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MEDIUM-TERM ATMOSPHERIC ^{14}C VARIATIONS

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ABSTRACT. High-precision ^{14}C measurements are presented, carried out on single tree rings from a section of the floating South German Neolithic tree-ring chronology. They confirm the existence of pronounced medium-term variations in the order of 2 percent during the 33rd to 38th centuries BC.

These variations turn out to be very regular while the precision of 1.5‰ allows a comparison with a geochemical model calculation. Good agreement is acquired for an input function with a periodicity of about 150 years and an amplitude of approximately 30 percent in the ^{14}C production rate.

INTRODUCTION

One of the few primary principles of radiocarbon dating (Libby, Anderson, and Arnold, 1949) is the constancy of atmospheric ^{14}C content. Based on this assumption, radiocarbon ages of material of known age appeared to deviate from true ages. By measuring present-day ^{14}C activity of tree rings, de Vries (1958) showed, that the constancy of atmospheric ^{14}C concentration is not entirely valid for, at least, the past few hundred years. After the extension of the North American bristlecone pine tree-ring chronology (Ferguson, 1970), several teams demonstrated convincingly a sinusoïdal change in ^{14}C concentration over the past 8000 years to a maximum of about 10 percent around 4000 BC (Suess, 1967; 1978; Ralph and Michael, 1970; Damon, Long, and Wallick, 1972). This long-term variation can very well be explained by observed sinusoïdal changes in the earth's magnetic field, affecting the penetration of cosmic radiation into the atmosphere (Bucha, 1970).

Superimposed on this general trend, Suess drew with "cosmic schwung", *ie*, with a fair degree of uncertainty in the statistical significance, medium-term variations, through his data (Suess, 1970). These 'wiggles' sometimes amount to a change in ^{14}C ages of a few hundred years within an historical period of one century. While discussing the validity of proposed wiggles and applied corrections to convert radiocarbon ages to calendar dates, it was, more or less, left to statisticians to provide a smoothed calibration curve (Renfrew and Clark, 1974).

Although a smoothed calibration curve can be applied for "long-lived" sampes with acceptable accuracy, it was believed unsatisfactory and misleading to use statistical methods to obtain a generally accepted calibration curve, without quantitative knowledge of medium-term variations (Mook, 1974). We thought it essential to measure present-day ^{14}C activity in single tree-ring samples and with much better precision than the usual 4 to 6‰. Moreover, only precise knowledge of the detailed nature of short- and medium-term variations allows comparison with geochemical model calculations.

EXPERIMENTAL METHODS

For more than 8000 years the deposition of oak logs along the river valleys in Southern Germany occurred more or less continuously. These subfossil trees are presently being excavated in fluvial gravel deposits for

tree ring analyses (Becker, 1979). This oak material, which is available in quantities up to 100g of dry material per single tree ring, also presents a magnificent opportunity for measuring natural ^{14}C variations with an accuracy of 1 to 2‰.

Our measurements were carried out on a section of the floating South German Neolithic tree-ring chronology. The subfossil oaks we used were sampled from sites within drainage basins of the Danube River (ca 48° 25′ N, 10° 5′ E). Single tree-ring samples, about 100g each were separated by hand from the bulk sections and chipped to matchstick size.

The subsequent chemical pretreatment was performed in three steps: 1) with 4 percent hydrochloric acid (80°C, 24hr) to remove resinous material, 2) 4 percent sodium hydroxide (80°C, 24hr) in order to resolve tannic acids and again, 3) 4 percent hydrochloric acid (80°C, several hours) to remove atmospheric carbon-dioxide, absorbed in Step 2. After each step, the samples were thoroughly washed to $p_H = 7$ with demineralized water. Although the resulting wood fraction probably does not originate from exactly the same time of the year as the α-cellulose fraction, it appeared acceptable for monitoring past ^{14}C levels (Tans, de Jong, and Mook, 1978). The individual year samples were combusted and purified to amounts of 50 to 55 l STP of carbon dioxide. A small amount of carbon dioxide was removed carefully from the mixed bulk for mass spectrometric analysis. The samples were kept under high pressure in small stainless steel containers. The special high-precision proportional gas-counting system has been described elsewhere (Tans and Mook, 1979).

