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RADIOCARBON ACTIVITY VARIATION IN DATED TREE RINGS GROWN IN MACKENZIE DELTA

C Y FAN*, CHEN TIE-MEI**, YUN SI-XUN**, AND DAI KAI-MEI†

ABSTRACT. Forty-five tree rings (1881-1925) were taken from a white spruce grown near Campbell River in Mackenzie Delta, Canada, for the measurement of ^{14}C activity variation. Because of the narrowness of the rings, 2 and sometimes 3 rings were combined to yield a total of 21 specimens. The ^{14}C content in these specimens was measured with a liquid scintillation-PM tube counter system of the History Department of Peking University. The data points exhibit a 10‰ variation, anti-correlated with sunspot numbers. The physical implication is discussed.

INTRODUCTION

Since Forbush (1954) discovered solar modulation of cosmic ray intensity, it has been assumed that there is a corresponding variation in the ^{14}C content in the earth's stratosphere. Lingenfelter and Ramaty (1970) calculated the production rates for 1953-1954 (solar activity minimum) and 1957-1958 (solar activity maximum) at various geomagnetic latitides; the difference between the average rates in these two periods was ca 22%. The question frequently asked was whether such a variation could cause a fluctuation in the ^{14}C concentration in the troposphere large enough to be measurable. Baxter and Walton (1971) reported an $\sim$ 30‰ $\Delta^{14}C$ variation from 1890-1950, and later, Baxter and Farmer (1973) reported a 10 to 20‰ variation from 1829-1865, both with an 11-year periodicity, anti-correlated with sunspot numbers. But the $\Delta^{14}C$ values in tree-ring samples from the same period measured by others (Broecker, Olson, and Bird, 1959; Cowan, Atluri, and Libby, 1965; Suess, 1965; Lerman, Mook, and Vogel, 1967; Damon, Long, and Wallick, 1973a; Tans, DeJong, and Mook, 1979; Burchuladze et al., 1980; Stuiver and Quay, 1980, 1981) do not show any large amplitude fluctuations, although some of them also exhibit 11-year periodicity. Since Baxter and Walton used grains grown in Scotland at latitudes $\sim$ 54°N, higher than that of most other sample locations, the disagreement may be due to

*Department of Physics, University of Arizona, Tucson
**Radiocarbon Laboratory, Archaeological Section, History Department, Peking University, Beijing, China
†Department of Physics, Nanjing University, Nanjing, China

the difference in latitudes as well as in the time and duration of photosynthetic assimilation of CO_2 by plants (Baxter and Farmer, 1973). Lerman, Mook, and Vogel (1970), in their thorough study of the variation of the ^{14}C content in the 19th- and 20th-century tree rings with geographic locations, found that, at 42°S latitude, contemporaneous tree rings contain 4.5 ± 1‰ less ^{14}C than those at 42°N latitude. Doubt persists as to whether the large amplitude reported by Baxter and Walton (1971) was partly real and partly due to a biased treatment of the measurements and selection of data (Damon, Long, and Wallick, 1973b). We describe here the results of our measurements of $\Delta^{14}C$ variation in 1881-1925 rings of a white spruce grown near Campbell River in Mackenzie Delta, Canada (68°N, 130°W). The data clearly exhibit a fluctuation with a periodicity of 11 years, anti-correlated with sunspot numbers. A plausible explanation is suggested.

SELECTION OF TREE RING SAMPLES

To study the possible latitudinal effect, we looked for trees from the utmost northerly region. We obtained two sections of dated white spruce from Dr ML Parker, Canadian Forestry Service, Vancouver; one of the sections contains 463 rings from AD 1510 to 1972. This sample enabled us to study the $\Delta^{14}C$ variation systematically over ca 40 solar cycles. We selected rings from AD 1881 to 1925 because 1) a number of existing $\Delta^{14}C$ measurements ca AD 1900 on tree rings from locations with a latitude spread from 32° to 63°N in the northern hemisphere can be compared with our result, 2) the AD 1908 ring permits re-examination of the anti-matter hypothesis of the Tunguska event (Cowan, Atluri, and Libby, 1965). The widths of the rings are narrow. Except for the AD 1925 ring, we had to combine 2 and sometimes 3 rings to yield enough carbon for one measurement. We obtained 22 samples but one (1917-1919) was contaminated by an accident.

EXPERIMENTAL METHOD

Measurements were made at the History Department of Peking University, using a liquid scintillation photomultiplier tube device. After being treated with routine HCl-NaOH-HCl procedure to remove resin, the samples, ca 10g each, were converted to CO_2 by burning and then to benzene by the following steps: $CO_2 \rightarrow CaC_2 \rightarrow C_2H_2 \rightarrow$ benzene. In the last step, $CrO_3 \cdot Al_2O_3 \cdot SiO_2$ was used as the catalyst.

