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LAMONT RADIOCARBON MEASUREMENTS VI*

WALLACE S. BROECKER and EDWIN A. OLSON

Lamont Geological Observatory (Columbia University), Palisades, New York

In contrast to previous radiocarbon measurement lists, this list contains only known-age samples, most of which formed during the past ten years. The measurements were made largely in order to gain an understanding of the distribution of radiocarbon within the dynamic carbon reservoir both today and at times in the past. Since all materials forming in this reservoir today do not have the same C^{14}/C^{12} ratio, such an understanding is necessary in order to arrive at the most accurate possible estimate of the age of samples submitted for dating. This is particularly important when high accuracy (i.e., <100 years error) is required on subaerially grown samples and also when attempting to extend the method to samples which formed in reservoirs other than the atmosphere (for example, the ocean and freshwater systems).

The data in this list are not reported with the idea of drawing new conclusions, for such conclusions as are possible have been reported elsewhere. However, republication in such a list as this has the following advantages: (1) if all laboratories summarize their measurements in this manner, the world data on C^{14}/C^{12} ratios in contemporary materials will be brought together in one place, in the same form, and in the same system of units; (2) by referencing the technical articles in which the data are discussed, the lists will act as a bibliography for such literature; (3) the summaries will be transferred directly to the punch cards published by Radiocarbon Dates Association, Inc., allowing a more complete and uniform coverage of the available data; and (4) such lists encourage the publication of isolated measurements which might otherwise remain in the files of individual radiocarbon laboratories.

EXPRESSING NATURAL RADIOCARBON VARIATIONS

In preparing this list the problem of the best way to express natural radiocarbon variations was carefully considered. A preliminary plan was submitted to a number of laboratories for criticism. Although a few investigators felt that the form chosen was too complex to be useful to the average geologist or archaeologist, the majority were in favor of the general plan. Numerous suggestions for changes in detail were made, some of these being adopted and some rejected. Even though the system adopted here may not be perfect in the eyes of some investigators, it is hoped that it will be accepted as the standard way of presenting C^{14} data on contemporary materials. The many advantages of a universal system should far outweigh the differences in opinion as to details of form.

The technical details of the system are given below. Following these is a section on the sources of natural radiocarbon variations, the purpose of which is to demonstrate to the nontechnical reader the advantages of the system and how it may be used.

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Ideally a tabulation of absolute radiocarbon measurements would be the best way to illustrate C^{14}/C^{12} ratio differences among a series of samples. As a consequence of numerous possible systematic errors, however, absolute radiocarbon measurements are exceedingly difficult to make, and so it is more practical to express results as a percentage difference from a universally accepted standard. In this paper, as in most isotopic work, this is done through the δ term,[1] defined for the isotope C^{14} as follows:

$$\delta C^{14} = \left[\frac{C_{sample} - C_{standard}}{C_{standard}}\right] \times 1000 \qquad (1)$$

where C denotes C^{14} concentration in any convenient units. Since a factor of 1000 is used instead of 100, δC^{14} is expressed in per mil, not percent.

In order to obtain radiocarbon concentrations, laboratories first measure activities—that is, C^{14} disintegration rates in a fixed quantity of carbon. Since the activity measured is directly proportional to the C^{14} concentration, it is convenient to use the directly measured activities in place of concentrations. Equation (1) then becomes:

$$\delta C^{14} = \left[\frac{A^* - A^0}{A^0}\right] \times 1000 \qquad (2)$$

where A stands for C^{14} activity, * denotes sample, and 0 denotes standard.

The purpose of a series of δC^{14} values calculated by equation (2) is to focus attention on the differences, in hope of understanding the factors which create inhomogeneities in the distribution of C^{14} throughout the dynamic reservoir.[2] Obviously, one possible explanation is simply that different samples have had different periods of C^{14} decay—that is, they were formed at different times in the past. By use of the standard radioactive decay equation, it is possible to convert measured sample activities to those that existed when the samples formed, thereby eliminating the age effect as a cause of δC^{14} differences. This will be referred to as age correction and is made by using the following equation:

$$A^* = A_M \cdot e^{\lambda t} \qquad (3)$$

where A^* is the age-corrected sample activity which one uses in equation (2), A_M is the measured sample activity, λ is the C^{14} decay constant, and t is the period between sample formation and activity measurement.

In equation (2) nothing specific was said concerning the reference standard. Although its selection is arbitrarily made, there are several requirements a good standard ought to meet:

[1] Use of this term should not obscure the simple concept it expresses. The statistician who says that John Doe makes 5% less money than the average American is expressing the same concept even though he does not call the figure 5% a δ term. If he did use the δ term, it would actually be –50, the minus sign denoting a value below standard and the 10-fold increase expressing per mil instead of percent.

[2] All the carbon in the Earth's crust can be considered as divided into two reservoirs. The dynamic reservoir consists of (1) the atmosphere, (2) the oceans and other water bodies, and (3) organic matter either living or undergoing decay. The dormant reservoir consists mainly of deposits of coal and oil, limestone and dolomite beds, and the organic fraction of shales. Within the dynamic reservoir, various mixing processes allow cosmic-ray produced radiocarbon to be distributed to all parts, replacing that lost by radioactive decay. On the other hand, the dormant reservoir, having had no access to fresh radiocarbon for millions of years, has lost by decay any carbon-14 once present.

1. It should be universally accepted.

2. It should be reliably uniform and readily available in sufficient quantities over a long period of time.

3. It should be invariant with time in the property for which it is to be a standard.

4. It should be a standard that one naturally and logically looks to for comparison.

At the present time the first two very practical requirements are adequately met by the oxalic acid C^{14} standard distributed by the National Bureau of Standards. The last two requirements can be met by modifying the measured oxalic acid activity in ways described in the following paragraphs.