RESULTS AND DISCUSSION

Results, calculated as conventional radiocarbon ages ($t_{1/2} = 5568$ yr), are plotted in figure 1. Measurements are related to the tree-ring number from the floating South German Neolithic master chronology (upper scale in fig 1). Recent comparison with the Bristlecone dates of Suess gave the absolute timescale for this floating chronology (lower scale in fig 1), which appears to start at 4035 ± 3 BC (de Jong, Mook, and Becker, 1979). Using this timescale, we applied corrections for ^{14}C decay ($t_{1/2} = 5730$ yr) since the time of formation of the wood. In order to obtain the actual medium-term changes in the atmospheric ^{14}C level, we had to separate them from the long-term ^{14}C change. For a first approximation, the long-term atmospheric ^{14}C level in the period, 3200 BC to 3900 BC, can be considered constant, but shifted with regard to 19th century natural ^{14}C level. The $\Delta^{14}C$ values, therefore, were all corrected by a constant factor. The resulting $\Delta^{14}C$ values (fig 2) represent the changes in atmospheric ^{14}C concentration, superimposed on, approximately, the constant atmospheric ^{14}C level during the Neolithic.

A first conclusion to be drawn from this figure is the quantitative confirmation of the existence of three pronounced wiggles during the 33rd to 38th centuries BC. Using a theoretical model (Tans, 1978) we calculated the relative changes in the ^{14}C production rate, premising that these are responsible for the observed medium-term atmospheric ^{14}C changes. This model theorizes on the exchange of ^{14}C between the ocean

and atmosphere, based on first-order kinetics and considering the chemical and isotopic equilibria. The size of the mixed layer is fixed, with a typical depth of 100m, which gives an N_m/N_a ratio of approximately 1.5. The observed apparent ages of the mixed layer of about 400 yr (Broecker and Li, 1970) are seen to restrict the atmosphere-ocean transfer rate k_1 fairly close to 0.15 yr^{-1}. The reverse transfer rate k_2 is coupled by mass conservation to k_1 by $k_1 \times N_a = k_2 \times N_m$. The exchange rate from the mixed layer to intermediate depths, k_3, has been taken as 0.125 yr^{-1}. The deep ocean to mixed layer transfer rate k_4 has been neglected, due to the observed fact that the average radiocarbon age of the deep sea is 1600 yr ($k_4 \cong 0.0007$ yr^{-1}). The resulting changes in ^{14}C production rate are calculated as changes above an assumed steady state production of 7.5‰ yr^{-1} and plotted in figure 3. This figure shows that observed variations in atmospheric ^{14}C might be explained by variations in production rate to a maximum of approximately 30 percent above the steady state. Without a quantitative knowledge of steady state ^{14}C production during the period, 3000 to 4000 BC, the amplitude of these ^{14}C production variations will remain under discussion. The calculated amplitude is necessarily smaller if a higher steady state production existed at that time during the smaller earth magnetic field (Bucha, 1970; Lingenfelter and Ramaty, 1970). This

