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SECULAR VARIATIONS OF COSMOGENIC ^{14}C ON EARTH: THEIR DISCOVERY AND INTERPRETATION

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ABSTRACT. Measurement of ^{14}C in samples of wood of precisely known age have shown that the cosmogenic ^{14}C content of the CO_2 in the atmosphere has not remained constant during the period of time covered by radiocarbon dating. As the terrestrial atmosphere mixes with a time constant of less than 3 years, these variations must be essentially independent of geographic location. The ^{14}C in atmospheric CO_2 must be a quantity that, at a given time, pertains to the terrestrial atmosphere as a whole. Not only is its knowledge necessary for deriving accurate radiocarbon dates, but it is also valuable in connection with many geophysical and astrophysical problems. Unfortunately, progress in our knowledge of ^{14}C variations in the terrestrial atmosphere has been delayed by hidden experimental errors in results obtained by many laboratories. By rigorous statistical analysis of the La Jolla results, it is now possible to show that the ^{14}C variations are not simple random fluctuations but show distinct regularities. Similar patterns of variations have been found in the growth rate of trees during the last 5000 years. Measurements of radiogenic ^{10}Be currently being done by European workers promise to conclusively elucidate the ^{14}C findings.

THE DE VRIES VARIATIONS

Willard F Libby, at the time when he and his co-workers published the first results of radiocarbon measurements around 1950 (Libby, 1952), believed that the exchange of the carbon dioxide in the atmosphere with the bicarbonate of the oceans occurred quickly compared to all other changes in the ocean-atmosphere system. He, therefore, assumed that ocean and atmosphere can be considered as one and the same large carbon reservoir, reacting, as Libby put it, to changes in the ^{14}C production rate "only extremely sluggishly." Libby considered variations such as those observed by H de Vries (1958) in the ^{14}C content of wood from the 15th and 17th centuries AD to be within the experimental error of the measurements. However, I had been able to show (Suess, 1953) that the exchange rate of carbon dioxide between atmosphere and ocean was not sufficiently fast to exclude the possibility of variations of a few per cent, as indicated by the measurements of de Vries.

In order to explain the results obtained by de Vries, Libby had also suggested the consideration of possible errors in the dendrochronologic dating that de Vries (pers commun, 1954) had used for his measurements. However, several other investigators used different tree-ring sequences to determine their sample ages. They at least qualitatively confirmed de Vries' results (Broecker, Olson & Bird, 1959; Willis, Tauber & Münnich, 1960; Ralph, 1959). As the cause of the ^{14}C variations, these investigators considered changes in the ^{14}C production rate through modulation of the cosmic ray flux by solar activity (see also Suess, 1958, 1968). This matter has been discussed from a more quantitative viewpoint by Stuiver (1961) and by Stuiver and Quay (1980). In any case, the time dependence of ^{14}C content of samples of known age seemed to be sufficiently firmly established for the period from $\simeq$AD 1000 to the present that a reliable calibration curve for a limited period of time was available to be published (Stuiver & Suess, 1966).

Unfortunately, Libby's view discouraged many investigators in the United States from following up on this type of measurement. It also created serious funding problems. Only three laboratories in the United States remained engaged in systematic investigations of the ^{14}C content in wood dated by its tree rings. C W Ferguson of the University of Arizona supplied tree-ring-dated samples to Arizona, Pennsylvania, and La Jolla laboratories. The samples were derived from bristlecone pines, trees that live more than 1000 years.

AGE CALIBRATION FOR BC TIMES

A year later, in 1959 (Suess, 1961), the laboratory in La Jolla observed another major irregularity. It clearly showed that a general ^{14}C trend existed during the 1st and 2nd millennia BC indicating ^{14}C levels increasing with sample age in BC time (see Fig 1). This irregularity caused the conven-