Because the C^{14} activity of oxalic acid decreases with time, the third requirement is not met directly; but it can be met by converting the measured oxalic acid activity to what it was on January 1, 1958. This is done in the following equation identical in form to equation (3):

$$A'_{ox} = A_{ox} \cdot e^{\lambda t'} \tag{4}$$

where A'_{ox} is the age-corrected oxalic acid C^{14} activity, A_{ox} is the measured activity,[3] and t' is the time between January 1, 1958 and the date of measurement. Since the half-life of radiocarbon (equal to $.693/\lambda$) is 5570 years, the correction of measured activities to obtain A'_{ox} will be negligible for a number of years, the exact number depending on the sensitivity of an individual laboratory's measuring equipment.

The fourth requirement for a good standard undoubtedly involves the most subjectivity. Nevertheless, if practical aspects were not considered, most radiocarbon workers probably would suggest as a standard their own modern control samples, most of which are woods grown in the 19th century. After age correction the activities of these woods ought to be about the same (i.e., to within 10 per mil). When age-corrected, the Lamont modern control—1890 oak wood—has a C^{14} activity about 95 percent of the NBS oxalic acid standard (as measured in 1958). Consequently, it is suggested that A^0, the standard activity in equation (2), be obtained by multiplying the measured oxalic acid activity by .950, allowing, of course, for age correction back to January 1, 1958. In equation form, A^0 and the measured oxalic acid activity (A_{ox}) will be related as follows:

$$A^0 = .950A'_{ox} = .950A_{ox} \cdot e^{\lambda t'} \tag{5}$$

where t' is again the elapsed time between January 1, 1958 and the date of measurement. The use of the factor .950, based on Lamont 1890 oak wood, does not imply that the Lamont modern control is superior to control samples used by other laboratories. It merely represents an attempt to place near zero the δC^{14} values of modern control samples used by most radiocarbon laboratories. Even if future work were to disclose that .955 or .945 is a more representative factor, such a second-order correction would not merit changing the .950 factor adopted here.

[3] A_{ox} is the measured activity provided that no isotope fractionation has occurred during laboratory preparation of the counting gas. If fractionation does occur, A_{ox} is the activity after corection for isotope fractionation.

After standard selection, age correction of sample activities, and calculation of δC^{14} values, it remains to explain the differences among such values. One cause is isotopic fractionation during sample formation or during preparation for activity measurement. Fortunately it is possible to make a precise correction for this source of variation and thereby obtain a new term, called ΔC^{14}, which is independent of fractionation. This is done in an operation called "normalization to the same C^{13}/C^{12} ratio"; it leads to the following equation:

$$\Delta C^{14} = \delta C^{14} - 2\delta C^{13}\left(1 + \frac{\delta C^{14}}{1000}\right) - 50.0 \qquad (6)$$

Details of the normalization procedure and of the derivation of equation (6) are given in the Appendix, while δC^{13} is discussed below. At this point it is sufficient to say only that ΔC^{14} values, defined in terms of the above equation, are equivalent to δC^{14} values except that fractionation has been removed as a source of difference among samples.

If in the transfer of a chemical element from one phase to another the istotopic composition of the portion transferred differs from that of its source, isotopic fractionation has occurred during the process. In the case of carbon (which is 98.9 percent C^{12}, 1.1 percent C^{13}, and about 10^{-10} percent C^{14}) there is a difference in the extent of fractionation among the isotopes, as first pointed out by Craig (1953). For example, if a wood sample grows with a 20 per mil lower C^{13}/C^{12} ratio[4] than its source, the atmosphere, it will have a 40 per mil lower C^{14}/C^{12} ratio than the atmosphere. Rafter (1955) obtained the first experimental verification of this difference in natural materials. Normalization, then, is simply an adjustment of a sample's living C^{14} activity to a level equivalent to the C^{13}/C^{12} ratio of a hypothetical dynamic reservoir having a uniform composition of the two stable carbon isotopes. Whether the adjustment is up or down depends on whether the sample is poorer or richer in C^{13} than the hypothetical reservoir.

In this paper all δC^{13} values are based on the *belemnite* standard of Craig (1957b). Although this standard is not universally available, Craig has carefully intercalibrated it with C^{13} standards used by other investigators. As yet, no direct intercalibration has been made between the belemnite standard and the NBS oxalic acid, but individual radiocarbon laboratories having mass spectrometers may do so indirectly using one of the other C^{13} standards.

For laboratories without access to a mass spectrometer, it is suggested that δC^{13} values for samples be estimated from literature data on samples of similar type (e.g. Craig, 1953). Such δC^{13} estimates, together with the ΔC^{14} values calculated from them, should be denoted by some such scheme as enclosure in parentheses. Errors affixed to such δC^{13} and ΔC^{14} values should reflect the δC^{13} spread indicated in the literature for a given sample type. Although this

[4] In the same manner that δC^{14}, defined by equation (2), indicates relative radiocarbon concentrations, so δC^{13} expresses C^{13}/C^{12} variations through the following similar equation (see Craig, 1953):

$$\delta C^{13} = \left[\frac{R_{sample} - R_{standard}}{R_{standard}}\right] \times 1000 \qquad (7)$$

where R stands for the ratio C^{13}/C^{12} as measured on the mass spectrometer. It is δC^{13} which is actually used in normalization; see equation (6) and the Appendix.

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scheme is certainly not satisfactory when high precision is necessary (approximately 3 to 8 per mil), it will be useful in dealing with problems where ΔC^{14} differences are much larger.

In equation (6) defining ΔC^{14}, the constant term 50.0 may cause some question. Just as age-corrected 19th century woods were made to have a δC^{14} value approximating zero, so introduction of the constant 50.0 into equation (6) puts the ΔC^{14} values of these woods near zero. Although the added constant is not strictly necessary, its inclusion creates a more convenient ΔC^{14} scale, one that generally should give immediate significance to individual ΔC^{14} values.

SOURCES OF VARIATION IN ΔC^{14}

What can be learned from radiocarbon data expressed in terms of ΔC^{14} values? The following section is designed to answer that question for those not familiar with the technical aspects of carbon-14 dating. Although the discussion must not be considered as "do-it-yourself instructions" for patching up apparently anomalous radiocarbon dates, it should serve a twofold purpose: (1) to point up certain factors that control the accuracy of radiocarbon dates and (2) to guide archaeologists and geologists in the selection of valuable control samples.