For one measurement, 5cc synthesized benzene was used. For smaller samples, we added background benzene to make up 5cc so that all samples were measured under the same experimental conditions. The samples were then sealed in a low potassium glass vial. After storing for ten days to allow for radon decay, ^{14}C activity was counted with two GDB-52L photomultiplier tubes in coincidence. The system was shielded against cosmic ray secondaries by 5 to 10cm of lead and a special circuit for pulse height discrimination was used to reduce cross-talk between the two photomultipliers. The net count rate for a 5cc modern sample was 39cpm; the background was 6cpm.

Precaution was taken to reduce uncertainties. Reliability and precision of the measuring system were checked by repeated preparation and counting of a ^{14}C calibration standard. An overall precision, which was defined as the standard deviation, was better than 5‰, including a pure statistical uncertainty of ca 3‰. Every run of the measurements was statistically tested. For each sample, we measured $^{13}C/^{12}C$ for fractionation correction.

EXPERIMENTAL RESULTS

Table 1 is a list of 21 tree-ring samples and their corresponding $\delta^{13}C$, $\Delta^{14}C$ (uncorrected) and $\Delta'^{14}C$ values

Table 1. Radiocarbon content in tree rings

Sample no.	Tree-ring dates	$\delta^{13}C$ ‰	$\Delta^{14}C$ ‰ (uncorrected)	$\Delta'^{14}C$ ‰ (corrected)
81301	1925	-26.05	-10.7 ± 4.4	- 2.6
81302	1923-1924	-26.40	+ 1.3 ± 4.7	+ 9.1
81303	1920-1922	-26.42	- 6.1 ± 3.8	+ 1.2
81304	1915-1916	-26.87	-10.4 ± 4.6	- 4.4
81305	1912-1914	-26.01	- 8.6 ± 4.4	- 3.2
81306	1910-1911	-25.57	- 1.0 ± 4.5	+ 3.9
81307	1908-1909	-26.01	-19.4 ± 4.2	-14.9
81308	1906-1907	-25.66	-16.9 ± 4.7	-12.8
81309	1904-1905	-26.45	- 1.5 ± 4.8	+ 2.2
81310	1902-1903	-26.25	+ 2.5 ± 4.4	+ 5.8
81311	1901-1902	-26.47	+ 1.7 ± 2.8	+ 4.8
82306	1899-1900	-26.75	+ 4.3 ± 3.0	+ 7.0
81313	1897-1898	-26.17	+ 2.8 ± 3.4	+ 5.1
81314	1895-1896	-26.18	+ 2.3 ± 2.8	+ 4.2
81315	1893-1894	-26.32	- 7.2 ± 3.1	- 5.7
81316	1891-1892	-25.90	- 6.4 ± 2.7	- 5.3
81301	1889-1890	-26.28	+ 3.8 ± 2.4	+ 4.5
82303	1887-1888	(-26.23)*	- 0.2 ± 2.9	+ 0.1
82304	1885-1886	(-26.23)*	+ 4.7 ± 3.6	+ 4.7
82305	1883-1884	(-26.23)*	- 0.8 ± 3.4	- 0.8
82302	1881-1882	-26.53	- 5.7 ± 3.0	- 5.7

*Estimated

(corrected for the Suess effects, linearly interpolated between 0‰ in 1885 and 8‰ in 1925). $\Delta^{14}C$ values were calculated according to Stuiver and Polach (1977):

$$\Delta^{14}C = \left\{ \frac{A_{sn} \exp[\lambda(1981 - x)]}{A_{abs}} - 1 \right\} \times 1000\ ‰$$

As a limited amount of oxalic acid SRN-4990 was available, A_{abs} was measured with Chinese sucrose charcoal standard which had been precisely intercalibrated against the international standard.

The data points are plotted in figure 1 along with other published $\Delta^{14}C$ values for comparison. To show the correlation with solar activity, we plotted, in the upper panel of figure 2, the sunspot numbers with the ordinates inverted and in the lower panel, $\Delta'^{14}C$ values. The cross-correlation coefficients with time differences -2, -1, 0, +1, and +2 years (- stands for a delay, + for an advance) are -0.26, -0.53, -0.58, -0.42, and -0.09, respectively. In view of the fact that sunspot numbers and the modulation of cosmic-ray intensity are only loosely correlated, the anti-correlation indicated in figure 2 is convincing.

We did not observe any measurable increase in $\Delta^{14}C$ in 1908, which might be caused by the Tunguska event. This agrees with the results of Lerman, Mook, and Vogel (1967).

