A 'state of the method' report, by its inventor

The Radiocarbon Dating Method

W. F. Libby

Professor Libby received the Nobel Prize in 1960 for his work in developing the radiocarbon dating method. A recipient of numerous other honors, Libby is currently director, Institute of Geophysics and Planetary Physics, University of California, Los Angeles.

This research was sponsored by the National Science Foundation under Grant No. GA-628, Contribution No. 3101 from the Department of Chemistry, UCLA.

Radiocarbon dating is a measurement of the age of dead matter by comparing the radiocarbon content with that in living matter. Radiocarbon (radioactive carbon C^{14}) is produced by the cosmic rays in the atmosphere and is assimilated only by living beings. At death, the assimilation process stops and the immutable radioactive decay loss no longer is compensated by the intake in food. The average life of a radiocarbon atom is 8,300 years so ample time for thorough mixing throughout the earth and through its atmosphere and oceans and the biosphere insures that living matter, wherever found on earth, always has the same radiocarbon concentration, i.e. the same ratio of radioactive carbon to ordinary carbon. This ratio is very small; about one in a million million atoms, but nevertheless, it is adequate for sensitive instruments and can be measured to somewhat better than I% accuracy.

The law of radioactive decay is that a given fraction is always lost in a given time. The half life of radiocarbon, 5,730 years, is the time for 50% loss. Thus, after a tree has fallen, 5,730 years later it will have half of the radiocarbon content of a living tree. For any other ratio larger or smaller, the age is lesser or greater. If the content is one quarter of that living material, the age is twice 5,730 years. This continues until it reaches an unmeasurably low figure at about 50,000 years. Thus, radiocarbon dating applies only to materials that have not been dead longer than 50,000 years. Most of human history falls in this span, however, and radio-carbon dating covers the great reaches of time most important to human history.

The assumption that the concentration of radiocarbon in living matter remains constant over all of time is a bold one. It appears to be nearly correct; however, deviations of a few percent do occur. These are determined by the measurement of radiocarbon in the wood in trees dated by the number of rings found. The Bristlecone Pine is the most famous and most useful tree yet discovered. In this way it has been possible to determine the accuracy of the basic assumption back to some 8,000 years, and a correction curve has been produced which allows absolute dating by radiocarbon back to 8,000 years. The deviation is about 8% at maximum although it is not entirely clear that the 8,000 years' deviation is decreasing from its maximum at about 6,000 years ago.

Radiocarbon dating's main contribution to human history so far has been to show the recency of many events and the antiquity of many other events. Man first came to the Americas apparently only 12,000 years ago, whereas animals have been here for a much longer time. On the other hand, the magnificent colored paintings of the Lascaux Cave in France are 16,000 years old. That is, the Lascaux color paintings were made 4,000 years before the first humans came to the Americas. Of course, we may find evidences of older man in America, but in twenty years, the firm radiocarbon dates for human occupation have never exceeded 12,000 years; whereas, in Europe and Asia Minor, they reach back to the limits of the radiocarbon method and go well beyond, as other dating methods have shown. Radiocarbon dates are a measure of simultaneous events, for the radiocarbon mixes quickly throughout the atmosphere and oceans. Thus, if the cosmic rays do vary by a few percent, so long as they are calibrated at any one place, thecalibration applies world wide.

The dating technique is somewhat demanding, requiring about one week in total time and requiring equipment cost--about $30,000.00.

W. F. Libby

PRINCIPLES

The cosmic ray production of radiocarbon in matter is the basis of radiocarbon dating. It is made from the most abundant atom in air-nitrogen of mass fourteen. Radiocarbon--carbon-14 or C^{14} --lasts 8,300 years on the average before reverting by radioactive decay to nitrogen-14. During this time it enters all living things as well as sea, water and air. Chemically, carbon dioxide is the food of life and presumably the freshly produced C^{14} atom is oxidized sooner or later (probably in a few days, although this time is not at all well known) to $^{14}CO_2$ which is mixed with the ordinary carbon dioxide (0.03% in air) by the winds. The process which converts CO_2 into plants--photosynthesis--is the means whereby the radiocarbon is introduced into living beings. If organisms were to live off of coal or oil, radiocarbon dating would not work for them because they would not be in touch with the cosmic rays through recent photosynthesis. The long time that coal or oil have been underground insures that the original radiocarbon in the plants which produced them would long since have disappeared.