In order to compute a radiocarbon age, the ΔC^{14}—and thus the initial C^{14}/C^{12} ratio—of the material to be dated must be estimated. Normally this is done by measuring ΔC^{14} in a similar material formed at a known time and in a similar environment. The accuracy of the age obviously depends on the accuracy of the ΔC^{14} estimate. An error of 1 percent in the estimate (10 on the per mil scale) will introduce an error of 80 years in the age of the sample; one of 20 percent (200 per mil) will introduce an error of 1800 years. Obviously, then, it is important to consider what factors are theoretically capable of producing ΔC^{14} variations in sample materials and to measure the ΔC^{14} values of samples representing different environments and different times in the past.

From the standpoint of precise radiocarbon dating, it would be ideal to find the same ΔC^{14} value for all samples regardless of age and growth environment. However, small ΔC^{14} differences do exist, demonstrating (1) that the Earth's radiocarbon is inhomogeneously distributed at the present time, (2) that the pattern of inhomogeneity may have been different in the part, and (3) that the total amount of terrestrial radiocarbon has perhaps varied significantly in the past. Regardless of which of these three situations is directly involved, there are two ultimate causes of ΔC^{14} differences:

1. C^{14}-free carbon transferred from the dormant carbon reservoir and cosmic-ray-produced radiocarbon are not mixed instantaneously throughout the dynamic reservoir.

2. the rates at which these two types of carbon are added have varied with time.

In the following paragraphs several aspects of these two causes are discussed and illustrated with examples from the literature.

The most serious problems in the choice of accurate control values for dating are due to the first cause—finite rates of mixing within the dynamic carbon reservoir. The first problem to be considered involves newly produced

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cosmic ray C^{14}. Since this is added directly to the atmosphere, it ought to give the atmosphere a slightly higher C^{14}/C^{12} ratio than the average for the entire dynamic reservoir. A similar situation exists when someone continuously drops red ink into a sinkful of water; the water is redder at the point of addition. Thus, it is not surprising to find that wood grown in the atmosphere averages 40 per mil higher on the ΔC^{14} scale than shells grown in surface ocean water (see Sample Descriptions, II). On the assumption that such a difference results from a finite mixing rate between atmospheric carbon dioxide and oceanic bicarbonate, Arnold and Anderson (1957), Revelle and Suess (1957), and Craig (1957a) have calculated that the average cosmic-ray-produced radiocarbon atom resides in the atmosphere from 5 to 20 years before entering the ocean. If this time were only a few days, the mixing between atmosphere and ocean would be a nearly instantaneous process, and there would be no ΔC^{14} difference between wood and shells.

It is clear, then, that carbon-14 age determinations on materials formed in the ocean must be based on control samples formed in the ocean. The actual situation, however, is not quite so simple; for mixing within the ocean itself is complex, and it is probable that the ΔC^{14} values for materials formed in surface ocean water vary considerably, perhaps over a 50 per mil range. Consequently, before age determinations on oceanic materials can be made to a precision of better than 200 years, a detailed study of oceanic ΔC^{14} variations must be made.

A more subtle example of ΔC^{14} variations produced by finite mixing rates involves movements of ocean water masses, important considerations in view of the fact that over 90 percent of the world's radiocarbon is oceanic. Oceanographers have long recognized both the existence of definite water masses and a circulation pattern which involves sinking in polar regions and upwelling at other points. The period of circulation is probably of the order of centuries, long enough for the radiocarbon concentration in deep masses to drop measurably as a result of radioactive decay. If for some reason the circulation rate were to rise suddenly, there would be a gradual drop in the atmospheric C^{14}/C^{12} ratio caused by the return of this C^{14}-deficient water to the surface and its equilibration with the air. If the circulation maintained this higher rate, the atmospheric radiocarbon concentration would ultimately level out at a lower average C^{14}/C^{12} ratio. On the other hand, if the circulation rate were to vary up and down, so would the atmosphere concentration—but with a lag of several years. The possibility of such variations was first suggested by Broecker and others (1958), following Worthington's (1954) suggestion of catastrophic overturn in the oceans during particularly cold years.

Recently de Vries (1958) has made precise measurements of the C^{14}/C^{12} ratio in tree rings, the growth dates of which cover the past several hundred years. He found irregular oscillations of about 1 percent on either side of the mean. Furthermore, he showed that these oscillations match climate variations in the manner expected if the oscillations were related to variations in oceanic mixing rates. Again it is clear that if radiocarbon dates are ever to be accurate within less than 100 years, such oscillations must be carefully defined.

Materials formed in freshwater systems also illustrate ΔC^{14} variations produced by finite mixing rates. Here, however, the carbon being added to the

dynamic reservoir is free of radiocarbon, being derived from ancient carbonate deposits. If mixing between atmospheric carbon dioxide and the dissolved carbonate were infinitely rapid, the ΔC^{14} values of materials forming within freshwater systems would be identical to those of atmospherically derived materials. The data in Sample Descriptions, III show that whereas this ideal situation is approached in some cases (for example, Walker Lake) the ΔC^{14} results for freshwater materials are often very low, a fact first demonstrated by Deevey and others (1954). Therefore, unless proper control samples are measured along with a given freshwater material to be dated, age errors up to at least 2000 years may result. For a detailed discussion of this problem, the reader is referred to a recent paper by Broecker and Walton (1959).

The other potential cause of ΔC^{14} variations, namely, variable addition rates of both radiocarbon and C^{14}-free carbon, can be demonstrated best by changes produced by man. This is so because presently there is no positive evidence for significant long-term natural variations in addition rates of either variety of carbon (Arnold and Libby, 1949; Münnich and others, 1958; Barker, 1958; Broecker, Olson, and Bird, in press). True, the rate of radiocarbon addition undoubtedly varies with observed short-term cosmic ray variations, but as yet nothing is known of any long-term cosmic ray fluctuations. Similarly, the natural transfer of ancient C^{14}-free carbon from the dormant to the dynamic carbon reservoirs is taking place continuously (and probably erratically on a day-to-day basis), but there is no evidence so far to show that the rate is great enough to significantly affect the radiocarbon dating method.

