curves 1 and 2 relate to a Seattle-Heidelberg and Seattle-Belfast comparison; curves 3 and 4 were calculated from Seattle Sequoia-La Jolla Bristlecone Pine and Seattle Douglas Fir-La Jolla Bristlecone Pine comparisons. Curve 5 gives the error multiplier relationship derived from a Seattle Douglas Fir and La Jolla German Oak comparison.

As discussed,  $\sigma_{11} = 27.7$  years for a series of 30 Seattle samples (table 2). A lower limit for the Seattle error multiplier,  $k_s$ , would be 1.0. Actually, the choice of 1.0 appears a not-too-optimistic estimate when considering the single-year data sets discussed previously (figs 6 and 7). The variance of the single-year data around the decade averaged trend (fig 12) appears to be entirely compatible with the quoted laboratory error,  $\sigma_1$ . This conclusion is valid when considering the scatter of the single-year data around ten-year averages, thus creating a "stepped" curve, as well as when considering the scatter around a "continuous" curve through the decade averages. Because the variance of the single-year data around these trends is not any larger than the  $\sigma_1^2$  variance, the quoted precision,  $\sigma_1$ , has, at least for the single-year data, to be close to the effective laboratory error. Therefore, an error multiplier,  $k_s = 1.0$ , appears possible.

The use of  $\sigma_{11}=27.7$  years,  $k_s=1$ , and  $\sigma_1=12.9$  years yields a  $\sigma_{tr}$  of 20.8 years.

Perhaps a  $\sigma_{\rm tr}$  value of 20 years is valid for other tree age comparisons as well. In that instance, we arrive at error multipliers for Belfast, Heidelberg, and La Jolla of, respectively, 1.0, 1.7, and 1.0 to 1.3. These error multipliers are smaller than those derived earlier because we now have assigned a portion of the variance between data sets to the tree  $^{14}$ C levels, and not solely to the laboratory measurements.

Although a  $k_s=1.0$  value is likely for the Seattle single-year measurements, such a low error multiplier is not necessarily valid for all our decade samples. The 20-year value for  $\sigma_{tr}$  should, therefore, be considered an upper limit only.

### CONCLUSIONS

The calibration curves of figures 2 to 4 are an improvement upon existing calibration curves because high-precision radiocarbon ages are given for each decade. The conversion for the current millennium should be considered as the most reliable because this part of the curve has been confirmed by the data of the Belfast laboratory (Pearson, 1980; pers commun, 1981).

The interlaboratory comparison with La Jolla and Heidelberg yields offsets between data sets. These offsets (up to 58 radiocarbon years) are most likely due to laboratory bias. A "real" offset, of perhaps 23 years, appears possible for the California Sequoia and German Oak radiocarbon ages. This offset may be due to differences in wood <sup>14</sup>C content, but may also be due to errors in the dendro-age determinations.

The variability in the radiocarbon ages of different laboratories leads to certain estimates of the error multiplier, *ie*, the constant with which the quoted laboratory precision has to be multiplied to obtain the overall laboratory variability. For the Seattle, Belfast, La Jolla, and Heidelberg laboratories, the error multipliers are, respectively, in the range of 1.0 to 1.53, 1.0 to 1.5, 1.1 to 1.4, and 1.7 to 2.0.

A complication in comparing radiocarbon age errors is the possible difference in <sup>14</sup>C content of wood samples of the same dendro-age. The influence of these <sup>14</sup>C differences on calibration curves appears limited

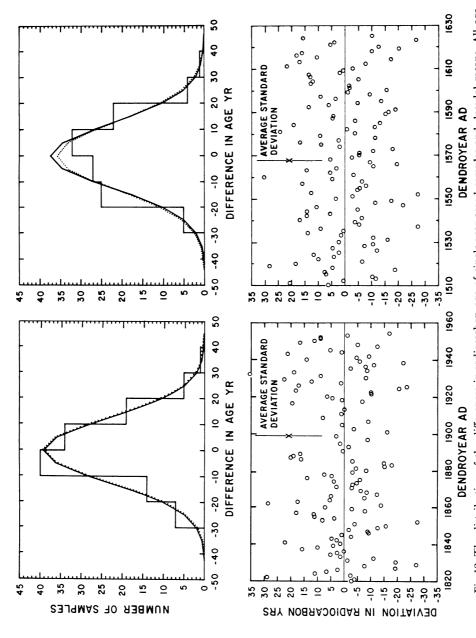


Fig 12. The distribution of the differences in radiocarbon age of single-year wood samples and decadal means. All age differences were obtained by comparing with the curve drawn through the decade averages. The dashed line is the expected Gaussian distribution calculated from the quoted radiocarbon age standard deviations (which are given in figures 6 and 7). The solid Gaussian curve is the actual distribution. The deviations from the decadal averages, as shown in the bottom figure, demonstrate the lack of a measurable 11-year cycle in the single-year radiocarbon ages.

because offsets are unlikely, and because the variability in  $^{14}$ C level of contemporaneous wood ( $\sigma_{tr}$ ) is, at the most, 20 years and probably less.