The cosmic rays actually produce radiocarbon only indirectly. In the first step of the process they strike the nuclei of the air atoms and disintegrate them. Among the fragments are many strange, short-lived particles, most of which transform almost immediately into longer-lived entities. Radiocarbon is produced by the interaction of one of these secondary particles, the neutron, with the nitrogen of the air. The neutron has been with us now since the early thirties and has come to be part of our daily life since it is the purveyor of atomic energy. Neutrons at high altitudes are found in the maze of general debris formed in the collisions of great cosmic-ray primaries with the nuclei of nitrogen or oxygen atoms.

The theoretical structure is in a sense simple: the cosmic rays make neutrons which in turn make radiocarbon atoms at a rate of about 2 per square centimetre of area of the Earth per second and have been doing so for tens of millennia. Thus at present there should be an equilibrium inventory in which about 2 radiocarbons revert to nitrogen every second for each square centimetre of area. Therefore, we should find about 2 disintegrations per second for every 8 grams of carbon in living beings, or dissolved in sea water, or in the atmospheric CO_2, for the total carbon in these three categories adds to 8 (7.5 in the oceans, 1/8 in the air, 1/4 in life forms and perhaps 1/8 in humus. Some of these figures are not accurately proven, but since the ocean is the largest and best known [5% error or better] the total is known to about 10%). Thus, we expect to find this concentration of radiocarbon in living matter.

There is a great saving grace--the 8,300 year average life of radiocarbon. In this great span of time, adequate opportunity exists for the atmosphere and oceans to mix, and for the biosphere to cycle many times, i.e, die, decay to CO_2, and be reborn in photosynthesis. This grand system is continually stirred. Living matter is a part of this until death occurs and thus all living things have the same ratio of $C^{14'}$ to natural and this ratio is two disintegrations per second per eight grams of carbon contained.

TABLE I

ACTIVITY OF TERRESTRIAL BIOSPHERE SAMPLES

Source	**Geomagnetic Latitude**	**Absolute Specific Activity** $d\ min^{-1}\ g^{-1}$
White spruce, Yukon (Frederick Johnson)	60E N	14.84 ± 0.30
Norwegian spruce, Sweden (Donald Collier, Chicago Natural History Museum)	55E N	15.37 ± 0.54
Elm wood, Chicago (author)	53E N	14.72 ± 0.54
Fraximus e elsior, Switzerland (Donald Collier)	49E N	15.16 ± 0.30
Honeysuckle leaves, Oak Ridge, Tennessee (C.H. Perry, Clinton Laboratory)	47E N	14.60 ± 0.30
Pine twigs and needles (3650 m. alt.) Mount Wheeler, New Mexico (Robert Fryxell)	44E N	15.82 ± 0.47
North African briar (John Hudson Moore, Inc.)	40E N	14.47 ± 0.44
Oak, Sherafut, Palestine (Donald Collier)	34E N	15.19 ± 0.40
Unidentified wood, Teheran, Iran (M. Hessaby)	28E N	15.57 ± 0.34
Fraximus mandshurica, Japan (Donald Collier)	26E N	14.84 ± 0.30
Unidentified wood, Panama (John Simpson)	20E N	15.94 ± 0.51
Chlorophora excelsa, Liberia (Donald Collier)	11E N	15.08 ± 0.34
Sterculia excelsa, Copacabana, Bolivia (9000 ft. alt.) (Donald Collier)	1E N	15.47 ± 0.50
Ironwood, Majuro, Marshall Islands (Donald Collier)	0E	14.53 ± 0.60
Unidentified wood, Ceylon (Donald Collier)	2E S	15.29 ± 0.67
Beech wood ('Nothafagus') Tierra del Fuego	45E S	15.37 ± 0.49
Eucalyptus, New South Wales, Australia (Donald Collier)	45E S	16.31 ± 0.43
Seal oil from seal meat from Antarctic (Byrd Expedition through H.J. Deason)	65E S	15.69 ± 0.30
Average	---------	15.3 ± 0.1*

* *Error of calibration of counter raises error on absolute assay to 0.5.*

At death, isolation occurs and the radiocarbon clock starts ticking. The isolation is complete so we can separate the ancient matter to be dated from modern contamination. The fact that it is possible to do this "laundry" job so completely is one of the real breaks of good fortune in radiocarbon dating.

The "laundry" of the dating materials is done by common sense and understanding. For example, charcoal is a favorite type of material for C^{14} dating since man is the only animal able to make fire. Charcoal is never attacked chemically. The first move with a charcoal sample is to examine the material under a low-power magnifying glass and to remove with tweezers foreign materials such as plant rootlets. Then an acid wash is used to remove carbonates and this is followed by an alkaline wash to remove humic acids. Normally this is adequate and the treatment ends with a thorough distilled water washing before drying and burning to give the carbon dioxide which is measured for radiocarbon in radiation sensitive instruments. Organic samples collected from widely dispersed places, and different materials such as wood, meat and oil show the same radiocarbon concentration of Table 1. Materials of historically known age or tree ring dated wood are used. The agreement obtained well within an uncertainty of a few centuries (1% in the count is 83 years in the radiocarbon age, since the average life is 8,3 00 years (1)).