Recently man has begun to add significant amounts of both C^{14}-free carbon and radiocarbon. The first fact was demonstrated originally by Suess (1955) who showed that woods grown between 1930 and 1950 have ΔC^{14} values 20 to 40 per mil below pre-1890 woods. This "Suess effect" is simply the result of diluting atmospheric radiocarbon with tremendous quantities of C^{14}-free carbon dioxide, a combustion product of the coal and oil that have powered the modern industrial revolution.

With the advent of large-scale nuclear bomb testing, the natural method of radiocarbon production has been simulated on a scale large enough to have world-wide effects. In fact, the Suess effect has been more than compensated for, and the ΔC^{14} value for newly formed plant material is currently rising at the rate of 20 to 50 per mil each year (see Rafter and Fergusson, 1957; de Vries, 1958; Münnich and Vogel, 1958; and Broecker and Walton, in press).

Perhaps a natural first reaction to the Suess and H-bomb effects is to conclude that the radiocarbon dating method will soon be valueless. This certainly is not true, for the addition of C^{14}-free industrial carbon dioxide and H-bomb radiocarbon are recent. After 19th century woods of known age are age-corrected, they provide the required C^{14} index of what present-day wood samples would be if man had not tampered with the natural regime. Actually, such tampering is equivalent to isotopic tracer experimentation on a grand scale and has provided valuable quantitative information on rates of mixing within the dynamic-carbon reservoir. This information is obviously of importance to a detailed understanding of the natural variations in ΔC^{14} values.

DATA PRESENTED IN THIS LIST

The data reported below were obtained at the Lamont Radiocarbon Laboratory during the past three years using techniques described by Broecker, Tucek, and Olson (in press); these involve basically the CO_2 method of de Vries and Barendsen (1953) and Fergusson (1955). The errors attached to δC^{14} values, though based on the reproducibility of the counting measurements, are approximately equal to the statistical errors. The δC^{13} values were calculated from C^{13}/C^{12} measurements on the CO_2 gas used for radiocarbon analysis. Dr. Wayne Ault and Mr. Glen Erickson supervised the measurements. Since no analyses of the oxygen isotope concentration in the CO_2 gas were made, the δC^{13} results have an uncertainty of about 1 per mil (see Craig, 1957b). In the case of samples for which no C^{13}/C^{12} ratios were measured, δC^{13} values are estimated from data on similar materials; these estimated δC^{13} values (see Craig, 1953) and the ΔC^{14} values based on them are enclosed in parentheses in order to distinguish them from experimentally determined values.

In addition to the samples reported here about 100 oceanic bicarbonate samples have been measured. These results will be published as soon as the present International Geophysical Year Program is completed.

For convenience, the outline of the tabulated data is presented below.

Section I. Atmospheric CO_2 together with plant or animal material deriving their carbon from atmospheric CO_2
- A. Pre-1900 samples
- B. Samples reflecting the "Suess effect" (1900-1952)
- C. Samples reflecting atomic bomb testing (1952-)

Section II. Oceanic dissolved bicarbonate together with samples deriving their carbon from oceanic dissolved bicarbonates

Section III. Dissolved bicarbonate from freshwater systems together with samples deriving their carbon from freshwater bicarbonate
- A. Samples from lakes
- B. Samples from rivers
- C. Samples from groundwater
- D. Samples from hot springs

If the reader keeps the following facts in mind as he scans the tables, he will find the data more meaningful.

1. ΔC^{14} values vary because the distribution of radiocarbon varies from place to place and/or from time to time.

2. δC^{14} values vary for the same reasons and also show the effects of isotopic fractionation.

3. The values of neither ΔC^{14} nor δC^{14} vary because samples are of different age, for such an effect has been corrected for.

4. δC^{13} values vary only because of isotopic fractionation.

5. All values tabulated are in per mil, which is one-tenth of a percent.

6. Negative values mean lower C^{14} or C^{13} concentrations than in the standards; the opposite is true for positive values.

7. For ΔC^{14} and δC^{14}, the standard approximates 19th century wood. For δC^{13}, the standard approximates marine shell.

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APPENDIX

Derivation of Equation Defining ΔC^{14} (equation 6 in text)

1. *Symbols and Abbreviations*

A^0 = .950 times the activity of the N.B.S. C^{14} standard corrected for decay (as defined by equation 5 in the text).

A^* = C^{14} activity of a sample after age correction.

A^*_N = A^* after normalization (that is, the C^{14} activity which an age-corrected sample would have if it had the same C^{13} concentration as the belemnite C^{13} standard).

$R_{sam.}$ = the C^{13}/C^{12} ratio of a sample.

$R_{stan.}$ = the $C^{13}/^{12}$ ratio of the belemnite standard.

k = a constant introduced in order that ΔC^{14} for 19th century woods will lie close to zero on the scale.

2. *Defined δ Terms* (equations 2 and 7 in the text)

$$\delta C^{14} = \left[\frac{A^* - A^0}{A^0}\right] \times 1000 \qquad \text{Equation (A)}$$

$$\delta C^{13} = \left[\frac{R_{sam.} - R_{stan.}}{R_{stan.}}\right] \times 1000 \qquad (B)$$

3. *Defined ΔC^{14} Term* (see section 6 for an evaluation of k)

$$\Delta C^{14} = \left[\frac{A^*_N - A^0}{A^0}\right] \times 1000 + k \qquad (C)$$

4. *Normalization Relation*

This asserts (see Craig, 1953, and Rafter, 1955) that the factor by which

the isotope C^{14} is either favored or repressed during fractionation is double the similar factor for isotope C^{13}. The negative sign on the δC^{13} term below results from the fact that samples having *less affinity* for C^{13} than the belemnite (hence, negative δC^{13}) should have their C^{14} activities *increased* in order to compensate for the C^{14} lost because of fractionation.

$$\left[\frac{A^*_N - A^*}{A^*}\right] \times 1000 = -2\,\delta C^{13} \qquad (D)$$

