ABSTRACT

GEOPHYSICAL IMPLICATIONS OF RADIOCARBON MEASUREMENTS

by

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The principle and basic assumptions for the existence of natural C^{14}, and its usefulness for dating are described. However, it is now known that the atmospheric inventory of C^{14} has not been constant over the past 7000 years, as originally assumed. Determinations by C-14 dating of tree-ring-dated sequoias and bristlecone pines have pointed out inconsistencies (oscillations) in the inventory (Fig. 7).

Possible reasons for the long-term trend and the smaller oscillations of shorter duration are discussed. It has been demonstrated by at least three separate experiments that the mixing rate of the atmosphere is rapid (of the order of two years) so that the results obtained with the dendro-dated samples from the southwestern U.S.A. may be applied on a worldwide basis. As for the basic causes of the deviations in the atmospheric inventory of C^{14}, a primary consideration is that of the constancy of the cosmic ray intensity. Measurements of the ratios of isotopes other than C^{14} in meteorites indicate that there have not been

pronounced changes at least within a precision of 5 percent.

However, the magnetic field of the Earth does have an effect upon the numbers of cosmic rays reaching our upper atmosphere that cause the creation of neutrons which react with N^{14} to form C^{14}. The stronger the field, the fewer cosmic rays and vice versa. Changes which have taken place have been determined by archaeomagnetic measurements of fired clays and lava flows. Calculation of the effect of the long-term magnetic changes over the past 9000 years with a sinusoidal solution for the relationship indicates that the change in the Earth's dipole field accounts for only about one-half of the long-term variation in C^{14} contents. "Recent" pole reversals may account for the cause of the magnetic change, but cannot be correlated precisely until the dendro-dates are extended to at least 11,000 years ago, or if we live through a pole reversal.

The shorter-term oscillations may be due to secular variations which, in turn, are caused by changes in the nondipole field. In Fig. 7, one sees a tendency for oscillations at 400 year intervals, but also one notes that there is no truly sinusoidal periodicity. This tends to confirm the correlation with the random magnetic changes.

Since the change in the dipole field is inadequate to

account for the entire long-term trend in C^{14} deviations, there may be a contribution from the magnetic field of the Sun. The Sun's field is comparable to that of the Earth's in strength, but is extremely variable, and has only recently been measured with any accuracy. As a secondary cause and effect, climatic changes must be considered.

In summary, part of the long-term deviation in the atmospheric inventory of C^{14} is due to changes in the Earth's dipole moment; the shorter-term oscillations may be related to variations in the Earth's nondipole field, the correlation is unclear except for a tendency for both to have periods of roughly 400 years, although there is no true sinusoidal periodicity; there may be a contribution from the varying magnetic field of the Sun; and there is a slight possibility of a contribution from the explosions of supernovae.

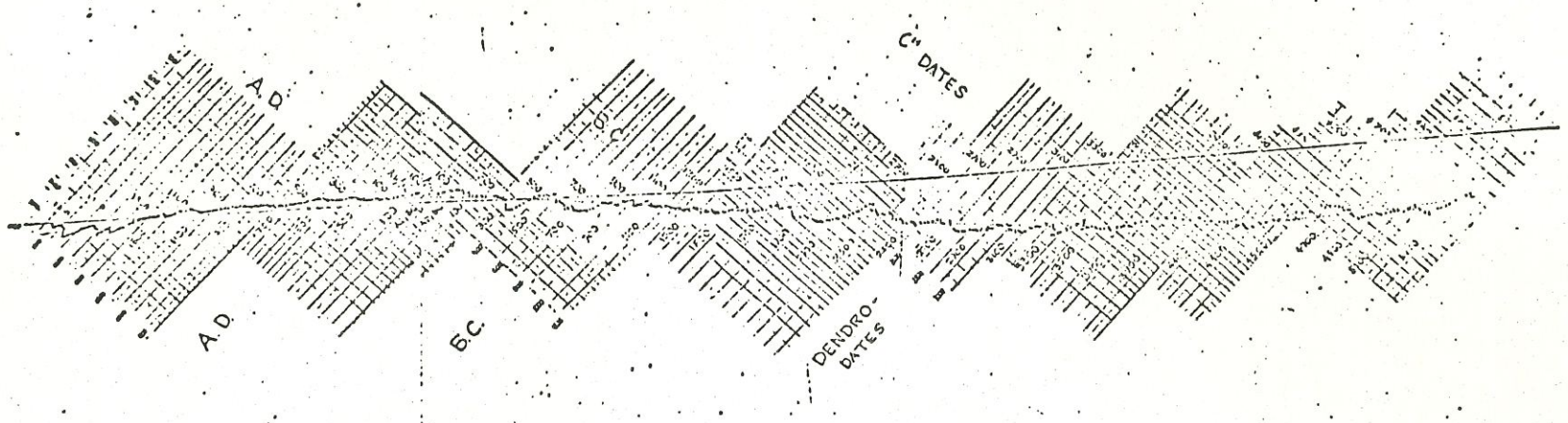

Fig. 7. University of Pennsylvania plot of weighted 9-sample averages along with curve from Eq. 4 in which is the best fit on the average for 600 dates (dashed line), and 45° correspondence line (solid line) are shown.

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The technique of radiocarbon (C^{14}) dating is almost twenty-five years old. In the beginning there was some reluctance to accept C^{14} dates; then, there was general reliance upon them. However, we have now learned that one of the basic assumptions of the method - namely, the constancy of the atmospheric inventory of $C^{14}O_2$ is not strictly valid. This problem and the means of assessing it are the main subjects of this thesis.

II. Historical Background

Shortly after the discovery of artificial radioactivity, the existence of natural "cosmic radioelements" was anticipated by A. V. Grosse (G2). More than a decade later in 1947, W. F. Libby in collaboration with Grosse and others (A1) demonstrated that natural C^{14} does exist. The proof was established by enriching methane from sewage gas in thermal diffusion columns. With this encouragement, Libby and his students then pioneered in the development of greatly improved techniques for the detection of this low-level radioactivity that enabled them to measure C^{14} without enrichment. They used a screen wall counter which had been developed by Libby (L3) and a surrounding ring of anticoincidence counters, all enclosed in a massive shield of iron.

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These improvements led to the reduction of the background of unwanted counts below the level of natural C^{14}, and, hence to its detection without enrichment.

The technique of counting C^{14} as solid carbon with both mechanical shielding of iron in combination with the more effective electronic shielding consisting of an anti-coincidence ring of cosmic ray counters surrounding the carbon counter is now well known (L4). In 1949, little was known about gas proportional or about scintillation counting, but geiger counting of solids was in vogue. Therefore, it was logical to convert samples to solid carbon - the element itself, not even a compound, which therefore contained as much carbon as possible per mole. This method was used by Libby and other laboratories for more than five years. However, there were two basic weaknesses in solid carbon counting - first, (β emitted has maximum energy of 160 kev) self-absorption which reduces the counting efficiency and second, liability to contamination, especially, from fall-out from atomic bomb tests. For these reasons, workers began to experiment with gas counting techniques and a few, with scintillation detection. The methods in use now are proportional counting of pure CO_2, CH_4, or C_2H_2 and liquid scintillation counting of C_6H_6. The results obtained at the

University of Pennsylvania were obtained by CO_2 counting.

III. Explanation of C^{14} Dating

Before we consider our present day problems in detail, we must understand why the existence of natural C^{14} provides a method of dating. First of all, where does it come from? It is formed in the upper atmosphere by the reaction of ordinary nitrogen (N^{14}) with neutrons (n).

$$N^{14} + n \rightarrow C^{14} + H'$$

Since there is an abundant supply of nitrogen, one notes that the constancy of this production is dependent upon the supply of neutrons. The neutrons are produced by cosmic rays, and the quantity is dependent upon the cosmic ray intensity and therefore also upon the intensity of the magnetic field of the earth--when the field is stronger, fewer cosmic rays reach the upper atmosphere and vice versa. After production of the C^{14}, it becomes approximately one part in 10^{12} parts of our atmospheric carbon dioxide (a very small amount). This, in turn, is in equilibrium with terrestrial life and with the oceans, the largest reservoirs of carbon. For dating, we must assume that the balance among the reservoirs has remained constant, and that the mixing rate of the entire atmosphere is rapid with respect to the average life of an atom of C^{14} --8033 years.

That all of these conditions are fulfilled within a precision of approximately 10 percent was first demonstrated by Arnold and Libby (A3) and a surprisingly accurate estimate of the world-wide distribution of natural C^{14} was made by Anderson and Libby (A2). The present best estimate of the mean specific activity of C^{14} in equilibrium with the biosphere is 13.56 ± 0.07 disintegrations per minute per gram, abbreviated dpm/g (K1).

Due to the mechanism of photosynthesis, all living vegetation is in equilibrium with this atmospheric $C^{14}O_2$ and almost all forms of terrestrial life, that consume this vegetation, contain the same proportion of C^{14}. An equilibrium balance between intake and radioactive decay is achieved. However, as soon as a plant or creature dies, it no longer "breathes" or eats and it ceases to acquire additional C^{14} from the atmosphere. Its radiocarbon content, therefore, begins to decrease and its rate of radioactive decay is dictated by the half-life of C^{14}. It decays back to N^{14} by emission of a beta particle as shown below:

$$C^{14} \rightarrow N^{14+} + \beta^-$$

Therefore, the accuracy of dating is dependent also upon the precision with which the half-life is known. In 1951 the best estimate of the half-life was 5568 ± 30 years

(E2,J1,M4,E3,H1) and this value continues to be used for the calculation of dates published in Radiocarbon. However, the present best estimate, as a result of three more recent determinations is 5730 ± 40 years (G1). The latter was accepted as the most probable value at the Fifth Radiocarbon Dating Conference held in Cambridge, England, in 1962. Because of the value of the half-life, the element carbon, found universally on Earth, provides a tracer for age determination for the periods of interest to most archaeologists, Pleistocene geologists, and many others.