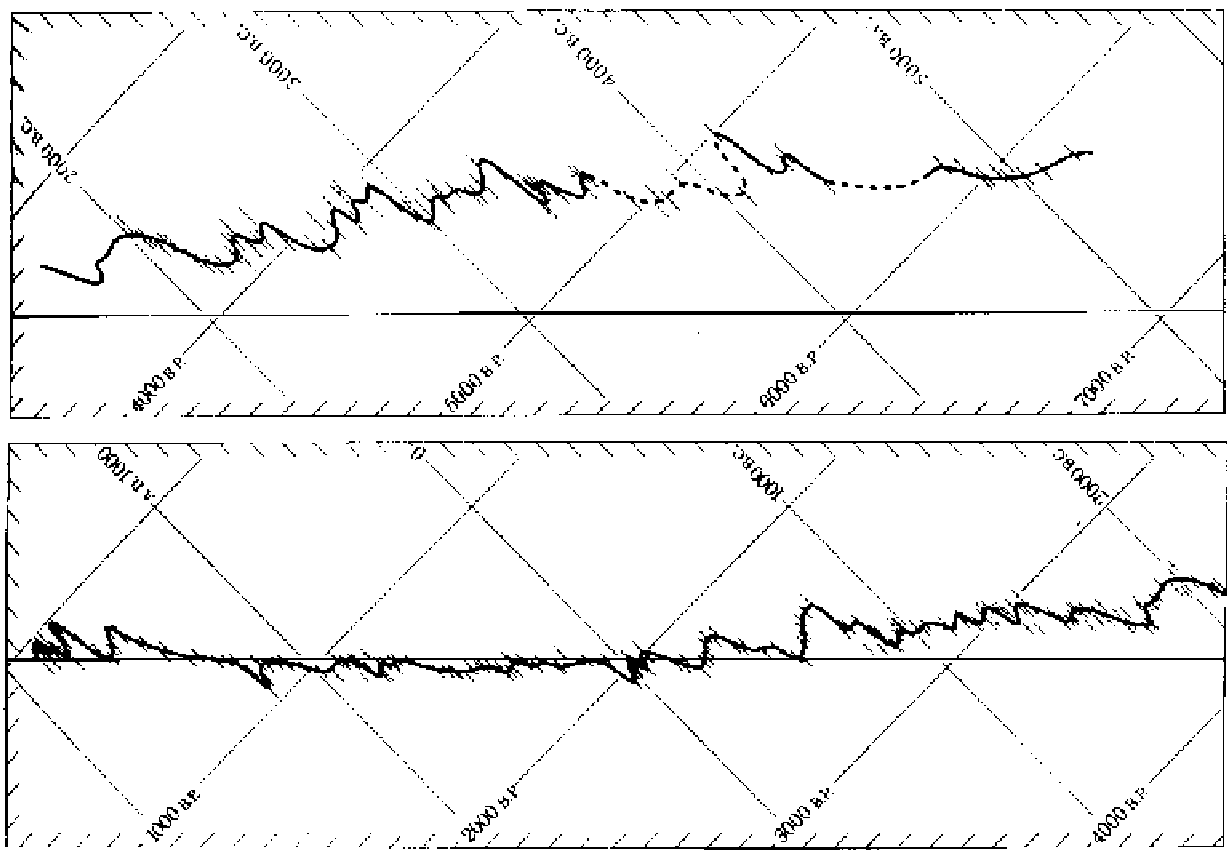


Figure 1. Empirical Correlation between conventional radiocarbon dates using the 5568-year half life and the bristlecone pine tree-ring ages after Suess and Ferguson. In the figures there are two parts: the lower represents the time period between the present and 4000 years ago; the top half of the figure represents the time from about 2000 B.C. to 5000 B.C. The base time for L.P. is the year 1950. The horizontal straight lines represent perfect agreement between the true ages measured by the tree rings and the actual radiocarbon age as shown by the wood in the trees. The radiocarbon dates are given in years before the present and the bristlecone dates in calendar years. Thus, a radiocarbon date of 6000 years before the present would correspond to a tree-ring age of 4900 B.C., or 6850 years before the present, so that the radiocarbon date makes the material appear to be too young by about 850 years. On the other hand, 1000 years ago little error would be found.