5. *Combination of Equations*

(a) Solve equation (D) for A^*_N and substitute in equation (C)

$$\Delta C^{14} = \left[\frac{A^*}{A^0}\left(1 - \frac{2\,\delta C^{13}}{1000}\right) - 1\right] \times 1000 + k \qquad (E)$$

(b) Solve equation (A) for $\frac{A^*}{A^0}$ and substitute in equation (E)

$$\Delta C^{14} = \delta C^{14} - 2\,\delta C^{13}\left(1 + \frac{\delta C^{14}}{1000}\right) + k \qquad (F)$$

6. *Adjusting Equation (F) so that ΔC^{14} for 19th Century Woods is about Zero*

On pages 112-113 of the text was a discussion of the C^{14} standard, the one adopted having made δC^{14} approximately zero for 19th century woods. Thus, for such woods, equation (F) becomes: $\Delta C^{14} = (-2\,\delta C^{13} + k)$. Craig (1953) has shown that woods have δC^{13} values clustering around –25 per mil, ranging from –22.5 to –27.9. Thus the ΔC^{14} value for pre-1900 woods will fall close to zero on the scale if k is chosen as –50.0 per mil. In this case:

$$\Delta C^{14} = \delta C^{14} - 2\delta C^{13}\left(1 + \frac{\delta C^{14}}{1000}\right) - 50.0 \qquad (G)$$

SAMPLE DESCRIPTIONS

Lamont No.	Locality	δC^{14}	δC^{13}	ΔC^{14}

I. SAMPLES UTILIZING ATMOSPHERIC CO_2

A. Samples Defining the Atmospheric C^{14}/C^{12} Ratio Before 1900

L-371E. Pompeii, Italy **8±5** **–21.1** **1±5**

Carbonized bread that was charred during the volcanic ash fall that buried the city (40° 45′ N Lat, 14° 29′ E Long) in A.D. 79. Coll. by A. Maiuri; subm. by Junius Bird, American Museum of Natural History (see Broecker, Olson, and Bird, in press).

L-108A. King's Canyon National Park **12±7** **–20.5** **3±7**

Sequoia wood from the A.D. 1057 to 1087 growth rings in a display section of the American Museum of Natural History forestry collection. Tree grew near the entrance to King's Canyon

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Lamont No.	Locality	δC^{14}	δC^{13}	ΔC^{14}

National Park, California (36° 48′ N Lat, 118° 40′ W Long), and was cut in 1891. Coll. and subm. by Junius Bird (see Broecker, Olson, and Bird, in press).

L-108B. King's Canyon National Park **−13±7** **−18.7** **−26±7**

Sequoia wood from the A.D. 570 to 578 growth rings of the same tree section described in L-108A.

L-113. Tikal, Guatemala **9±7** **−21.8** **3±7**

Zapote wood from a carved lintel marked 9.15.10.0.0 in a Mayan temple (17° 12′ N Lat, 89° 36′ W Long). According to the Spinden correlation the date is equivalent to August 30, 481 A.D. The wood represents between 20 and 40 growth years. Based on the Goodman-Thompson correlation, which equates the Mayan date to June 30, 741 A.D., a value of -30 ± 7 is obtained for ΔC^{14}. Subm. by Junius Bird (see Broecker, Olson, and Bird, in press).

L-353A. U. S. Pacific Northwest **19±6** **−24.9** **19±6**

Wood from the A.D. 1632 to 1636 growth rings of a Sitka spruce section which is part of the American Museum of Natural History forestry collection (Cat. No. 456). The tree was cut in 1881. Exact growth location unknown. Coll. July 1956 by Junius Bird, F. Scherer and W. S. Broecker (see Broecker, Olson, and Bird, in press).

L-353C. U. S. Pacific Northwest **28±6** **−18.0** **15±6**

Wood from the A.D. 1675 to 1678 growth rings of the same Sitka spruce described in L-353A.

L-353E. U. S. Pacific Northwest **12±5** **−20.9** **4±5**

Wood from the A.D. 1734 to 1742 growth rings of the same Sitka spruce described in L-353A.

L-353B. U. S. Pacific Northwest **16±5** **−24.1** **15±5**

Wood from the A.D. 1807 to 1812 growth rings of the same Sitka spruce described in L-353A.

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Lamont No. Locality	δC^{14}	δC^{13}	ΔC^{14}
L-353F. U .S. Pacific Northwest Wood from the A.D. 1821 to 1826 growth rings of the same Sitka spruce described in L-353A.	**13±5**	**–22.0**	**8±5**
L-353D. U. S. Pacific Northwest Wood from the A.D. 1870 to 1881 growth rings of the same Sitka spruce described in L-353A.	**0±6**	**–21.4**	**–7±6**
L-314. Palisades, New York Wood from the A.D. 1888 to 1892 growth rings of an oak tree grown on the grounds of the Lamont Geological Observatory (41° 00′ N Lat, 73° 55′ W Long). Coll. February 1955 by C. S. Tucek, presently at Bethel College, St. Paul, Minnesota, and W. S. Broecker (see Broecker, Olson, and Bird, in press).	**1±5**	**–24.0**	**0±5**
L-315. Palisades, New York Wood from the A.D. 1888 to 1892 growth rings of a pine tree grown on the grounds of the Lamont Geological Observatory (41° 00′ N Lat. 73° 55′ W Long). Coll. February 1955 by C. S. Tucek and W. S. Broecker (see Broecker, Olson, and Bird, in press).	**–2±6**	**–23.0**	**–6±6**

B. Samples Defining the Atmospheric C^{14}/C^{12} Ratio During the Time of the Suess Effect (1900-1952)

L-313. Palisades, New York Wood from the 1936 to 1940 growth rings of an oak tree grown on the grounds of the Lamont Geological Observatory (41° 00′ N Lat, 73° 55′ W Long). Coll. by W. S. Broecker and C. S. Tucek (see Broecker, Olson, and Bird, in press).	**–29±5**	**–24.1**	**–32±5**
L-288M. Lake Winnemucca, Nevada Woody portion of sage brush growing on the east shore of the dry lake in the Fishbone Cave area (40° 15′ N Lat, 119° 18′ W Long). Although this sample was collected in September 1955, it was a 1-in.-diam stalk which grew mostly during the time of the Suess effect and is so categorized. Coll. by P. C. Orr, Western Speleological Institute, and W. S. Broecker (see Broecker and Orr, 1958).	**4±7**	**–11.2**	**–23±7**

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Lamont No.	Locality	δC^{14}	δC^{13}	ΔC^{14}
L-184E.	**Palisades, New York**	**−37±15**	**−23.8**	**−41±15**

Twigs from an oak tree grown on the grounds of the Lamont Geological Observatory (41° 00′ N Lat, 73° 55′ W Long). Coll. during the summer of 1952 by B. Eckelmann and J. L. Kulp, Lamont Geological Observatory (see Carr and Kulp, 1954).