IV. Age Calculation and Statistical Uncertainty

In order to calculate a C^{14} date, let us assume for the moment that the half-life is known to our satisfaction and that the production rate of C^{14} is constant. If we ignore other factors, terrestrial creatures and plants, in equilibrium with the atmosphere, will have a constant amount of C^{14}. (This is not absolutely true, especially for the past 100 years during which the atmospheric C^{14} was first depleted by the combustion of inert fossil fuels not containing C^{14} and then augmented by nuclear bomb explosions.) This static condition persists until the living subject dies. Then, its C^{14} is no longer replenished and it decays only. Since this is the decay of a radioactive element, it follows

the familiar exponential decay equation:

$$I = I_o e^{-\lambda t} \qquad (1)$$

where

I = activity of the sample when measured.

I_o = original activity of the sample (this value is obtained from samples of known age, approximately 100 years old, but corrected for zero age).

λ = decay constant = $0.693/T_{\frac{1}{2}}$ with $T_{\frac{1}{2}}$ the half-life.

t = time elapsed.

If we feed the value of the half-life into this equation (5568 years in this case), we have the simple formula

$$t = \log\frac{I_o}{I} \times 18.5 \times 10^3 \text{ years} \qquad (2)$$

for routine age calculations.

Not so obvious in these simple equations is the fact that radioactive disintegration is a random process. One cannot predict when any particular atom will decay; one can say only that after a certain length of time, on the average a certain quantity will have disintegrated. In radioactive parlance this uncertainty is quoted in terms of statistical deviation. For uniformity in radiocarbon dating, it has been customary to quote the standard statistical deviation (or one sigma) with each date. This so-called one sigma tolerance means that there is the probability in two cases out of three that the date lies within the range quoted. If the one sigma tolerance is doubled,

then there is greater assurance that the true date lies within such a range--specifically, 21 times in 22.

In most laboratories, the tolerance quoted includes the statistical uncertainty inherent in the counting of the unknown sample, and of the background, and calibration samples. Since, at the moment, we have neglected other possible discrepancies between radiocarbon and true ages, this is the minimum uncertainty and it cannot nor may not be ignored in quoting C^{14} dates.

In most laboratories, samples are counted overnight for 1000-minute intervals, and each sample is counted at least twice. If the second count is made a week after the first, this provides a check on possible radon conatmination. (In the purification of the CO_2 or other gas for counting, radon which may have come from traces of uranium and thorium in the soil mixed with the sample, is the most difficult contaminant to remove.) Since the half-life of radon is only 3.82 days, it will have decayed a measurable amount in the second week. In our laboratory, if the two counts are not statistically consistent, the sample is then counted a third or more times. Our method of statistical analysis has been explained in Satterthwaite and Ralph (S1.)

A specific example, including an age calculation, is given below.

The first step is to determine the average total counting rate ($\bar{n}_t$) and the average net counting rate ($\bar{n}_n = \bar{n}_t - b$) where b = background counting rate which is an average of all of the once-a-week checks, usually for a period of 10 weeks (unless there has been a shift in background). These are set forth as follows:

Date	Counter	n_t	n_n	$n_n - \bar{n}_n$	$(n_n - \bar{n}_n)^2$
5/1/69	I	22,623	13,085	-131	17,161
5/9/69	I	21,377	13,346	+130	16,900
		$\bar{n}_t$ = 22,000	$\bar{n}_n$ = 13,216	Σ = -1	Σ = 34,061

The chi-square test is then applied, where

$$\chi^2 = \frac{\Sigma(n_n - \bar{n}_n)^2}{\bar{n}_t} = \frac{34,061}{22,000} = 1.55$$

From a table of chi-square factors (S3), we find $P(\chi^2) \geq 0.2$. The criterion for standard statistical consistency is that $P(\chi^2)$ be > 0.1. Therefore, these two counts pass the test, and since they are consistent, the statistical deviation of the average, σ_s equals $\sqrt{\frac{(\bar{n}_t + b)N}{N}}$ where N equals the number of counting runs. In this example,

$$\sigma_s = \frac{\sqrt{(22,000 + 8785)2}}{2} = 124$$

(Note that in this particular example, there was a shift in background between the two counts. This was due to the replacement of a geiger counter which became faulty after May 1, 1969.) During this counting interval in May, the average "zero age" (I_o) counting rate, based on the 16 consistent measurements of our 100-year-old oak calibration sample (based on A.D. 1950) was:

$$I_o = 31.666 \pm .054 .$$

Next, we divide σ_s by 1000 (the standard counting time) and add the sum of the squares of $\sigma_s/1000$ and .054, and take the square root, i.e.,

$$\sigma_t = \sqrt{(.124)^2 + (.054)^2} = .135$$

We then subtract and add σ_t from $\bar{n}_n$ and calculate the maximum and minimum ages from the formula

$t = \log \frac{I_o}{I} \times 18.5 \times 10^3$. With the use of a desk calculator and tables of logarithms, our worksheet appears as follows:

13.216			
.135			
13.081	2.4208	.38396	7103.3 (maximum age)
13.351	2.3718	.37507	6938.8 (minimum age)
			164.5 (difference)

164.5/2 = 82

6939 + 82 = 7021 = B.P. (before present) age

We next subtract 19 years (in 1969) from this to standardize

the age to B.P. 1950, and then subtract 1950 to find its A.D. - B.C. age. To convert the date to the 5730 half-life, we multiply the B.P. age by 1.030 and obtain the following dates:

Half-life	B.P. 1950	A.D. - B.C.
5568	7002±82	5052±82 B.C.
5730	7213±84	5263±84 B.C.

For a sample of known age, all of which are counted at least three times and are corrected for C^{13}/C^{12} (natural fractionation), a typical worksheet is shown on the next page.

V. Interlaboratory Calibration and Mass-Spectrometric Measurement of C^{13}/C^{12}

Our calibration sample on which I_o is based consists of a section of tree-ring dated white oak. From this, we select samples which span no more than 10 tree rings in the range of 110 to 130 years B.P. From the counting rate, I, and the known age, t, we obtain I_o from equation 2.

For interlaboratory calibration, however, a sample which could be made in bulk with a specific amount of C^{14} was desirable. The National Bureau of Standards provided this in the form of oxalic acid (N.B.S. No. 4990). From the measurement of this and wood calibration samples in the

P-1856 P - SW - INY - 33

Known Age
4125 ± 5 B.C.
6098 B.P. 1973
6075 B.P. 1950

Pinus aristata

	I_o	n_t	n_n	$n_n - \bar{n}_n$
I 3/3/73	29,845	25,366	15,718	-57
I 3/4/73	"	25,305	15,657	-118
I 3/5/73	"	25,597	15,949	174
		25,423	15,775	= -1

$\Sigma^2 = 47,449$

$\chi^2 = 1.866, p(\chi^2) \sim 0.40$ $\sigma_s = 108$

$I_o = 29.845 \pm .049$

6098/18,500 = .32962

log 15.775 = .19797

log I_s = .52759, then $I_s = 33.697 \pm .108$

$I_o = 29.845 \pm .049$

$+ 3.852 \pm .119 =$

$+ 12.907 \pm .399\% =$

$+ 129.07 \pm 3.99$ mils

Correction for C^{13}/C^{12} $2 \times \delta C^{13} = + 7.76$ mils

$\Delta = 129.07 - (7.76 \times 1.129) = 120.31 \pm 3.99$ mils

$- 8.761$ mils

15.775 (1- .009) = 15.633 ± .108

15.633 ± .108

.119

15.514	1.9237	.28414	5256.6
15.752	1.8947	.27754	5134.5
			122.1

(5568 half-life) 5196 ± 61; 5173 ± 61; 3223 B.C.

(5730 half-life) 5352 ± 63; 5329 ± 63; 3379 B.C.

range of A.D. 1840 to A.D. 1860, in many laboratories, it has been found that the age-corrected wood samples have activities equal to 95 percent of the oxalic acid standard sample. An absolute calibration of the oxalic acid standard sample was made also (K1).

The oxalic acid furnishes a sample homogeneous in C^{14}, but this compound was an unfortunate choice because it is easily fractionated in the process of converting it to CO_2. The amount of fractionation of this and of other samples can be determined by the mass spectrographic measurement of C^{13} - C^{12} ratios. It is then assumed that the effect upon the C^{14} isotope is double that of the C^{13} (C3). Craig (C4) made careful measurements of CO_2 samples from the combustion of the NBS oxalic acid standard submitted by 13 radiocarbon laboratories and found that

δC^{13} = -19.3 mils (dry combustion)

δC^{13} = -19.6 mils (wet combustion)

whereas $\delta C^{13} = 1000\ [R/R_{PDB} - 1]$. $R = C^{13}/C^{12}$ of oxalic acid.

$R_{PDB} = C^{13}/C^{12}$ of PDB - the Chicago limestone C^{13} standard. For average wood δC^{13} = -25 mils relative to PDB. The C^{13} of the oxalic acid is, therefore, enriched relative to wood by about 6 mils.