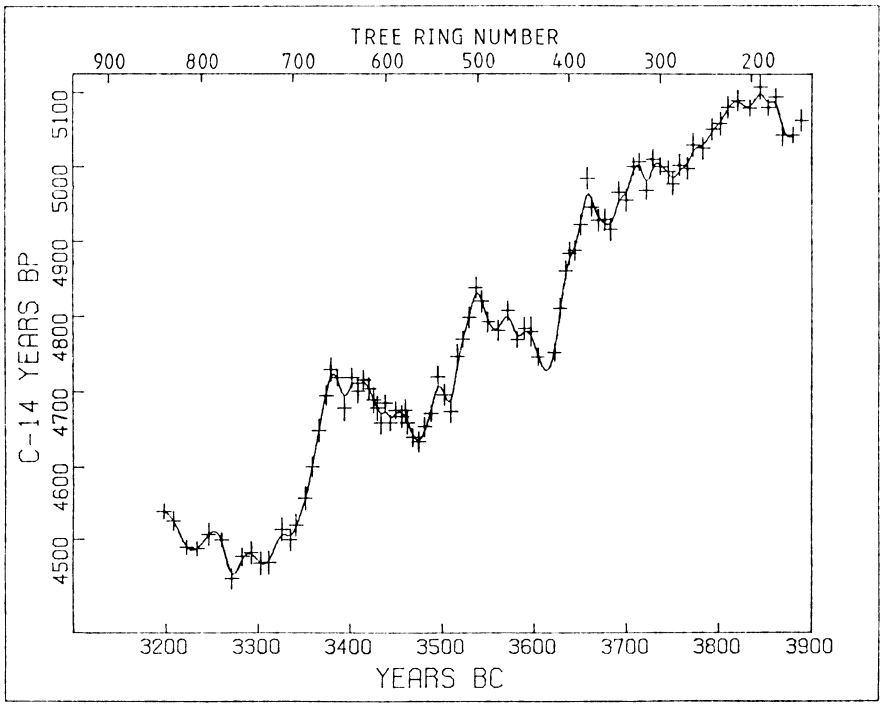

Fig 1. Conventional radiocarbon age ($t_{1/2}$ = 5568 yr) versus tree-ring number for the South German Neolithic masterchronology (upper scale) and versus the recently established absolute timescale (lower scale). Note that there is a slight difference with the previously published dates (de Jong, Mook, and Becker, 1979).

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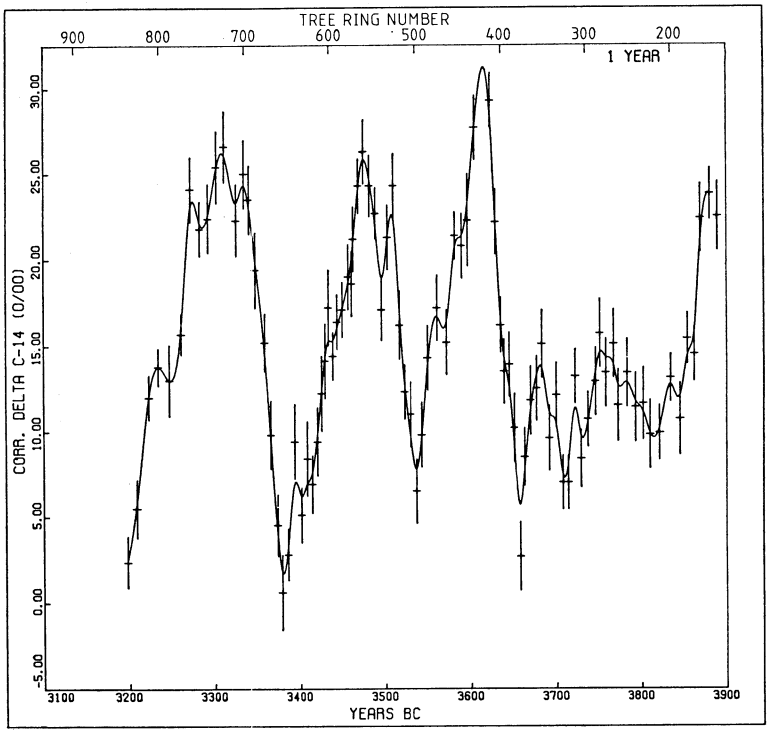

Fig 2. Deviations of $\Delta^{14}C$ from the approximately constant atmospheric ^{14}C level ($t_{1/2} = 5730$ yr) during the Neolithic.

would not affect periodic behavior of the variations. ^{14}C production rate rises to its maximum value in a relatively short period of approximately 50 years, while the decrease takes approximately 100 years. This results in an oscillating behavior with an average period of about 150 years.