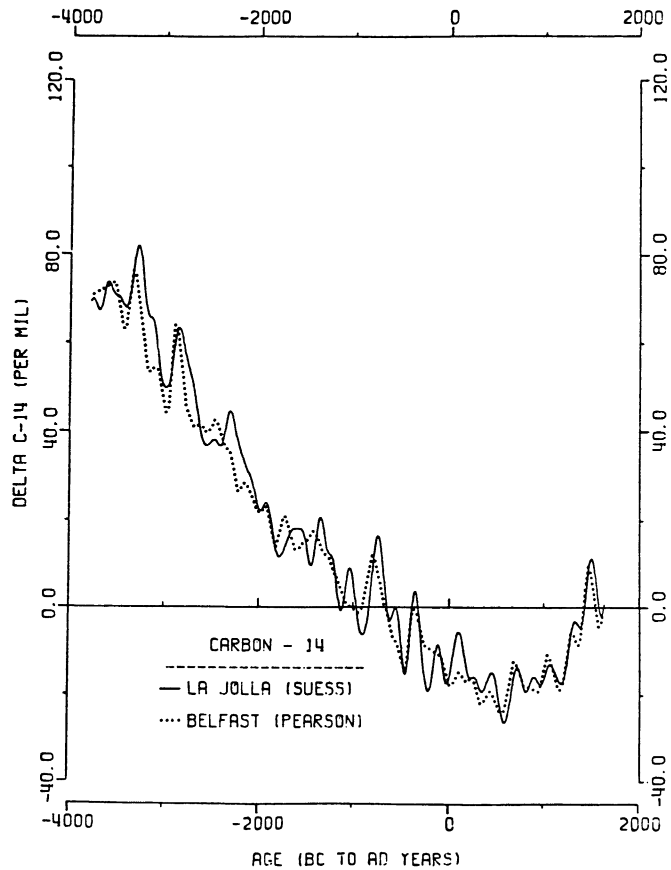

Fig 1. La Jolla and Belfast ^{14}C sequences truncated to common length (3980 BC–AD 1880) and filtered appropriately. Courtesy of Charles Sonett (1985b).

tional ^{14}C dates to appear too young. Samples from BC times were older than these dates had indicated. This had a profound effect upon the Bronze age chronology and on the relationship of cultural developments in the Near East and in Europe. According to Colin Renfrew (1973) it led to the "second revolution in archaeology."[1]

The observed decrease of the atmospheric ^{14}C level during the last millennia BC meant that all the radiocarbon dates so far determined, the so-called "conventional radiocarbon dates," had to be revised. Obviously, the correct age of a wood sample is the same as that of wood of known age that shows the same ^{14}C content.

In 1969, at the Twelfth Nobel Symposium on "Radiocarbon Variations and Absolute Chronology," a calibration curve was presented based on measurements carried out in La Jolla (Suess, 1970). This curve reached back to 5400 BC, as far back in time as bristlecone pine samples dated by their tree rings were available from Ferguson. The curve could be approximated by a sine wave with a peak-to-peak amplitude of ca 10% and a period of ca 10,000 years. This large change in the specific ^{14}C activity was, in general, attributed to a change in the magnetic dipole moment of the earth. Recently, however, it appeared that the calculated agreement between geomagnetic moment and ^{14}C production rate was not as perfect as it had seemed. The high ^{14}C values of 10,000 years ago could possibly be an after-effect of the great ice age (Suess, 1968). Measurements of ^{10}Be certainly will allow this to be decided.

THE WIGGLES

The general trend of this curve was in agreement with measurements by other laboratories, but the curve showed variations, so-called "wiggles," that could not be recognized from measurements by other workers. At that time, the general concensus was that the observed trend was well established; however, the "wiggles" were considered questionable. An exhaustive article by Damon, Lerman, and Long (1978) illustrates the situation at that time.

As expected, Willard Libby was reluctant to accept our experimental results. Privately, however, he remarked, "If your data were to reflect true atmospheric ^{14}C variations, then they would, indeed, be an extremely interesting and important geophysical parameter." Thus, this strengthened my efforts to speedily obtain as much information as possible on this geophysical parameter. I assigned the highest priority to measurements of suitable bristlecone wood samples that I obtained from Ferguson.

In the early 1970s, the question of the nature of this geophysical parameter was still open. Even the very existence of variations in the ^{14}C level with amplitudes on the order of 1 or 2% appeared to be controversial at that time. In particular, R M Clark of Monash University stated that the

[1] The so-called "first revolution in archaeology" was caused by Libby's Chicago measurements such as the dating of the famous "Two Creeks Forest wood" from Wisconsin. They showed that large parts of North America (and also of Scandinavia) had been covered by ice until ca 10,000 yr ago, *viz*, much longer than had been thought before. Hence, the end of the Paleolithic era had occurred much later than had in general been assumed.

assumption of "wiggles" in the calibration curve was "not statistically justified" (Clark, 1975).

CYCLES

I, personally, did not have the slightest doubt that these variations had indeed occurred. For me, the next open question was whether they were random fluctuations or periodic, in some way regular. To me the variations seemed to show cyclic recurrences of distinct features, as indicated by their time spectrum, which showed well-defined lines superimposed upon a noisy background (Neftel, Oeschger & Suess, 1981). However, these results were ignored. Funding during the 1970s had become increasingly difficult. In 1980, the National Science Foundation terminated its support completely. A request for money for data reduction and for the publication of the results was denied.

At that time, and with the loss of all funding for further measurements, the data so far obtained in La Jolla, somewhat revised and including some unpublished results, were compiled by Linick and sent on computer cards to seven prominent persons working on time series analysis, with the request that they impartially analyze the series. The only investigator who complied with this request was Charles Sonett of the University of Arizona, who was interested in cosmic ray variations and solar activity. His very careful and sophisticated study of the La Jolla ^{14}C time series fully confirmed the more elementary approach of my previous co-workers (Sonett, 1985).

There can be no doubt now that the short-term ^{14}C variations as observed for Holocene times, such as shown in Figure 1, are primarily caused by the modulation of the cosmic ray flux through interplanetary magnetic fields originating from the sun. The most obvious observation that shows this is the correlation of the atmospheric ^{14}C level with the sunspot index during the so-called Little Ice age in the 15th and 17th centuries AD. That this correlation is not just qualitative has been shown by Stuiver and Quay (1980).