Since Craig reported these measurements in 1961, most

laboratories have processed many batches of oxalic acid and have found that fractionation is severe and erratic in the wet conversion to CO_2, but is also variable in dry combustion. Therefore, it is necessary to measure δC^{13} for each batch. By informal agreement among several laboratories, the value of $\delta C^{13} = -19$ mils is assumed to be standard and the corrected activity is obtained by the formula:

$$0.95\ A_{ox} = 0.95\ A'_{ox}\left[1 - \frac{2(19 + C_{ox}^{13'})}{1000}\right]$$

where A'_{ox} and $\delta C_{ox}^{13'}$ are based on the actual counts and mass-spectrometric measurement (F5).

Because of these difficulties with oxalic acid and also because the supply is almost exhausted, many laboratories have selected a secondary standard, namely wood, which was described previously. After checking against the oxalic acid, these provide each laboratory with an inter-calibrated standard which is more representative of the types of materials to be dated.

For these woods and for other tree-ring dated series which are important for geochemical studies, and also for materials such as shells that may differ significantly from average wood, it is advisable to make spectrographic measure-

ments of C^{13} - C^{12} ratios. The deviation from normal is then expressed as

$$\Delta = \delta C^{14} - (2\delta C^{13} + 50) \quad (1 + \frac{\delta C^{14}}{1000})$$

where Δ is the per-mil deviation from the modern C^{14} standard (i.e., from 0.95 A_{ox} as defined previously or from age corrected wood) and δC^{14} and δC^{13} are the observed per mil deviations from C^{14} and C^{13} standards (F5). Fortunately, for most wood and charcoal samples the δC^{13} variation is small, and may be neglected in dating samples of unknown ages.

VI. Basic Assumptions

We now return to the fundamental assumption which was mentioned briefly in the beginning of this paper - namely, that the biospheric inventory of C^{14} has remained constant during the past 50,000 years or so. We know now that this assumption is not precisely true. Papers and discussions of most of these problems were reported at the Nobel Symposium XII held in Uppsala, August, 1969 (01). The problem is illustrated in Fig. 1.

A. Mixing Rate

Fortunately, numerous experiments (L1, T1, and others) have demonstrated that the mixing rate of C^{14} in the atmos-

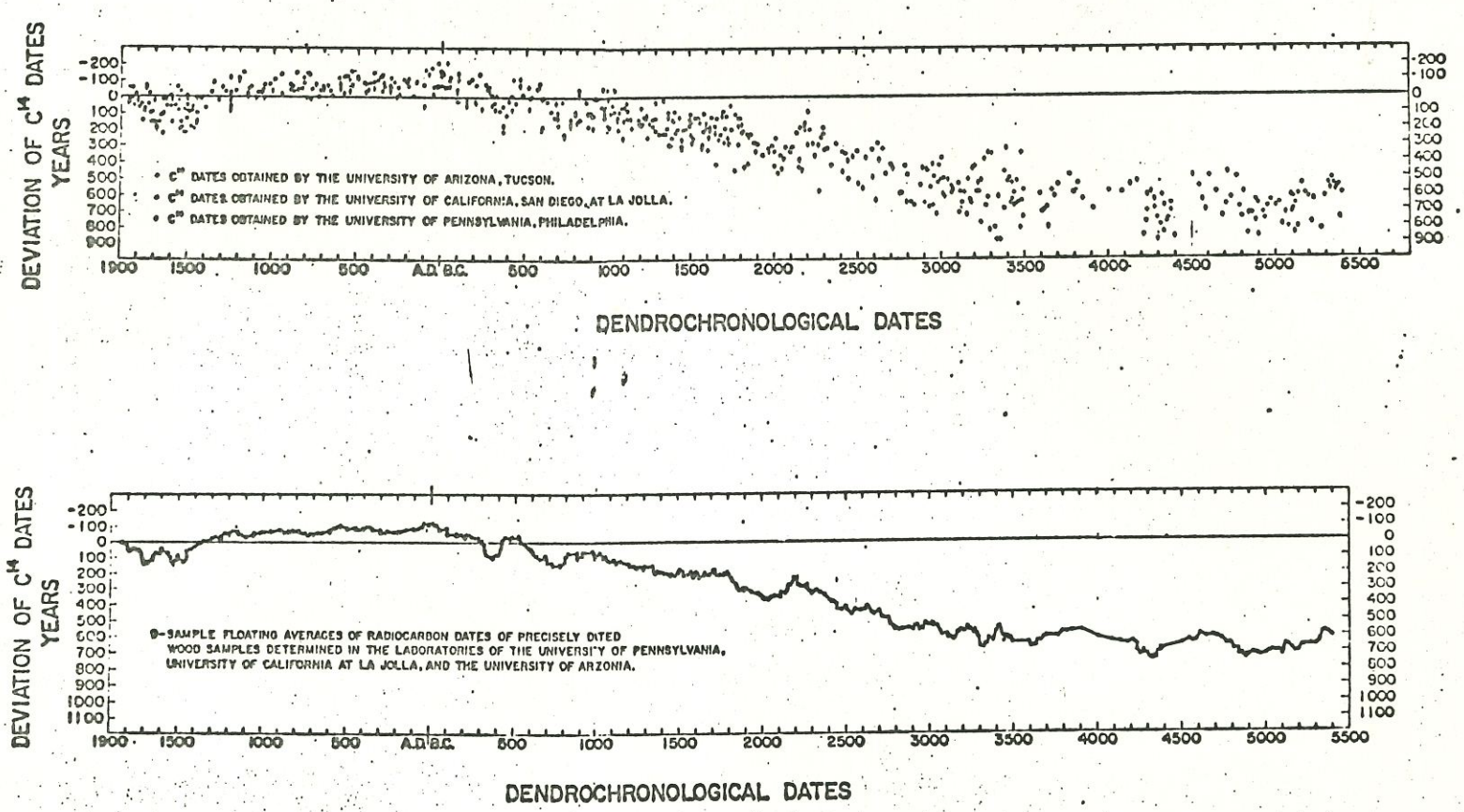

Fig. 1. C-14 versus Dendrochronological Dates copied from Michael and Ralph (M3).

Top Figure: Individual dates obtained by the three laboratories.

Bottom Figure: 9-sample floating averages (not weighted), of the individual dates shown above.

phere is rapid--of the order of two years even between hemispheres--so that we can consider the atmosphere as a whole. A study by Haugen (H2) in which he compared major trends in tree ring indices for Alaska, the Urals, Scandinavia and Labrador resulted in highly significant correlations among these areas from A.D. 1650 on, and supports the proposition of a rapid mixing rate. Incidentally, this demonstrates also that a C^{14} date for a particular age will be the same all over the world, except for small "pockets" with poor circulation.

B. Constancy of the Cosmic Ray Intensity

It is more difficult to obtain a precise assessment of the constancy of the cosmic ray intensity since we are looking for comparatively small variations in the concentration of C^{14} activity. Studies of the decay series of other nuclides in meteorites indicate that there have been no major changes in cosmic ray intensity during the past 300,000 years (H3). Crevecoeur (C5) has extended this period to 5 million years. For more recent time ranges (less than 2000 years), a study of Ar^{37}/Ar^{39} ratios in iron meteorites by Fireman and Goebel (F4) indicates that the cosmic ray flux near 1 AU (AU = average distance between Earth and Sun), did not differ significantly in the spring

of 1969 from the previous several hundred years. The accuracy of measurement of these ratios was 5 percent--much better than most previous determinations of isotopic ratios in meteorites. However, the basic difficulty has been that the orbits in space of the meteorites have been unknown and that the cosmic ray flux received varies with distance from the Sun. Fortunately, this difficulty will soon be overcome because tracking stations, set up in seven middle Western states in 1964 have, at last, recorded the trajectory of a meteorite which fell near Lost City, Oklahoma on January 3, 1970 (S9). Also at Pribram in Czechoslovakia the precise trajectory of a meteorite was recorded in 1959. When these and others have been studied, more will be known about the constancy of the cosmic ray intensity.

C. Sunspots

Numerous attempts have been made to correlate short-term C^{14} fluctuations with sunspot activities and cycles. Results have given correlations both negative (L6,B5) and positive (S5,H4). The difficulties are that first, the statistical uncertainty in C^{14} dates is greater than the short-term sunspot cycles, and second, the sunspot data prior to A.D. 1750 are scarce and unreliable.

D. Introduction to the Problem of the Magnetic Intensity

Another important factor is the constancy of the

magnetic field of the Earth in past times. The strength of the magnetic field affects the intensity of cosmic rays which reach the upper atmosphere to react with nitrogen. When the magnetic field is stronger, more cosmic rays are deflected from the upper troposphere and, therefore, do not produce C^{14}. This, as one of the more likely causes, will be discussed later.

VII. Tree-Ring Dated Samples

We may not have direct and precise methods to measure the constancy of production of C^{14}, the subsequent equilibrium balance between the atmosphere and the oceans, and its absolute decay rate, but we can evaluate radiocarbon years in terms of true ages by means of samples of known age, as well as shed some light upon the basic causes of the oscillations.

Dendrochronology, by means of the tedious process of cross-dating, has provided wood samples of known age, accurate to within a year. For the correction of C^{14} dates, the first prerequisite was a series of long-lived trees. (It has been found that only the outer growth ring of a tree is in equilibrium with the atmosphere and that the C^{14} content of the adjacent inner ring is one year old and so on until one reaches the maximum age or "pith" of the tree.

These long-lived series were first provided by the sequoias (Sequoia gigantea) (D4) which afforded a scale reaching 3100 calendar years back in time. More recently, starting in 1954, Schulman (S2) and then Ferguson (F2,F3) working with bristlecone pines (Pinus aristata), have succeeded in extending the range of known-age trees to approximately 8000 years before the present. However, because of the paucity of wood of this extreme age, precisely dated samples reaching to only 7350 calendar years before present have so far been available for radiocarbon dating.