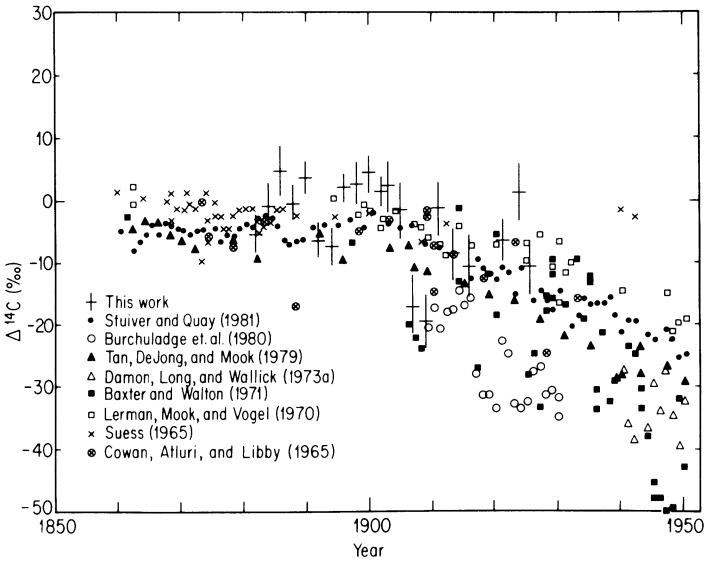

Fig 1. $\Delta^{14}C$ (uncorrected for the Suess effect)

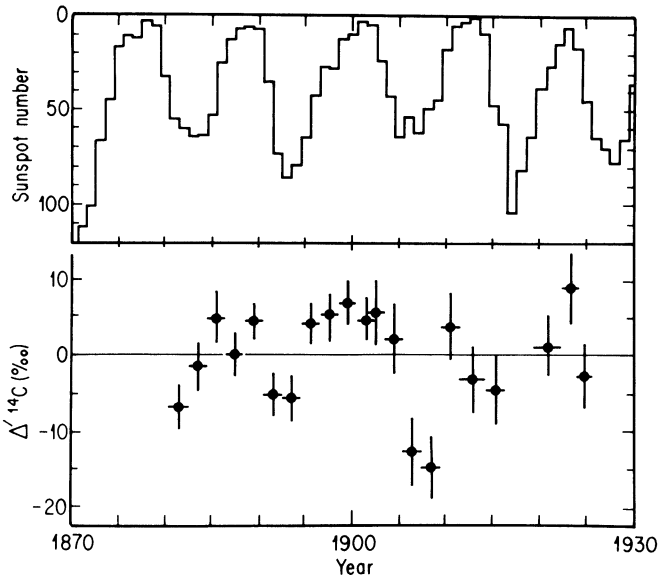

Fig 2. Upper panel: sunspot numbers
Lower panel: $\Delta'^{14}C$ (corrected for the Suess effect)

DISCUSSION

To explain the observation by de Vries (1958) of the variations of $\Delta^{14}C$ with time and location, Stuiver (1961) was the first to suggest that they are caused by the solar modulation of the ^{14}C production in the upper atmosphere. While the long-term $\Delta^{14}C$ change attributed to a variable sun has been studied by many investigators and the correlation established beyond any doubt (e g, Stuiver and Quay, 1980 and references therein), the puzzle remains as to why trees grown in high latitudes seem to respond to the 11-year cycle of cosmic-ray intensity change with higher sensitivity than those of lower latitudes. We have made the following observations: 1) The amplitude of the variation in ^{14}C production rates in a solar cycle increases with geomagnetic latitudes. Table 2 lists the ^{14}C production rates calculated by Lingenfelter and Ramaty (1970) in 1953-54 and 1957-1958 at various latitudes. The weighted averages of the amplitudes over the latitude ranges 0-60° and 60-90° are 11% and 33%, respectively. 2) After the last and most powerful high-altitude atomic bomb test in 1962, atmospheric ^{14}C concentration measured at latitudes 28-71°N were higher than those at latitudes 9-15°N, for almost three years (Nydal, 1968). This indicates that the time scale for the transport of ^{14}C

Table 2. ^{14}C production rate by cosmic ray

Geomagnetic latitude	1953-1954 ^{14}C production rate (per cm^2 sec)	1957-1958 ^{14}C production rate (per cm^2 sec)
0°	0.91	0.86
10°	0.94	0.89
20°	1.13	1.07
30°	1.70	1.51
45°	2.80	2.27
50°	4.20	3.19
60°	4.88	3.50
70-90°	4.99	3.50

across latitudes in the stratosphere is much longer than that in the troposphere. 3) The ocean surface area in the latitude range 60-70°N is only ca 30% of that of the land mass; beyond 70°N, it is frequently covered with ice. These physical conditions would cause the mixing rate of CO_2 with sea water to be much lower in this region than in lower latitudes.

We suggest that the atmosphere of the northern hemisphere can be divided into two boxes, 60-90°N (box 1) and 0-60°N (box 2), and the transport of ^{14}C may be approximately represented as follows:

Q_1, stratosphere ^{14}C source $\rightarrow$ ^{14}C(box 1) $\rightarrow$ ^{14}C(box 2) $\rightarrow$ ocean

Q_2, stratosphere ^{14}C source $\rightarrow$ ^{14}C(box 2) $\rightarrow$ ocean

$\hookrightarrow$ ^{14}C(box 1)

with $\delta Q_1 \sim 3\delta Q_2$. A preliminary calculation indicated that this model seems to explain the observed 10‰ fluctuation and the difference reported by Lerman, Mook, and Vogel (1970) and by Baxter and Farmer (1973) between the northern and southern hemispheres. We are presently continuing our measurements and working out the details of the proposed model.

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