During the period, AD 200 to 700, which was previously thought to be "quiet", similar variations are observed (Bruns, Münnich, and Becker, 1980). The comparable amplitude and frequency indicate that medium-term variations not only are more common than originally believed, but are also likely to be caused by a more regular mechanism.

CONCLUSION

Now that we have, unambiguously and quantitatively, established the existence of medium-term variations, the possible phenomena that might cause such changes in the ^{14}C production rate have to be investigated. In this respect, we must consider variations in the terrestrial and interplanetary magnetic field. As mentioned above, the long-term trend in radiocarbon content of the atmosphere over the past 8000 years has been dominated by the geomagnetic effect (Bucha, 1970). Fluctuations on the smaller time-scale of a few hundred years, on the other hand, seems to be caused neither by the main geomagnetic dipole nor by oscillations of the non-dipole earth magnetic field (Stuiver, 1978). It is more likely to

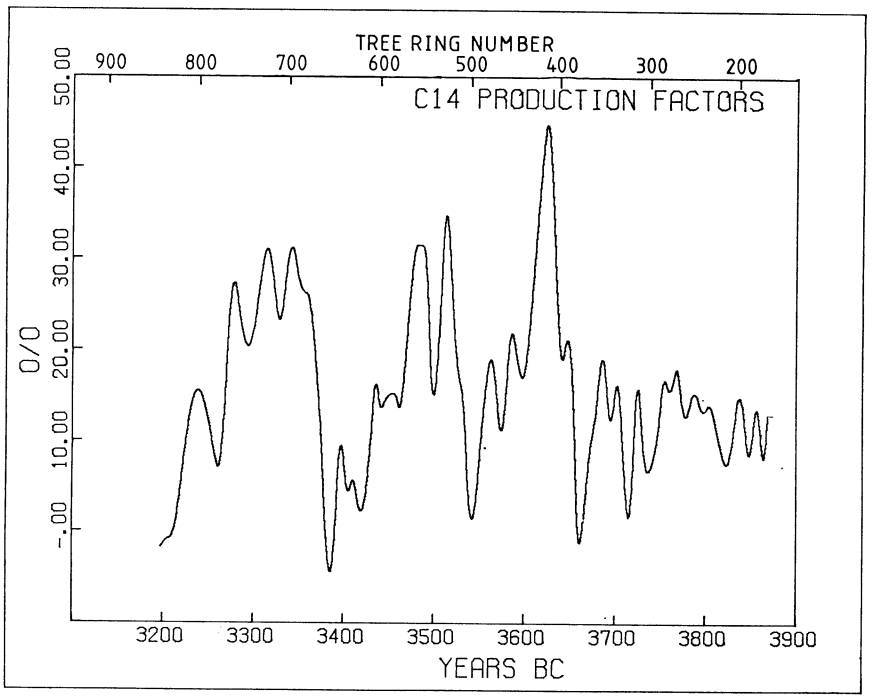

Fig 3. Deviations in the ^{14}C production rate above an assumed steady state production of 7.5‰ yr^{-1}.

assign medium-term fluctuations to changes in solar activity leading to modulation of the galactic cosmic-ray flux penetrating the atmosphere. More high-precision ^{14}C measurements, particularly over the last centuries, are needed for more pertinent information about the correlation between changes in solar activity and atmospheric ^{14}C. Unraveling this relation is also essential in establishing the dilution effect of fossil fuel CO_2 during the last decades. Because of natural ^{14}C fluctuations, the Suess effect cannot yet be established to a sufficient accuracy (Tans, de Jong, and Mook, 1979).

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