C. Samples Defining the Atmospheric C^{14}/C^{12} Ratio Since Large-scale Bomb-testing Began (1952-)

L-316A.	**Palisades, New York**	**−24±8**	**−22.2**	**−30±8**

Pine needles coll. during late fall of 1954 on the grounds of the Lamont Geological Observatory (41° 00′ N Lat, 73° 55′ W Long) by C. S. Tucek and W. S. Broecker.

L-316B.	**Palisades, New York**	**−17±6**	**−23.2**	**−21±6**

Oak leaves coll. during the fall of 1954 on the grounds of the Lamont Geological Observatory (41° 00′ N Lat, 73° 55′ W Long) by C. S. Tucek and W. S. Broecker.

L-367A.	**North Atlantic Ocean**	**62±8**	**−9.0**	**31±8**

Atmospheric CO_2 coll. on the Columbia University research vessel *Vema* by pulling air through CO_2-free KOH. Continuous absorption from 36° 15′ N Lat, 64° 40′ W Long to 32° 27′ N Lat. 63° 47′ W Long. Coll. June 1956 by R. Gerard, Lamont Geological Observatory (see Broecker and Walton, in press).

L-367B.	**North Atlantic Ocean**	**54±6**	**(−8±4)**	**(21±7)**

Atmospheric CO_2 coll. on the Columbia University research vessel *Vema* by pulling air through CO_2-free KOH. Continuous absorption from 32° 27′ N Lat, 63° 47′ W Long to 32° 48′ N Lat, 54° 23′ W Long. Coll. June 1956 by R. Gerard (see Broecker and Walton, in press).

L-367C.	**Mediterranean Sea**	**88±8**	**−7.4**	**54±8**

Atmospheric CO_2 coll. on the Columbia University research vessel *Vema* by pulling air through CO_2-free KOH. Continuous absorption from 41° 02′ N Lat. 12° 45′ E Long to 40° 30′ N Lat. 14° 01′ E Long. Coll. June 1956 by R. Gerard (see Broecker and Walton, in press).

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Lamont No. Locality	δC^{14}	δC^{13}	ΔC^{14}
L-367D. Mediterranean Sea Atmospheric CO_2 coll. on the Columbia University research vessel *Vema* by pulling air through CO_2-free KOH. Continuous absorption from 36° 34′ N Lat, 23° 14′ E Long to 35° 10′ N Lat, 23° 35′ E Long. Coll. June 1956 by R. Gerard (see Broecker and Walton, in press).	**64±6**	**–5.3**	**25±6**
L-371A. Rome, Italy Fresh-sprouted wheat coll. November 7, 1956 near Rome (41° 45′ N Lat, 12° 15′ E Long) by L. Lodi. Subm. by Junius Bird (see Broecker and Walton, in press).	**23±6**	**–25.1**	**24±6**
L-371B. Rome, Italy Poplar twigs from the same area as L-371A above. Coll. November 7,1956 by L. Lodi; subm. by Junius Bird (see Broecker and Walton, in press).	**17±5**	**–25.0**	**18±5**
L-415B. Kearney, Nebraska Tree leaves grown near the point where Nebraska Route 44 crosses the Platte River (40° 40′ N Lat, 99° 02′ W Long). Coll. August 1957 by A. Walton, National Physical Laboratory, Teddington, England, and W. S. Broecker (see Broecker and Walton, 1959).	**48±7**	**–31.8**	**65±7**
L-415G. Evanston, Wyoming Tree leaves grown near the point where U. S. Route 30 crosses the Bear River (41° 16′ N Lat, 110° 58′ W Long). Coll. August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).	**83±7**	**–28.2**	**94±7**
L-415HH. Wadsworth, Nevada Tree leaves grown near the point where U. S. Route 40 crosses the Truckee River (39° 38′ N Lat, 119° 17′ W Long). Coll. August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).	**85±7**	**–27.8**	**95±7**
L-415QQ. Truckee, California Tree leaves from 3 mi E of Truckee, California on U. S. Route 40 (39° 20′ N Lat, 120° 14′ W Long). Coll. August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).	**91±7**	**–27.2**	**100±7**

Lamont No. Locality	δC^{14}	δC^{13}	ΔC^{14}
L-415EEE. Great Salt Lake, Utah	**89±7**	**–26.0**	**96±7**
Tree leaves from a swamp maple growing on the Provo terrace at the north end of the Oquirrh range (40° 42′ N Lat, 112° 16′ W Long). Coll. August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).			
L-445. Palisades, New York	**53±7**	**–25.6**	**57±7**
Oak and maple leaves from the grounds of the Lamont Geological Observatory (41° 00′ N Lat, 73° 55′ W Long). Coll. October 1957 by W. S. Broecker (see Broecker and Walton, 1959).			
L-452. Palisades, New York	**79±9**	**–21.0**	**75±9**
Forsythia blossoms from the grounds of the Lamont Geological Observatory (41° 00′ N Lat, 73° 55′ W Long). Coll. during May 1958 by W. S. Broecker.			
L-456. Oxalic acid	**53**	**–16.9**	**39±2**
National Bureau of Standards C^{14} standard. (Note that the δC^{14} for this sample is a defined rather than a measured value. The error in the ΔC^{14} value results from the uncertainty in the δC^{13} measurement).			
L-464C. North Atlantic Ocean	**131±7**	**–10.8**	**105±7**
Atmospheric CO_2 coll. on the Columbia University research vessel *Vema* by pulling air through CO_2-free KOH. Continuous absorption from 29° 57′ N Lat, 61° 41′ W Long to 21° 43′ N Lat, 51° 07′ W Long. Coll. November 1957 by T. Takahashi, Lamont Geological Observatory (see Broecker and Walton, 1959).			
L-466B. North Atlantic Ocean	**109±6**	**–7.2**	**75±6**
Atmospheric CO_2 coll. on the Columbia University research vessel *Vema* by pulling air through CO_2-free KOH. Continuous absorption from 15° 04′ N Lat, 39° 49′ W Long to 7° 22′ N Lat, 29° 59′ W Long. Coll. December 1957 by T. Takahashi (see Broecker and Walton, in press).			
L-466C. South Atlantic Ocean	**94±9**	**–8.8**	**63±9**
Atmospheric CO_2 coll. on the Columbia University research vessel *Vema* by pulling air through CO_2-free KOH. Continuous absorption from 30° S Lat, 44° W Long to 35° S Lat, 55° W Long. Coll. January 1958 by T. Takahashi (see Broecker and Walton, in press).			