Data averaging would not improve the calibration curves of figures 2 to 4. For such averaging, the magnitude of the laboratory bias has to be established first through improved interlaboratory calibration. Assignment to each laboratory of a realistic error multiplier, and perhaps an estimate of  $\sigma_{\rm tr}$ , is also needed. A first attempt in averaging will be made by combining the Seattle data with those of the Belfast laboratory when the latter laboratory has finished the calibration of the last 2000 years of the Irish chronology.

Evidently, errors other than Poisson counting statistics increase the variability of the <sup>14</sup>C measurements. However, their influence is moderate. When the entire laboratory variability is taken into account, the uncertainty region of the calibration curves (one standard deviation) increases from about 12 years to a maximum of 18 years.

# ACKNOWLEDGMENTS

Many members of the Quaternary Isotope Laboratory contributed to this research. P M Grootes critically read the manuscript, R L Burk collected several of the tree sections, and P J Reimer handled most of the statistical calculations. For many years, P J Wilkinson dedicated much time to the high-precision measurements.

The Vancouver Island Douglas Fir was collected and cross-dated by M L Parker, L Joza, and P Bramhall of the Western Products Forestry Laboratory in Vancouver, British Columbia. The cross-check samples of German Oak were provided by Bernd Becker of the University of Hohenheim (Stuttgart), West Germany. The Bristlecone Pine samples were provided by C W Ferguson of the University of Arizona, Tucson.

The study of the relationship between atmospheric <sup>14</sup>C variations and climatic change, supported by National Science Foundation grant ATM-8022240 of the Climate Dynamics Program, resulted in many of the radiocarbon measurements reported in this paper. The interpretive portion and some of the earlier measurements were supported by the NSF geochemistry program, Grant EAR-8018141.

# REFERENCES

- Bruns, Michael, Münnich, K. O. and Becker, Bernd, 1980, Natural radiocarbon variations from AD 200 to 800, in Stuiver, Minze and Kra. Renee, eds, Internatl radiocarbon conf, 10th, Proc: Radiocarbon, v 22, no. 2, p 273-277.
- Burchuladze, A A, Pagava, S V, Povinec, P, Togonidze, G I, and Usačev, S, 1980, Radiocarbon variations with the 11 year solar cycle during the last century: Nature, v 287, p 320-322.
- v 287, p 320-322.

  Douglas, A E, 1919, Climatic cycles and tree growth, Vol 1: Carnegie Inst Washington Pub 289.
- Lerman, J C, Mook, W G, and Vogel, J C, 1970, C14 in tree rings from different localities, in Olsson, I U, ed, Radiocarbon variations and absolute chronology, Internatl radiocarbon conf, 7th, Proc: New York, John Wiley and Sons, p 275-299.
- Pearson, G W, 1980, High precision radiocarbon dating by liquid scintillation counting applied to radiocarbon timescale calibration, in Stuiver, Minze and Kra, Renee, eds, Internatl radiocarbon conf, 10th, Proc: Radiocarbon, v 22, no. 2, p 337-345.

  Scott, E M, Baxter, M S, and Aitchison, T C, in press, <sup>14</sup>C dating reproducibility:
- Scott, E M, Baxter, M S, and Aitchison, T C, in press, <sup>14</sup>C dating reproducibility: Evidence from a combined experimental and statistical programme, in Internatl symposium on C-14 and archaeology, 1st, Proc: Univ Groningen, in press.

- Stuiver, Minze, 1978, Radiocarbon timescale tested against magnetic and other dating methods: Nature, v 273, p 271-274.
- Stuiver, Minze and Polach, H A, 1977, Discussion reporting of <sup>14</sup>C data: Radiocarbon, v 19, p 355-363.
- Stuiver, Minze and Quay, P D, 1980, Changes in atmospheric carbon-14 attributed to
  - a variable sun: Science, v 207, p 11-19.

    1981, Atmospheric <sup>14</sup>C changes resulting from fossil fuel CO<sub>2</sub> release and cosmic ray variability: Earth and Planetary Sci Letters, v 53, p 349-362.
- Stuiver, Minze, Robinson, S W, and Yang, I C, 1979, <sup>14</sup>C dating to 60,000 years BP with proportional counters, in Berger, Rainer and Suess, H E, eds, Radiocarbon dating, Internal radiocarbon dating conf, 9th, Proc: Berkeley/La Jolla, Univ California Press, p 202-215.

  Suess, H E, 1978, La Jolla measurements of radiocarbon in tree-ring dated wood: Radiocarbon, v 20, p 1-18.