For the past 15 years, three laboratories have been obtaining C^{14} dates from samples (spanning about 10 years each) of these long-lived dendro-dated trees. About 10 percent of the samples are exact duplicates or triplicates; others are not. The three laboratories are at the University of Arizona in Tucson, The University of California at San Diego in La Jolla, and the University of Pennsylvania in Philadelphia.[1]

In all three laboratories, these C-14 dates have been obtained with extreme care. In our laboratory each sample

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[1]I acknowledge with gratitude financial support from the National Science Foundation for our known-age dating program. This has been supported since 1957 with NSF grants G-3281, G-5608, G-14094, GP-405, GP-3778, GA-993 and GA-12,572.

has been counted at least 3 times (1000 minutes each), and each has been corrected for C^{13}/C^{12} even though these variations are small. The resultant statistical uncertainties are of the order of $\pm$ 0.5%. All three laboratories presented results individually, in one form or another at the Twelfth Nobel Symposium held in Uppsala, Sweden in 1969 (D1,S7,M2). At Uppsala, it was clear that the C-14 dates of all three laboratories were in excellent agreement on the average, but that there were differences among individual or small groups of dates. (See Reference 01, pp. 110, 597, 615-618, 619-624, and separate plates I and II.)

At the more recent international C^{14} conference in Lower Hutt, New Zealand, in October 1972, it was anticipated that after three more years spent in obtaining greater numbers of C^{14} dates, from precisely dated wood samples, better agreement could be achieved. With this in mind, the laboratories of Arizona (D2) and Pennsylvania (M3) presented separately composite plots of all C^{14} dates obtained by all three laboratories, with the Arizona laboratory adding a few dates determined by Stuiver some time previously (S6). La Jolla presented its 1969 (Uppsala) "calibration curve" derived from La Jolla dates only and containing many doubtful short-term oscillations based on very few dates. Again,

there was good agreement on the average, but no decision was reached as to which calibration method best expressed "true" calendric dates. However, from the radiocarbon dating of tree-ring-dated sequoias and bristlecone pines, fundamental geophysical and geochemical information is beginning to emerge. These dating efforts have been in process for more than ten years, but it is just now that the dendrochronological time scale is being extended sufficiently back in time to see more than a half-period of the long-term C^{14} deviations. This result (Fig. 1), which is of great interest in itself, is essential also for the interpretation of the indicated shorter-term deviations superimposed upon it.

VIII. Calculation of the Magnetic Effect

It now seems most likely that part of the long-term divergence of C^{14} dates was caused by changes in the intensity of the Earth's magnetic field. As illustrated in Fig. 2 from Bucha (B6) the magnetic intensity, probably on a worldwide basis, reached a maximum in Roman times and a minimum around 4000 B.C. (in terms of radiocarbon years). From these data, one may now calculate the effect of these changes in magnetic intensity upon the atmospheric C-14 inventory.

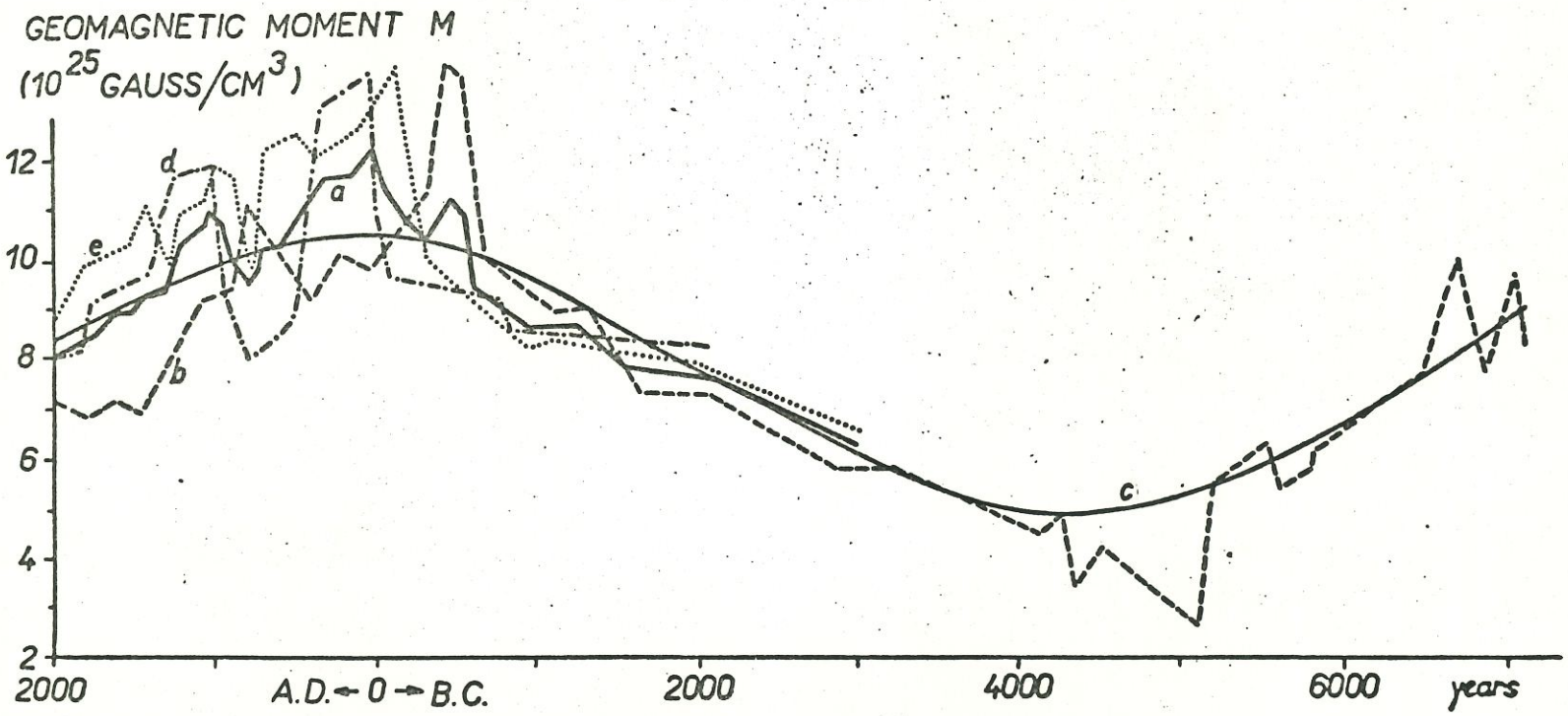

Fig. 2. Changes of the averaged reduced Earth's magnetic moment (solid line a) determined from European (dashed line b), southwestern North American (dash-dotted line d), and Japanese (dotted line e) results. The smoothed curve c expresses the average. After Bucha (B6, p. 82).

The rate of production of C^{14}, P(t) is proportional to the average cosmic ray intensity which is influenced to some extent by the magnetic field of the Earth. The relationship suggested by Elsasser, Ney, and Winckler (E1) is:

$$P(t) = \frac{C_2}{[M(t)]^{0.52}} \quad \text{where} \qquad (3)$$

C_2 is a constant and M(t) is the magnetic dipole moment (dependent upon time, t). Since the experimental evidence now indicates that the changes were cyclic (at least for one cycle), let us try a cyclic solution rather than the exponential one proposed by Elsasser et al.. If we take our boundary conditions from the magnetic data of Bucha (B6) as shown in Fig. 2, and assume that these changes have been worldwide, a solution may be found as follows:

Let $P(t) = \frac{K}{[M(t)]^{\frac{1}{2}}}$ where $M(t) = 1 - A \sin\omega t$

K is a constant and A is the amplitude of the cycle and is estimated to be $\frac{1}{2}$ from Bucha's archaeomagnetic data (B5, p.72). When we take the square root of M(t) as shown in Fig. 3, we find that the average A becomes 0.26 or approximately $\frac{1}{4}$. We now have