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Lamont No.	Locality	δC^{14}	δC^{13}	ΔC^{14}
L-466D.	**Straits of Magellan** Atmospheric CO_2 coll. on the Columbia University research vessel *Vema* by pulling air through CO_2-free KOH. Continuous absorption from 53° 40′ S Lat, 60° 40′ W Long to 54° 50′ S Lat, 68° 30′ W Long. Coll. February 1958 by T. Takahashi (see Broecker and Walton, in press).	**103±6**	**–13.1**	**82±6**
L-487B.	**Evanston, Wyoming** Tree leaves from the point where U. S. Route 30 crosses the Pear River (41° 16′ N Lat, 110° 58′ W Long). Coll. September 1958 by W. S. Broecker (see Broecker and Walton, in press).	**142±7**	**–23.7**	**146±7**
L-487C.	**Great Salt Lake, Utah** Swamp maple leaves from the Provo terrace at the north end of the Oquirrh range (40° 42′ N Lat, 112° 16′ W Long). Coll. September 1958 by W. S. Broecker (see Broecker and Walton, in press).	**111±7**	**–22.6**	**111±7**
L-487D.	**Wadsworth, Nevada** Tree leaves grown near the point where U. S. Route 40 crosses the Truckee River (39° 38′ N Lat, 119° 17′ W Long). Coll. August 1958 by W. S. Broecker (see Broecker and Walton, in press).	**131±9**	**–26.1**	**140±9**
L-487E.	**Truckee, California** Tree leaves coll. 3 mi E of Truckee (39° 20′ N Lat, 120° 14′ W Long) on U. S. Route 40 August 1958 by W. S. Broecker (see Broecker and Walton, in press).	**126±7**	**–26.1**	**135±7**
L-487S.	**Palisades, New York** Tree leaves from the grounds of the Lamont Geological Observatory (41° 00′ N Lat, 73° 55′ W Long). Coll. October 1958 by W. S. Broecker (see Broecker and Walton, in press).	**77±5**	**–27.0**	**85±5**
	II. SAMPLES UTILIZING OCEANIC HCO^-_3			
L-317A.	**Long Island Sound** Clam shells coll. during the fall of 1954 in the Port Jefferson area (40° 57′ N Lat, 73° 05′ W Long) by W. S. Broecker.	**–2±9**	**0.0**	**–52±9**

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Lamont No.	Locality	δC^{14}	δC^{13}	ΔC^{14}
L-317B.	**Long Island Sound**	**−44±10**	**−14.3**	**−67±10**

Clam meat from the same clams described above in L-317A.

L-317C.	**Long Island Sound**	**−35±8**	**1.4**	**−88±8**

Dissolved bicarbonate from water where clams described above in L-317A,B were collected. Coll. during the fall of 1954 by W. S. Broecker.

L-241A.	**Tipasa, Algeria**	**4±10**	**(0±3)**	**(−46±10)**

Living shells coll. 1954 on Kouali Point (36° 40′ N Lat, 2° 30′ E Long) by L. C. Briggs; subm. by C. E. Stearns, Tufts University.

III. SAMPLES UTILIZING CO_2 FROM TERRESTRIAL WATERS

A. Lake Samples

L-288C.	**Pyramid Lake, Nevada**	**−11±7**	**6.3**	**−74±7**

Currently forming unlithified $CaCO_3$ which coats portions of the base of the Pyramid at the present water level (39° 59′ N Lat, 119° 30′ W Long). Coll. September 1955 by P. C. Orr and W. S. Broecker (see Broecker and Orr, 1958).

L-288I.	**Pyramid Lake, Nevada**	**−75±10**	**−22.7**	**−83±10**

Algae floating on the surface of the lake on the west side of Anaho Island (39° 58′ N Lat, 119° 31′ W Long). Coll. September 1955 by W. S. Broecker and P. C. Orr (see Broecker and Walton, 1959).

L447B.	**Pyramid Lake, Nevada**	**−59±7**	**−22.3**	**−68±7**

Chubs living in the lake (40° 00′ N Lat,

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Lamont No.	Locality	δC^{14}	δC^{13}	ΔC^{14}
	Long) at Sutcliffe. The water was processed immediately by acidifying, then sweeping with CO_2-free air. The CO_2 released from the water was collected in CO_2-free KOH. Coll. August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).			
L-447A.	**Walker Lake, Nevada** Chubs living in the lake (38° 46′ N Lat. 118° 46′ W Long). Coll. February 1958 by K. Johnson (see Broecker and Walton, 1959).	**−15±10**	**−20.9**	**−24±10**
L-415ZZ.	**Walker Lake, Nevada** Plants growing completely under water on the west side of the lake (38° 43′ N Lat, 118° 43′ W Long). Coll. August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).	**−3±7**	**−20.6**	**−12±7**
L-415CCC.	**Mono Lake, California** Dissolved bicarbonate extracted from 20 gal of water taken 50 ft offshore 2 mi N of Leevining (37° 59′ N Lat, 119° 08′ W Long). Sample was processed by the same procedure described in L-415KK. Coll. September 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).	**−130±7**	**2.3**	**−184±7**
L-415FFF.	**Great Salt Lake, Utah** Brine shrimp coll. near shore (40° 50′ N Lat, 112° 35′ W Long) June 1958 by R. Cohenour of Salt Lake City, Utah (see Broecker and	**−111±7**	**−16.3**	**−132±7**

Lamont No.	Locality	δC^{14}	δC^{13}	ΔC^{14}

August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).