DEVIATIONS

The long experience with radiocarbon dating has taught us two things: on simultaneity it apparently is reliable because mixing is relatively rapid, but on absolute dates, it can be incorrect by as much as 600 or 700 years. The peak of the deviation apparently occurs some 7,000 years ago. The simultaneity principle states that two samples taken from any place in the world for any past epoch will give the same date.


Bristlecone pine, Inyo National Forest

The bristlecone pine tree ring chronology of Ferguson & Bannister (2) has made possible the determination of the extent of the deviations of the radiocarbon dates by Suess, Ralph & Damon.(2) back to some 8,000 years ago. The dates appear to start falling slightly too young about 3,000 years ago and continue deviating in that sense until what may be the peak deviation of some 700 years is reached about 5,000 years ago. The deviation then appears to level off. There is some evidence of a decrease in the variation back toward agreement at 10,000 to 11,000 years. This evidence is from the Swedish varve chronology, according to Tauber and others (2). In addition to the broad general sweep of the main deviation, there appears to be a short-term fine structure of somewhat erratic nature, according to Suess (2) (cf. Figure 1).

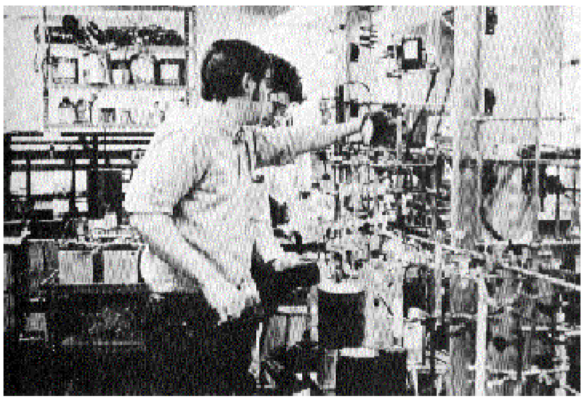
The speculation at the moment is that the main deviation is due to a weakening of the Earth's magnetic field observed by Bucha and others (2) according to the principle pointed out by Elsasser long ago. This deflects fewer cosmic rays and makes more radiocarbon and shows younger ages. The fine structure then is maybe due to variations in the intensity of the solar wind which fends off the cosmic ray. The Earth's field normally deflects about half the cosmic rays so weakening of the magnetic field could cause the observed effect. A third possibility is that solar cosmic rays also play a role. It appears to be unlikely that the intensity of the galactic cosmic rays varies, since the radioactivities found in meteorites seem to agree only with the assumption of constancy, i.e. the long-lived and shorter-lived radioactivities occur in intensities which fit only this assumption. Unfortunately the accuracy with which this assertion can be made is limited due to the paucity of data. A benefit for radiocarbon dating has been gained here from the Moon samples. Their large size and freshness allows more accurate measurement of the intensities of the radioactivities induced by the cosmic rays in the surface rocks. High energy protons from accelerators are used to calibrate for the relative efficiencies of production of the various radioactive atoms.

The question of the solar proton contributions remains somewhat open at the moment. Counters on space probes seem to say that there must be some such contribution but the energy spectrum and the intensity remain uncertain. The Earth's magnetic shield is so strong that it may be difficult for these relatively low-energy cosmic rays to play a role.

The argument for the finely-structured deviations being due to some solar cause is persuasive, but just how the effects occur is less clear and further work is necessary. It may be anticipated that when these points are settled the information may prove to be of value to astrophysics.

The main effort now underway is the Bristlecone Pine Program of Ferguson, Suess and Damon (2), but the work of Stuiver (2) on lake sediments is very promising as well. His results seem to agree with the bristlecone curve in many respects but they do not seem to agree with the Swedish varve results in the older periods beyond 8,000 years ago. Since the method is questionable (the counting of annual layers of sediment and the association of organic matter in a particular layer with that layer), as is true also of the Swedish varve method in some respects, we are left uncertain about the course of the curve prior to 7,500 years ago. So we are driven back to the bristlecone pine method to extend the chronology backward to glacial times about 11,000 years ago.

Ferguson reports finding a piece of wood in the White Mountain (California) area which radiocarbon dates at about 9,000 years. So, presumably, if an overlapping piece or pieces can be found there, the chronology could be extended from the present 8,000 year limit in the White Mountain area back to 9,000. A second area near Ely (Nevada) has a bristlecone chronology reaching back to about 5,000 years. There are several bristlecone stands which have not been dated dendochronologically, but it would seem that they offer some additional hope for future work. Every effort should be made to preserve the ancient wood on the ground in these forests, for they are of prime scientific value, possibly embodying our main opportunity to check radiocarbon dates back to 10,000 years.