$$P(t) = K\,(1 - \tfrac{1}{4} \sin\omega t)$$

If I is the amount of C-14 in the atmosphere, we then

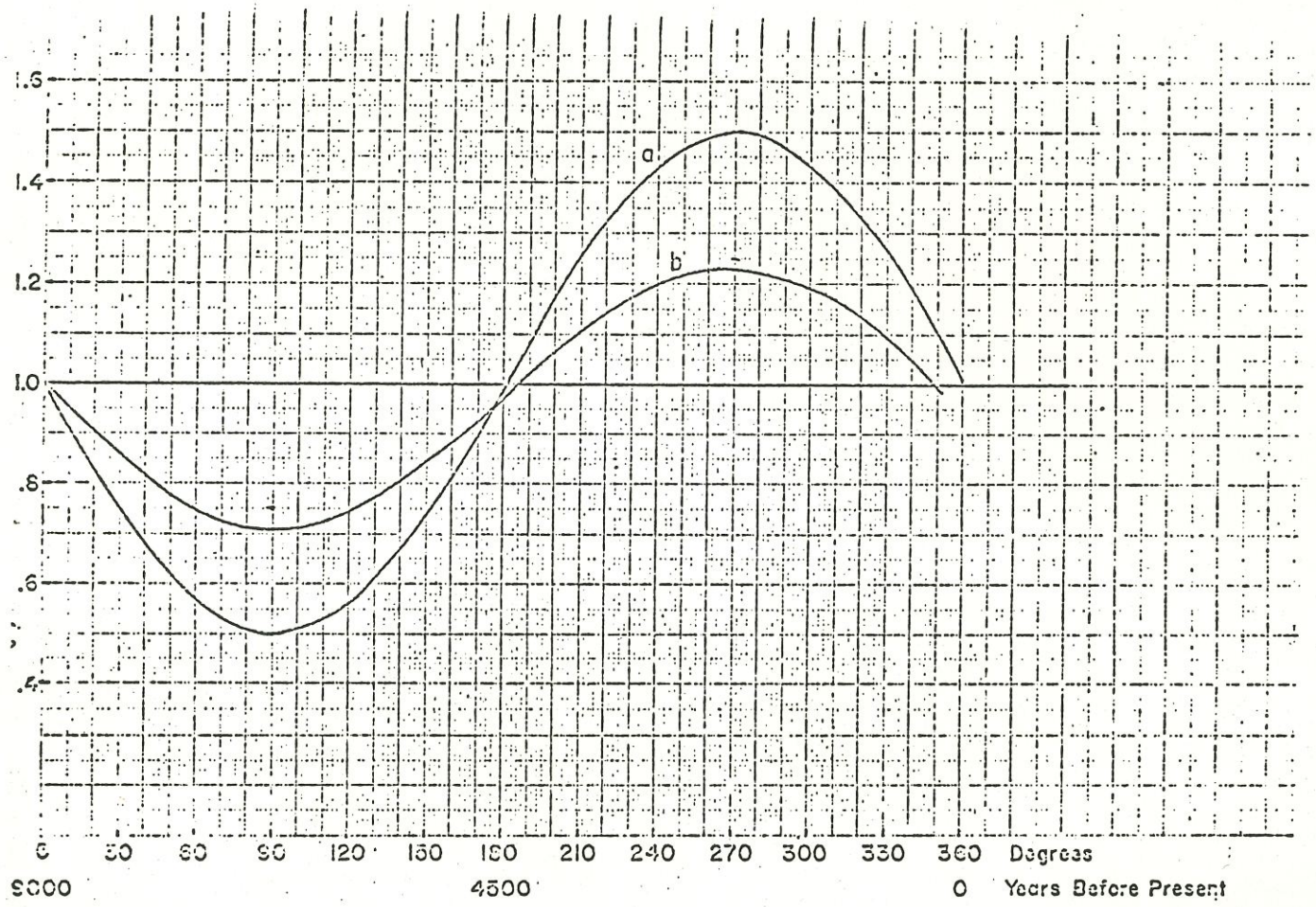

Fig. 3. F_t/F_o is the ratio of the change in magnetic intensities, past and present, respectively. The values have been estimated from Bucha's Fig. 3.11, p. 72 (B6).

$$\text{Curve a} = 1 - \frac{1}{2}\sin\omega t \qquad \text{Curve b} = \left(1 - \frac{1}{2}\sin\omega t\right)^{\frac{1}{2}}$$

have the differential equation

$$\frac{dI}{dt} = - \lambda I + P(t)$$

(where λ = decay constant of C-14) .

Neglecting the interchange of atmospheric C-14 with the biosphere and oceans, we assume that

$$I = \frac{C}{\lambda} + E \cos\omega t + F \sin\omega t + Ge^{-\lambda t}$$

We then have the derivative

$$\frac{dI}{dt} = - E\omega\sin\omega t + F\omega\cos\omega t - G\lambda e^{-\lambda t}$$

Then,

$$\frac{dI}{dt} + \lambda I = C + (E\lambda + F\omega)\cos\omega t + (F\lambda - E\omega)\sin\omega t$$

$$= K - \frac{K}{4}\sin\omega t .$$

From this, we find that

$$C = K$$

$$F = - \frac{K\lambda}{4(\lambda^2 + \omega^2)}$$

$$E = \frac{k\omega}{4(\lambda^2 + \omega^2)}$$

and that the term $Ge^{-\lambda t}$, which represents the transient solution, has been eliminated. Then,

$$I = K[\frac{1}{\lambda} + \frac{\omega}{4(\lambda^2 + \omega^2)}\cos\omega t - \frac{\lambda}{4(\lambda^2 + \omega^2)}\sin\omega t].$$

For this cycle with A = $\frac{1}{2}$ and a period of approximately 9000 years, we have

$$\omega = \frac{2\pi}{9000} = 0.70 \times 10^{-3}$$

and for the 5730 half-life,

$$\lambda = .693/5730 = 1.21 \times 10^{-4} \text{ per year.}$$

Then,

$$I = K [8300 + 343 \cos\omega t - 59 \sin\omega t] .$$

At t = 9000 years, $\omega t = 6.3^{o}$ and

$$\cos\omega t = 0.994$$

$$\sin\omega t = 0.110$$

$$I = K [8300 + 334]$$

or I has changed by 4%.

To find the lag (t_L) in this effect, we have the ratio of the sine and cosine terms from the equation for I:

$$\frac{-59}{343} = - 0.172 = \omega t_L$$

and $t_L = \frac{-0.172 \times 9000}{2\pi} = - 246$ years.

Therefore, within a magnetic cycle of 9000 years' duration, the effect of changes in magnetic intensity upon the atmospheric inventory of C-14 will be periodic (also, for that reason, within a period of 9000 years) with an

amplitude of 334 years and a lag of 246 years. As yet we are not certain that C^{14} contents are conforming to the cyclic pattern of the magnetic changes. This will not be known until the tree-ring chronology has been extended back in time another 3000 years and more archaeometric measurements are made covering the interval of 10,000 to 7,000 B.P.

The magnitude of the observed C^{14} deviation at 7000 B.P. is now known to be much greater (see Fig. 1)-namely, 750 years or about 10%. If the assumptions made in these calculations are correct, one may state that changes in the magnetic intensity in past times are not the sole cause nor the dominant cause of changes in atmospheric C^{14} contents. The magnetic effect may be found to have even less importance if it is found that the magnetic changes described by Bucha (B6) are not worldwide. Even though similar results have been reported from France (T2), the Soviet Union (B8), Japan (N1), Czechoslovakia (B7), and the Americas (B7), measurements made by Athavale (A4) in India show only a 15 per cent negative change in magnetic intensity during the past 4000 years.

There are other uncertainties in regard to these calculations. One uncertainty, suggested by Lal and Venkatavaradan (L2), is that the ratios F_t/F_o given in

Fig. 2 may not be truly equivalent to M_t/M_o (the dipole moments, past and present, respectively). The quantities measured, F_t and F_o, (past and present) are based only on determinations of intensities. Since the field directions have not been measured, there may be possible corrections due to nondipole components and to dipole wobble.

Furthermore, values of the exponent for M(t) (Eq. 3) may range from 0.52 to 0.58 (W1, p. 487 and D3, p. 2316).

Also, for the production of C-14, the effect of vertical cut-off rigidities as functions of geomagnetic latitudes has not been taken into consideration. (Rigidity in this sense refers to the curvature of the radius of orbit of a cosmic ray. For example, at the magnetic north pole, all cosmic rays may head into the Earth's atmosphere, whereas at the equator, only the most energetic are not bent away.) These have been discussed by Lingenfelter and Ramaty (L5) and Fanselow and Stone (F1). However, since the mixing rate of the world-wide atmosphere is rapid - of the order of a few years, the neglect of the problem of rigidities may not be important.

However, if we accept these uncertainties as not very great, it now seems likely that part of the long-term divergence of C^{14} dates was caused by changesin the intensity of the Earth's dipole field, but additionally, there may have been contributions to the oscillations from climactic cycles and changes in the cosmic ray intensity prior to the rays' reaching the earth's magnetic field. These questions may be

answered when the dendrochronological scale is extended. There are, of course, the fascinating speculations as to the very basic causes of these particular changes - variations in the solar wind, "recent" pole reversals and their mechanisms, the various sunspot theories, etc.

IX. Pole Reversals

For example, in regard to the "recent" pole reversals, There is now evidence that two have occurred during the past 35,000 years, but that there was an absence of reversals between about 30,000 and 13,000 years ago (B2,B4,M5,B3 and others).[2] Until a few years ago, it was thought there had been none in the Brunhes epoch (that is, for the past 700,000 years). An almost complete pole reversal would cause a change in the Earth's magnetic intensity, and thereby affect the atmospheric inventory of C^{14}. The difficulty is that the occurrence of these fairly rapid pole reversals had not been detected previously and, therefore, the theories for their causes are not yet defined. In a way, similar to

[2] In June 1972, a search for "recent" reversed lava flows in the U.S.A. was made by the writer in the San Francisco Mountains north of Flagstaff, Arizona. One reversed lava flow may have been found at the base of Mt. O'Leary close to the point where the Bonito flow from Sunset Crater ran against the flank of Mt. O'Leary. Aerial photographs revealed that it came from a side vent. This reversal was detected in the field with a portable fluxgate magnetometer, but has not yet been measured precisely. A thermoluminescent date of one of the lava samples collected, which may easily be in error by 100%, places the flow at 8000 B.P.

their causes are not yet defined. In a way, similar to the C^{14} problem, it may not be possible to define them until more archaeo- or geo-magnetic data are determined. Also, theories for their basic mechanisms need to be developed. For example, one new attempt is that of Won and Kuo (W2) in which they have suggested that the oscillations of the inner core of the Earth "...may be linked to the mechanism of the Earth's magnetic field (W2, p. 911)". Or, conversely, an oscillation of the inner solid core of the Earth caused by an earthquake or other upheaval may affect the magnetic field. Cox (C1) had suggested that reversals with period of the order of 10^4 years "... may hold the key to understanding how polarity reversals are related to intensity fluctuations (C1, p. 237)". Cox and Cain (C2) have also presented and summarized many of these questions and problems.