The 200-year sunspot cycle seems to have prevailed at least during the past 2000 years (Schove, 1983). Also, the frequency of observations of *Aurorae Borealis* (Link, 1964) seems to have varied periodically in 200-year intervals. The ^{14}C variations, with time constants on the order of 100 years, appear to be almost as regular as the 11-year sunspot cycle. Thus, other effects, such as changes in the rate of ocean-atmosphere CO_2 exchange, can hardly be major contributing factors to these short-term ^{14}C variations in the atmosphere. Measurements of ^{10}Be concentrations now in progress in Europe (Beer *et al,* in press; Raisbeck, ms in preparation) can be expected to settle this matter conclusively.

^{14}C AND CLIMATE

For many years I had had the impression that a correlation existed between the ^{14}C level and the global climate. This was particularly true for the time of the Little Ice age in the 15th and 17th centuries. During this time of a world-wide low temperature, the ^{14}C level was rising as a result of a quiet sun during the so-called Spörrer and Maunder sunspot minima.

By good fortune, Sonett learned that at the University of Arizona tree-ring laboratory another complete tree-ring series had been worked out independently from that of Ferguson which had been used for ^{14}C determinations. This series was established by LaMarche and Stockton (LaMarche, 1974) specifically in order to investigate the effects of climatic variations upon the growth rate of trees as shown by the thickness of the annual tree rings (see Fritts, 1976). Sonett used growth rate values that had been normalized by experts in such a way as to eliminate effects from factors other than climate. He could show that time variations in the thickness of the tree rings revealed a spectrum similar to variations of ^{14}C in contemporaneous wood (Sonett & Suess, 1984). Thus, it appeared that variations in cosmic ray-produced ^{14}C in the atmosphere and variations in climate that affect the growth rate of trees are mediated by the same factor, in all probability, the sun. In what way this happens is as yet not clear. It seems that the phase relationships in the two time series are complicated. Still, I believe that this relationship between the growth rate of trees and cosmic-ray-produced ^{14}C shows more convincingly than any other empirical correlation that solar activity influences the global terrestrial climate.

CALIBRATION

Thirty years have now passed since Willard Libby obtained his first results of radiocarbon dating. Since then it has become obvious that these results, though most valuable in some fields of science, were not accurate and needed corrections. These corrections are a function of the time at which the samples originated; they are now known as far back in time as wood samples dated by their tree-rings have become available (Suess, 1979). They reflect variations of the specific activities of ^{14}C in the carbon dioxide of the atmosphere at the time of growth of the wood. As discussed above, this activity has not fluctuated randomly, but to a certain extent has undergone periodic variations. Hence, a spectrum of these variations shows distinct lines superimposed upon a noisy background which is largely caused by experimental error. This makes is possible to distinguish between accurately measured and less accurately measured ^{14}C time series, because errors in the ^{14}C determinations as well as mistakes in the dendrochronologically determined tree-ring ages obscure the spectral lines and enhance the background.

The most complete and longest tree-ring series so far established is that of Ferguson, for wood from the bristlecone pine of the White Mountains, California. The La Jolla laboratory has, over the years, measured some 700 samples obtained from Ferguson, many of them twice. Never have indications been observed of an error in this tree-ring sequence. The La Jolla ^{14}C data for this sequence were used by Sonett for investigations of the properties of the ^{14}C time series. Recently, Pearson (1980) of the Belfast ^{14}C laboratory published ^{14}C values for a tree-ring series established by Brown *et al* (1986) for wood grown in Northern Ireland. A comparison by Sonett (1985a,b) of the Belfast results with the bristlecone data showed perfect agreement (see Fig 1). A large number of ^{14}C measurements have also been carried out in La Jolla on wood from floating tree-ring series of oak trees from central Europe as established by Becker (1980). There, however, dendrochronologic difficulties appeared to exist that only quite recently were eliminated. This was done by "wigglematching" the ^{14}C values to those of the bristlecone data (Linick, Suess & Becker, 1985). Wood from this Becker chronology is also being used now in Seattle by Stuiver. Unfortunately, the results of the large number of measurements already performed there have not yet been published in a form that can conveniently be used for statistical analyses. It is to be expected that, in view of the meticulous care and patience with which the Seattle measurements are now being made, they are undoubtedly more accurate than the La Jolla data that have been obtained during the past 20 years. In any case, I hope very much that the considerable work that still remains to be done using conventional high-precision counting techniques will be done without rivalry but with mutual collaboration of the few laboratories still left which are capable of doing this important work.

ACKNOWLEDGMENTS

After the retirement of G Bien in the early 1970s, Timothy Linick supervised the technical operations of the La Jolla Radiocarbon Laboratory and took care of reduction and evaluation of data. Technical operations at the laboratory were carried out by Carol Hutto, and at times also by Terry Jackson. Their work was excellently done, with unusual care and efficiency in every respect. Linick, currently working at the Radiocarbon Laboratory of the University of Arizona, Tucson, is now, with the courteous consent of his present employers, compiling a list of the La Jolla bristlecone pine results (which until now have only been privately distributed to interested colleagues due to lack of funds).

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