L-415E. Woodruff, Utah **−138±7** **−22.0** **−150±7**

Plants growing completely under water on the silt bottom of the Bear River 2 mi E of the intersection of Utah Routes 39 and 3 (41° 30′ N Lat, 111° 06′ W Long). Coll. August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).

L-415F. Woodruff, Utah **−122±7** **−6.7** **−160±7**

Shells from Bear River at same locality as L-415E above. Coll. August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).

L-415N. Minkcreek, Idaho **−244±8** **−25.9** **−254±8**

Plants growing completely under water in the Bear River 5 mi E of Route 34 (42° 13′ N Lat, 111° 45′ W Long). Coll. August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).

L-415P. Corinne, Utah **−152±13** **−21.9** **−165±13**

Carp caught in the Bear River near Route 191 bridge (41° 30′ N Lat, 112° 07′ W Long) by L. Jensen during September 1957 (see Broecker and Walton, 1959).

L-415GG. Wadsworth, Nevada **−33±9** **−27.4** **−30±9**

Plants growing completely under water in the Truckee River near the U. S. Route 40 bridge (39° 38′ N Lat, 119° 17′ W Long). Coll. August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).

L-415BB. Dunphy, Nevada **−189±7** **−10.2** **−233±7**

Shell *Anodonta californiensis* Lea living in the Humboldt River near the Route 40 bridge (40° 42′ N Lat, 116° 30′ W Long). Coll. August 1957 by A. Walton and W. S. Broecker.

L-415II. Nixon, Nevada **−112±7** **−34.6** **−101±7**

Plants growing completely under water in the Truckee River 5 mi S of the agency bridge (39° 46′ N Lat, 119° 20′ W Long). Coll. August 1957 by W. S. Broecker and A. Walton (see Broecker and Walton, 1959).

Lamont No.	Locality	δC^{14}	δC^{13}	ΔC^{14}
L-415LL.	**Reno, Nevada** Plants growing completely under water in the Truckee River 1 mi N of the center of town (39° 31′ N Lat, 119° 48′ W Long). Coll. August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).	**6±7**	**–18.8**	**–7±7**
L-415NN.	**San Francisco Flycasting Club** Plants growing completely under water in the Truckee River about 5 mi E of Truckee, California (39° 23′ N Lat, 120° 07′ W Long). Coll. August 1957 by W. S. Broecker and A. Walton (see Broecker and Walton, 1959).	**–40±7**	**–18.8**	**–53±7**
L-415OO.	**San Francisco Flycasting Club** *Margaritifera margaritifera falcata* (Gould) shell living on a sandy shoal in the Truckee River at the same locality as L-415NN above. Coll. August 1957 by W. S. Broecker and A. Walton (see Broecker and Walton, 1959).	**6±7**	**–4.7**	**–34±7**
L-415PP.	**San Francisco Flycasting Club** Meat from Sample L-415OO above.	**–23±7**	**–27.0**	**–20±7**
L-415RR.	**Wellington, Nevada** Plants growing completely under water in the West Walker River at the Nevada Route 3 bridge (38° 46′ N Lat, 119° 23′ W Long). Coll. August 1957 by W. S. Broecker and A. Walton (see Broecker and Walton, 1959).	**–4±7**	**–10.0**	**–34±7**
L-415YY.	**Schurz, Nevada** Plants growing completely under water in the Walker River at the Route 95 bridge (38° 57′ N Lat, 118° 48′ W Long). Coll. August 1957 by A. Walton and W. S. Broecker (see Broecker and Walton, 1959).	**5±9**	**–24.4**	**4±9**
L-448.	**Composite of Shells** Equal portions of 14 shell samples coll. between 1885 and 1931 from nine different mid-continent rivers, including the Ohio, Allegheny, Schuylkill, Tuscarawas, and Mahoning Rivers. All but two samples come from Ohio or Pennsylvania. Subm. by J. Parodiz, Carnegie Museum. For details as to species and exact collection localities, see Broecker and Walton (1959).	**–287±9**	**(0±5)**	**(–328±11)**

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Lamont No. Locality δC^{14} δC^{13} ΔC^{14}

C. Groundwater Samples

L-393. Wayne County, Indiana –420±10 (0±7) (–470±12)

Calcareous tufa forming in water flowing out of a glacial gravel deposit 5.5 mi S and 1.25 mi W of the Wayne County Courthouse, Richmond, Indiana (39° 50′ N Lat, 84° 53′ W Long). Coll. during the summer of 1957 by J. Thorp, Earlham College.

L-443A. New River Cave, Va. –252±15 (–5±15) (–292±21)

Outer 1 mm of material scraped from a currently forming stalagmite at a cave near New River, Virginia (37° 09′ N Lat, 80° 35′ W Long). Coll. 1956 by R. Gurnee, National Speleological Society, about 1 mi back in the cave (see Broecker and Walton, 1959).

L-373. Bronx Zoo, New York –130±15 (0±7) (–180±17)

Stalactites forming in a pipe gallery under the Rockefeller Fountain (40° 51′ N Lat, 73° 57′ W Long). Coll. November 1956 by D. Revelle, National Speleological Society; subm. by R. Gurnee.

D. Hotsprings Sample

L-288J. Pyramid Lake, Nevada –81±10 1.0 –133±10

$CaCO_3$ forming 3 ft above the lake level just below the orifice of a hot spring which flows out of the west side of the Pyramid (39° 59′ N Lat, 119° 30′ W Long). Coll. August 1955 by P. C. Orr and W. S. Broecker.

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