If we consider the situation during a pole reversal as a time of minimum magnetic intensity and therefore of maximum cosmic ray intensity, this situation could be equated to the present intensity at the magnetic poles. On the basis of the work of M. Merker (M1), Wada and Inoue (W1), and Dergachev (D3), this situation could cause an

increase in the C^{14} inventory by a factor of 2. To verify this, we need to extend the tree-ring time scale back to at least 13,000 calendar years or to live long enough to witness and record the next pole reversal.

X. Reservoirs of C^{14}

In regard to the world-wide atmospheric inventory of C^{14}, probably the most difficult factor to evaluate quantitatively is that of the balance among the reservoirs. The main reservoirs are the atmosphere and the associated almost contemporaneous biosphere, the mixed (surface) ocean layer, and the deep oceans (the largest reservoirs of all). Many attempts to evaluate the mixing parameters have been made. One of the most recent is that of Houtermans, Suess, and Oeschger (H5) in which box models and electrical analogs are presented. However, more specific data were published by Gulliksen and Nydal (G3). The answers to the fundamental problem of the exchange rates on this enormous global scale cannot be obtained until the magnitude and duration of the changes in atmospheric C^{14} are determined extending back in time to, at least, 11,000 B.P.

XI. Polynomials and Oscillations

So far, the factors which may affect the long-term deviation (Fig. 1) have been discussed. Curves have been

derived by means of various computer programs to fit this trend such as those by Damon (D2) and Suess (S8). Our best fit, based on the 600 C^{14} dates is shown in Fig. 4. It is derived from the following third order polynomial:[3]

$$T_{C-14} = -43.96 + 0.918 \times T_D + 7.17 \times 10^{-5} \times T_D^2 + 1.18 \times 10^{-8} T_D^3 \quad (4)$$

where T_{C-14} and T_D are positive for A.D. and negative for B.C. $T_{C-14} = C^{14}$ dates. T_D = dendro date.

However, as we can see in the simple regression curve of Fig. 1, there are shorter term oscillations which depart from the main trend.

To determine which of these are real, rather than due to statistical uncertainties and errors in measurement, we again drew the time-scale on a straight line, and plotted the C^{14} dates as differences from those calculated from equation 4, and included the one sigma uncertainties of each C^{14} date. This produced a forest of lines that was difficult to interpret. Therefore, for the purpose of simplification, we eliminated all points whose error bars crossed the base-line. We then created a new plot with the remaining dates. Then, for emphasis, we encircled the C^{14} dates

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[3] I am indebted to Douglas P. O'Brien of GeoMetrics, Palo Alto, California, for deriving this equation.

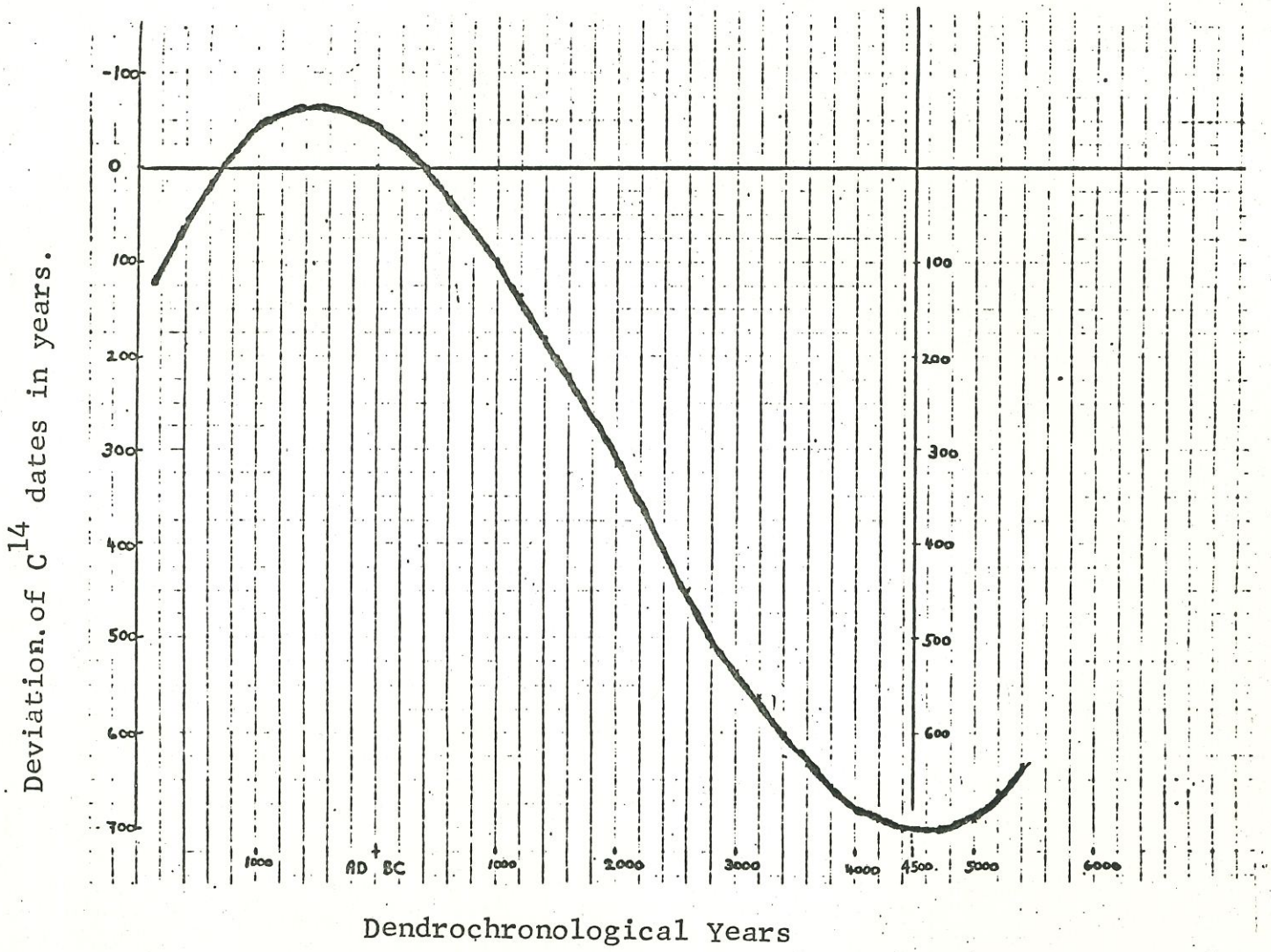

Fig. 4. Average Curve based on Eq.4 which is the best fit for 600 C^{14} dates for dendro-dated samples.

that deviated from the baseline by two sigma or more (see Fig. 5). We next drew an oscillating curve through the remaining deviations. The significant variations have an amplitude of 100 to 200 C^{14} years, and tend to have a time constant of 400 real years, although this is far from regular.

In a computer program devised by Kenneth Meyer in the Department of Geography at Temple University, averaging was done at 20-year intervals. This produced a greater forest of apparent short-term oscillations which tend to mask the possible 400-year cycles. (See Fig. 6.)

We turned again to plotting with 9-sample weighted averages of both dendro- and C^{14} dates. (These averages were calculated at the University of Pennsylvania from the data in Fig. 1.) The results are shown in Fig. 7.

The first question is: which of these plots represents the best display? Since the curves are conveniently compared only on large worksheets, I have compiled Tables I and II. One notes from the left-hand columns that many of the peaks do not occur with the same periodicity from plot to plot. From a simple visual comparison of Figs. 5 and 7, however, there is a correspondence among about ten peaks, roughly two-thirds of the total. Also, in both figures, approximately