The University of California (Los Angeles) carbon-dating laboratory

The principle of simultaneity means that radiocarbon dates are the same at any given epoch over the entire Earth so a calibration at any one locality is equivalent to a world-wide calibration.

GEOPHYSICS AND ASTROPHYSICS

The corrections to the radiocarbon dates are of fundamental interest to the geophysicists and astrophysicists. The source of the Earth's magnetic field remains unknown although evidence accumulates suggesting that it is related to the metallic nature of the Earth's interior and the Earth's rotation. Venus has no magnetic field and is of about the same size as the Earth; it presumably has a metallic interior but does not rotate: 241 Earth days to a Venusian day. Mars also has no magnetic field, is much smaller, and may have no metallic interior even though it rotates at about the same rate as the Earth. The Moon is smaller still. None of these bodies has a magnetic field. Jupiter, on the other hand, has a very strong field--about fifty times stronger than the Earth--and it rotates more rapidly, 10 hours, and is larger than the Earth.

Thus, the evidence seems to indicate that the over-all strength of the Earth's magnetic field decreased substantially perhaps 7,000 or 8,000 years ago, and then subsequently renewed its intensity about 5,000 years ago. It is only the overall strength that matters, since world wide mixing is so efficient. A mere shift of direction (which is well known to have occurred in historic times) would not be recorded by radiocarbon.

On a longer time scale--millions of years--it has been discovered that complete reversal of the direction actually occurs repeatedly. These very ancient data seem to give little evidence about over-all intensity. However, it is natural to suppose that the fact of reversal at least suggests the possibility of intensity variation.

The astrophysicists expect to learn about the constancy of solar activity over the last 40 millennia as more work is done on radiocarbon dating. They also can expect strict limits to be set on the intensities of past super novae bursts since these could have given (-ray bursts which would have given short-termed peaks (of the order of 50 years wide) in the deviation curve of Lingenfelter (2). A short-termed perturbation as we have had recently in the atmospheric nuclear explosions which have raised the C^{14} content of the atmosphere and biosphere by about 50% lasts about 50 years before mixing with the ocean occurs and gives a dilution of some thirty-fold. Thus, radiocarbon is particularly sensitive to short-term perturbations but the method requires samples from the particular years involved. Thus, it has been shown that the Siberian meteorite of 1908 could not have contained antimatter (Cowan, Atluri & Libby, 1965) (3) by measuring wood from tree rings in the years following. Had it contained anti-matter in the amount needed to cause the large explosion observed, it would have produced neutrons (and thus C^{14}) in about the same amount as the tests of 1962 which gave about 50% increase in the biospheric concentration of C^{14}.

HISTORY AND ARCHAEOLOGY

The correction curve itself is primarily of interest to historians and archaeologists. With it we now can say that the Egyptian chronology currently accepted probably is nearly correct. Further work is needed to clarify detailed points and the possibilities of substantial clarification of the history of the first dynasties appear to be good. The opportunities for pre-dynastic Egypt extending back into palaeolithic seem to be very substantial (Save-Soderbergh & Wentdorf) (2).

In Europe the main new result seems to be a redating of the neolithic (Neustupny (2)), at about two millennia older than previously believed, although further work is needed.

In the Americas it has given a quantitative chronology with relatively few surprises, except for the continued failure to firmly establish preglacial man. A great deal of information about the history of the climate has been obtained (cf. for example Wells and Berger) (4).

Earth scientists are interested in the curve itself for dating vertical earth movements and the eustatic rise of the seas following the last glacial period. Climatologists use radiocarbon dates to establish climatic changes on a world-wide basis.

The method itself has been improved in several respects. It now is possible to date bone using the small protein content. The prospects for developing a reliable method for shell samples appear to be brighter (Wentdorf) (2).

The study of the nuclear test radiocarbon and its rate of movement into the sea promises to give detailed understanding of the mechanism and the rate of uptake of CO_2 from the air by the sea; a matter of concern as the rate of burning of coal and oil continues to increase. It will also allow more quantitative evaluations of the fine structure in the deviation curve. A somewhat unexpected result that the rate may be controlled by an enzyme has recently been obtained (Berger and Libby) (5) by studying sub-surface sea water (60 meters deep) and finding that it equilibrates more rapidly with atmospheric CO_2 than does surface water (Santa Monica, California beach) and that the surface water can be brought into agreement by addition of the enzyme carbonic anhydrase in a few parts per million.

A general early treatment of the method is available (6) and the dates themselves are published in *Radiocarbon,* a journal from the Yale University Press.

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