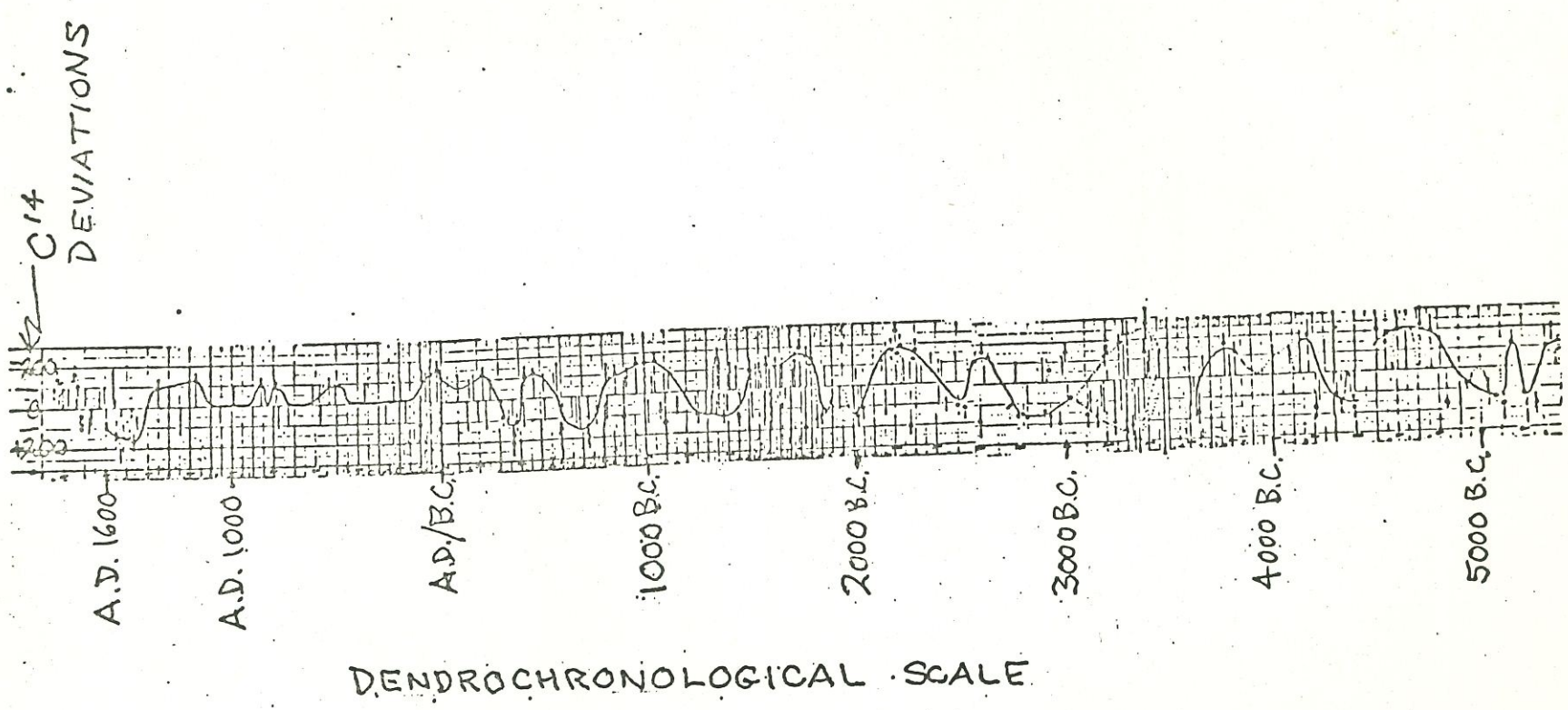

Figure 5. Plot of significant residual deviations from the average curve shown in Figure 4. C^{14} dates with solid circles deviate by 2 sigma; others, by 1 sigma.

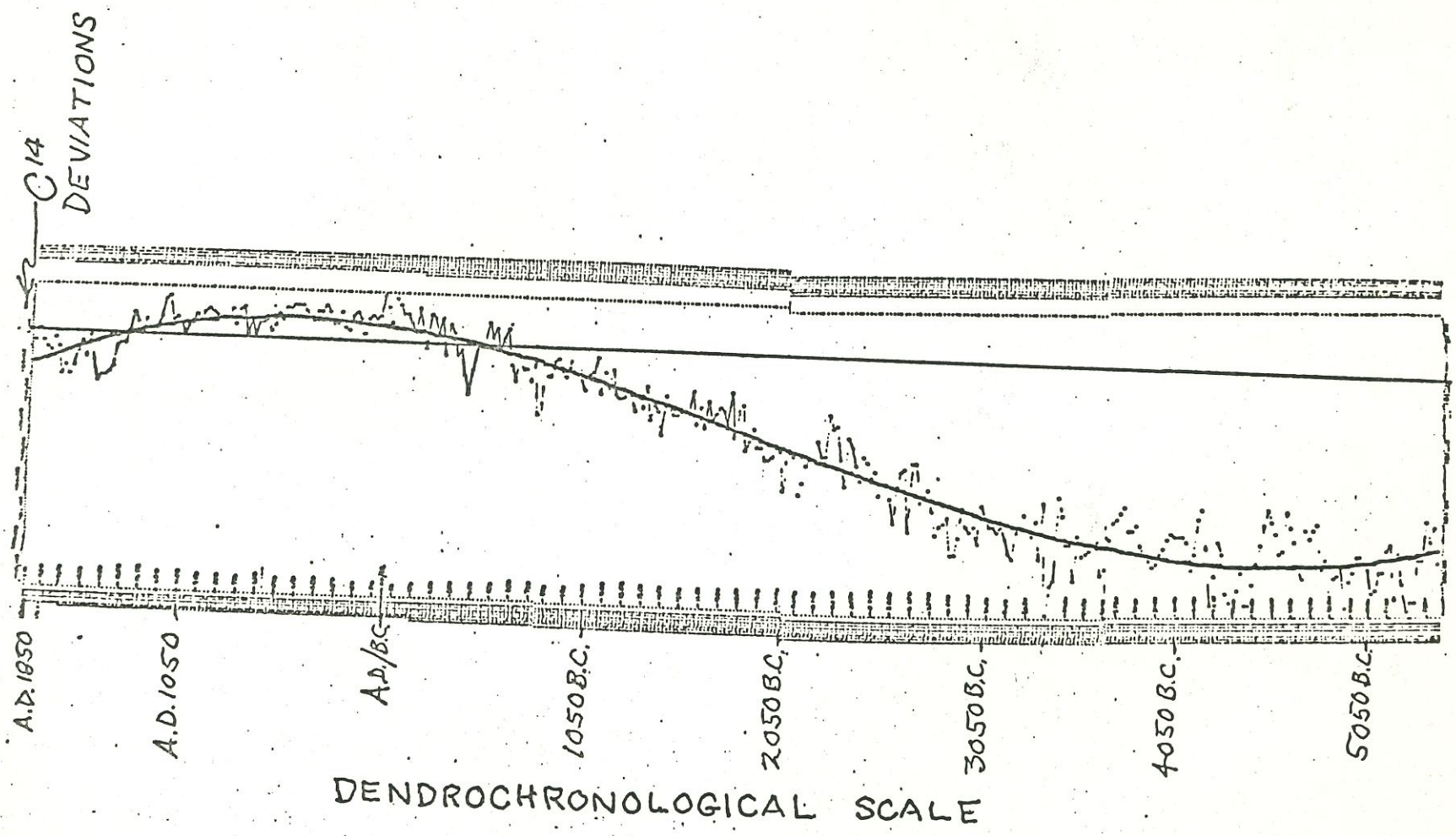

Figure 6. Plot prepared at Temple University of the average fit and residual deviations averaged over 20-year intervals.

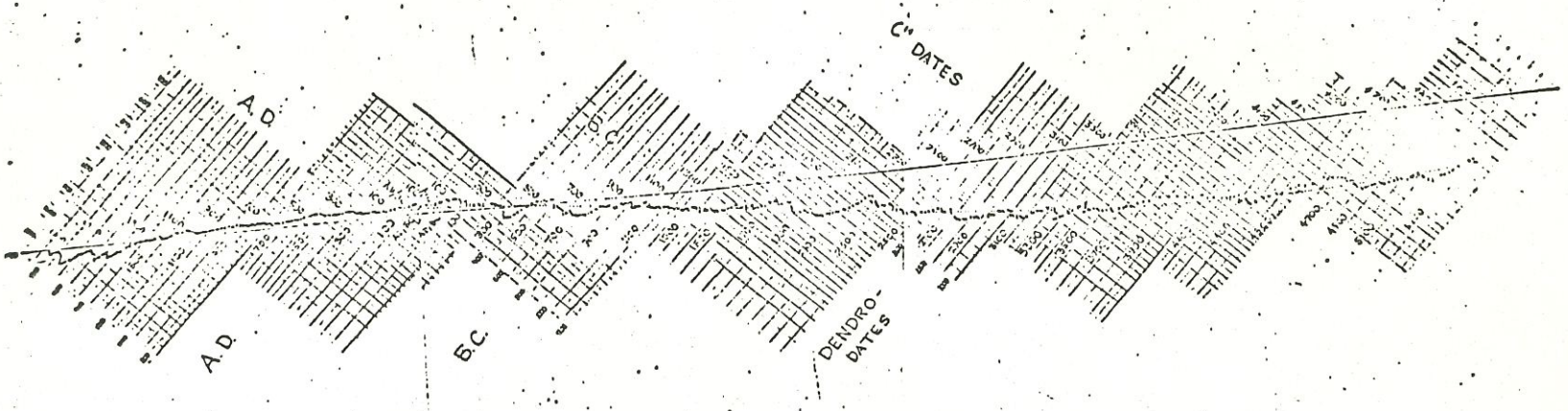

Fig. 7. University of Pennsylvania plot of weighted 9-sample averages along with curve from Eq. 4 in which is the best fit on the average for 600 dates (dashed line), and 45° correspondence line (solid line) are shown.

Table I. Analysis of Oscillations shown in Figure 5.

Calendric Dates of Adjacent Peaks	Interval between Peaks in Calendar Years
A.D. 1500 - A.D. 1200	300
A.D. 1200 - A.D. 20	1180
A.D. 20 - 220 B.C.	200
220 B.C. - 360 B.C.	140
360 B.C. - 470 B.C.	110
470 B.C. - 700 B.C.	230
700 B.C. - 1000 B.C.	300
1000 B.C. - 1400 B.C.	400
1400 B.C. - 1750 B.C.	350
1750 B.C. - 2325 B.C.	375
2325 B.C. - 2625 B.C.	300
2625 B.C. - uncertain	uncertain
3780 B.C. - 4170 B.C.	390
4170 B.C. - 4670 B.C.	500
4670 B.C. - 5070 B.C.	400
5070 B.C. - 5150 B.C.	80
5150 B.C. - 5220 B.C.	70
5220 B.C. - 5350 B.C.	130
	Total = 17

Note: Criterion for the selection of peaks was that each one should have an amplitude of 100 C^{14} years or more.

Table II. Analysis of Oscillations shown in Figure 7.

Calendric Dates of Adjacent Peaks	Interval between Peaks in Calendar Years
A.D. 1700 - A.D. 1500	200
A.D. 1500 - 1 B.C.	1500
1 B.C. - 350 B.C.	350
350 B.C. - 700 B.C.	350
700 B.C. - 1725 B.C.	1025
1725 B.C. - 1800 B.C.	75
1800 B.C. - 2200 B.C.	400
2200 B.C. - 2350 B.C.	150
2350 B.C. - 2800 B.C.	450
2800 B.C. - 3150 B.C.	350
3150 B.C. - 3325 B.C.	175
3325 B.C. - 3425 B.C.	100
3425 B.C. - 3850 B.C.	425
3850 B.C. - 4650 B.C.	800
4650 B.C. - 5240 B.C.	590
	Total = 15

Note: Criterion for the selection of peaks was that each one should have an amplitude of 50 C^{14} years or more.

one-third of the intervals between the peaks occur roughly at 400-year periods.

As a further test for possible periodicity, Douglas O'Brien tested the residuals from his Eq. 4 (page 32) with various statistical procedures including harmonic analysis by means of a Fourier transform. He also averaged the residuals over 50-year intervals. (See Fig. 8.) The results from both tests were inconclusive, but they are in reasonable accord with the results from Fig. 7 for the A.D. era. However, for the B.C. range, where dates are fewer, this analysis does not clarify the situation. As one can see in Fig. 8, there are pronounced oscillations in the B.C. era based only on one 50-year interval each. One may state, however, that this harmonic analysis confirms the fact that there is no true periodicity in the oscillations of the C^{14} dates.

For these reasons and because of the uncertainties in the plot of Fig. 5, I prefer the technique and plot of Fig. 7 as representing the closest approach to the true situation.

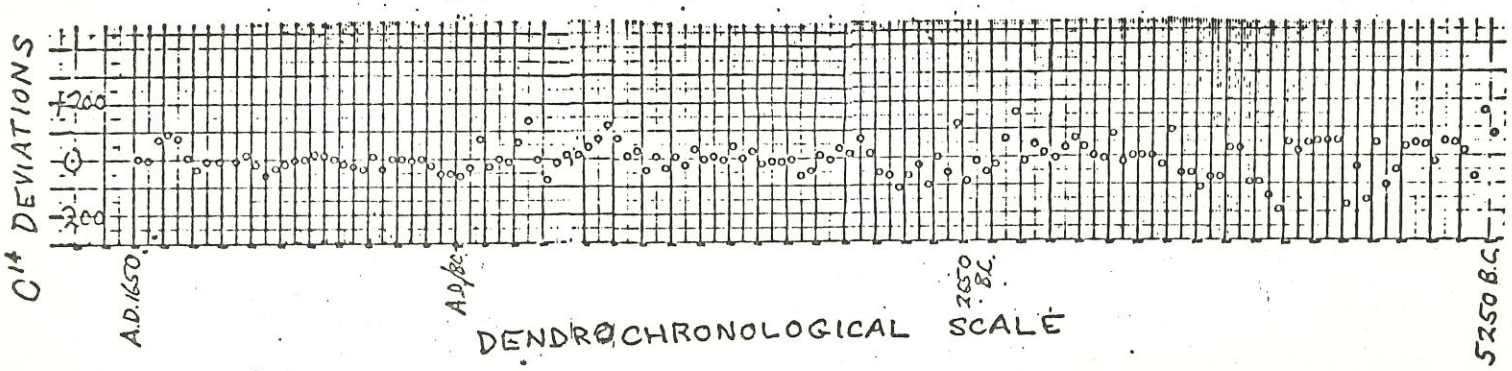

Fig. 8. Plot of the deviations from Eq. 4 (derived by D. P. O'Brien) with spectral analysis, averaged over 50-year intervals.

Conclusions

I demonstrated in Section VIII that part of the long-term deviation in the atmospheric inventory of C^{14} was due to the known average changes in the intensity of the Earth's magnetic field. It is believed that this was caused by a comparatively gradual change in the Earth's dipole moment.

The dipole moment now has a strength of about 8×10^{25} emu, and the axis is inclined about 11^{o} to the Earth's geographic axis. (A recent reference for these facts and some that follow is Stacey's Physics of the Earth (S4). Also the best-fitting theoretical dipole field (to explain the cause of the magnetic field) is centered about 300 km off the Earth's geographic center. Therefore, since there is asymmetry, it is not surprising that the main dipole field has changed fairly frequently. It also has been changing in time.

The main dipole field, however, is not far from the center of the Earth. In addition to this, the nondipole field, at any given time, can be approximated by eight non-central dipoles at about 0.25 Earth radii (about halfway between the core-mantle boundary and the center of the Earth). However, these are idealized dipoles and not extensive volumes so that they may actually be at lesser depth, but still in the liquid core. Secular variation and

westward drift (approximately 0.18^{o} per year) are probably a result of the nondipole field, and due to the fact that the mantle of the Earth may be rotating faster than the core.

The separation of the magnetic field into dipole and nondipole components does not have true significance, but it does provide a means of explanation of the behavior of the field. (In physical reality the two must be summed together.) The strength of the nondipole is about 20 percent of that of the dipole field (C1 and S4). Since the dipole field changes also, it contributes to the secular variation. Recently, the dipole field has been decreasing slowly, but the nondipole field may have been changing at a greater rate. This might account for the two very pronounced peaks in recent times (A.D. 1700 and A.D. 1500, Fig. 7). However, during the past 200 years, the field has been growing stronger, which may explain the decrease that began to appear before we started to deplete the atmospheric inventory of C^{14} by the combustion of fossil fuels during the past 100 years. This is really unfortunate since the past 400 years constitute the only period for which we have accurate magnetic data, that is, direct recordings of the magnetic intensity, declination, and inclination. For all earlier periods, we must

rely upon secondary archaeomagnetic measurements of fired clays, lava flows, etc. with attendant uncertainties in both the measurements and the dating.

However, if, tentatively, we accept the archaeomagnetic results and compare the deviations of curve a of Fig. 2 (and curve b for the 5000 B.C. interval) from the smoothed curve c with Figs. 5 and 7, we arrive at the results shown in Table III. The correlation is only slightly better than 50% The one gratifying observation about Table III is that the two methods of plotting gave the same results. One difficulty is that most of the archaeomagnetic dating is based upon associated C^{14} dates. Some, therefore, may not have been truly associated, and since we do not yet have correction factors (based on the bristlecone pines) for the 5000 B.C. peak, the very slight apparent agreement may be fortuitous.

However, if we accept the magnetic data as being reasonably accurate, we find that the correlation with the long-term trend is good, except for inadequate amplitude, but with the short-term C^{14} oscillations it is very poor, except that there is a tendency for 400-year intervals in the C^{14} data and the measured secular magnetic variation has a similar period. The harmonic analysis by Douglas

Table III. Comparison of Archaeomagnetic (Fig. 2) and C^{14} Results (Figs. 5 and 7).

Approximate Date of Magnetic Change	Direction of Magnetic Change from curve C	Comparison with Fig. 5	Comparison with Fig. 7
A.D. 1000	Up	No correlation	No correlation
A.D. 10	Up	Correlates	Correlates
450 B.C.	Up	Wrong Direction	Wrong Direction
600 to 1000 B.C.	Down	Correlates	Correlates
5000 B.C.	Down	Very slight correlation	Correlates but effect upon C^{14} is small even though the magnetic deviation is large.

O'Brien suggests, however, that there is no true sinusoidal periodicity. This tends to confirm the fact that the magnetic changes are among the basic causes. However, since the correlation with the Earth's magnetic oscillations is not good, we must look for other possible causes. It is my thought that the most probable is the varying magnetic field of the Sun.

Babcock and Babcock (B1), by a series of careful measurements from 1952 to 1954 (during a quiet sunspot period) determined that the intensity of the magnetic field near the poles of the Sun is of the order of 1 gauss on the average, but that there are appreciable fluctuations. In the middle latitudes, that is, the equivalent of equatorial regions, the field tends to be about 0.2 gauss, but again it is variable. At any rate, the average for the whole Sun appears to be at least as great as that for the Earth with an average value of 0.5 oersteds (1 gauss in B units is equivalent to 1 oersted in H units.) Supporting evidence for the Sun's field is the fact that magnetic fields are essential to create sunspots.

A pertinent correlation between the Sun and Earth may be obtained from the earthly diurnal changes which sometimes have magnitudes of 50 gammas (1 gamma = 10^{-5}

oersteds), but are usually of the order of 25 gammas. The diurnal changes must be related to the relative motions (and fields) of the Sun and Earth. Thus the field of the Sun may be responsible for some terrestrial magnetic effects. Again, we are stymied in correlating the effects upon the atmospheric inventory of C^{14} because the varying magnetic field of the Sun has only recently been measured. Climatic effects which would be caused by variations in the behavior of the Sun, may now be considered as secondary causes. However, changes in climate such as the warming of the oceans at the end of the Wisconsin Glacial Period, would affect the atmospheric inventory of C^{14}, i.e., the balance between the atmosphere and the oceans.

Explosions of supernovae, as suggested by Dergachev and Kocharov (D3), as possible causes of the variations in the atmospheric inventory of C^{14}, should also be considered. However, the possible effect would be due to the generation of excess cosmic rays from the time of the explosion of the supernova. If we believe the data from meteorites (Section VIB), then probably the cosmic ray intensity has been constant, at least within 5 percent and within the period of the radiocarbon time scale.

In summary, part of the long-term deviation in the

atmospheric inventory of C^{14} is due to changes in the Earth's dipole moment; the shorter-term oscillations may be related to variations in the Earth's nondipole field, the correlation is unclear except for a tendency for both to have periods of roughly 400 years, although there is no true sinusoidal periodicity; there may be a contribution from the varying magnetic field of the Sun; and there is a slight possibility of a contribution from the explosions of supernovae.