# BOMB-PRODUCED <sup>14</sup>C IN TREE RINGS

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ABSTRACT. The <sup>14</sup>C content in 1961–1967 and 1970–1983 tree rings of a spruce grown in Dailing, China (47°N, 129°E) was measured by liquid scintillation. As a result of USSR bomb tests at Novaya Zemlya (72°N, 53°E),  $\Delta^{14}$ C values rose dramatically from 250‰ in 1961 to a maximum 909‰ in 1964, and then gradually decreased to 238‰ in 1983. We compared  $\Delta^{14}$ C values in the rings of an oak tree grown at 43°N, 74°W and that of a pine grown at 49°N, 9°E, and atmospheric  $\Delta^{14}$ C values in both northern and southern hemispheres. We observe that: 1) annual tree rings grown in the same latitude zone have the same  $\Delta^{14}$ C values, reflecting rapid longitudinal mixing of the atmosphere; 2) atmospheric <sup>14</sup>C concentrations reached a global equilibrium distribution at the end of 1968, and tree ring <sup>14</sup>C content reflects atmospheric <sup>14</sup>C concentration; 3) 1976–1982 rings of the Dailing spruce show excessive <sup>14</sup>C, likely due to the effect of 1976 and 1980 Chinese bomb tests; 4)  $\Delta^{14}$ C decreases exponentially, halving every 17 yr.

#### INTRODUCTION

The USSR conducted a series of nuclear tests during 1961–1962 at Novaya Zemlya, located at 72°N, 53°E, which contributed about 338 Mt (megatons of TNT equivalent) to the atmosphere. The very high concentration of <sup>14</sup>C produced at high northern latitudes offered a unique opportunity to study atmospheric mixing of CO<sub>2</sub> before uptake by the biosphere and ocean. To study latitudinal mixing in the troposphere, we obtained three spruce pines grown at three different latitudes: Mackenzie Delta (68°N, 130°W), Dailing (47°N, 129°E) and Mingyin (27°N, 100°E); we measured the <sup>14</sup>C content in 1961–1967 annual rings.  $\Delta^{14}$ C values rise dramatically from the 1961 level (~250‰) to their respective maxima, 964‰, 909‰ and 743‰ in 1964, and then fall to ~680‰ in 1967. The meridional gradient in <sup>14</sup>C concentration was clearly demonstrated. We also noted that the Mackenzie spruce responded quickly to bomb-produced <sup>14</sup>C, the Dailing spruce responded with a slight delay, and the Mingyin spruce responded even later; these differences presumably reflect the mechanism of meridional mixing in the troposphere. These results were reported at the 12th International Radiocarbon Conference (Dai & Fan 1986).

The Dailing spruce was cut down in 1985, and we were able to obtain useful ring samples up to 1983. We measured  $\Delta^{14}$ C for two reasons:

- Levin et al. (1985) measured the Δ<sup>14</sup>C values in the 1966-1982 rings of a pine grown at Obrigheim, Germany (49°N, 9°E) to study the effect of a 300-megawatt nuclear plant 4 km from the pine. Because Obrigheim and Dailing are practically along the same latitude, but separated by 120° of longitude, it would be interesting to check two high Δ<sup>14</sup>C values believed to be affected by the power plant. Further, from 1964 to 1980, China conducted 22 atmospheric nuclear tests at Lop Nor (40°N, 90°E), 8 of which had yields ≥1 Mt. Dailing is 3200 km downwind from Lop Nor, and may have been affected.
- 2. Telegadas (1971) found that by the end of 1968, bomb-produced <sup>14</sup>C had reached a global equilibrium distribution. Thus, it is desirable to measure the decrease of  $\Delta^{14}$ C in tree rings, which reflects the decay rate of <sup>14</sup>C concentration. This is important for calculating the residence time of <sup>14</sup>CO<sub>2</sub> in the atmosphere.

We report here our measurements and discuss the results.

## 754 Dai Kaimei, Qian Youneng and C. Y. Fan

## MEASUREMENTS AND RESULTS

Measurements were made with the Nanjing University liquid scintillation counter (LSC) system. After repeated HCl-NaOH-HCl treatment, each of the wood samples was converted to  $\text{Li}_2\text{C}_2$  by reaction with lithium, then to  $\text{C}_2\text{H}_2$  by  $\text{Li}_2\text{C}_2$  reaction with H<sub>2</sub>O, and finally catalytically to benzene ( $\text{C}_6\text{H}_6$ ). A plastic scintillation anticoincidence device and a lead shield, 6–10 cm thick, were used to reduce LSC background. Each measurement used a 5-cc benzene sample mixed with 1 cc scintillation liquid in a quartz counting vial. The count rate of a background sample was 2.0 cpm when the counting efficiency was 70%. We made certain that identical experimental conditions existed for all measurements.

Table 1 lists the  $\Delta^{14}$ C values of 20 rings of the Dailing spruce; seven were reported earlier (Dai & Fan 1986). The 1968 and 1969 rings were not measured because of human error. The results, together with the  $\Delta^{14}$ C measured in an oak grown in Bear Mountain Park, New York (43°N, 74°W) by Cain and Suess (1976) and in a pine grown in Obrigheim, Germany (49°N, 9°E) by Levin *et al.* (1985) are plotted in Figure 1; error bars are too small to be shown. To show the latitude effect in  $\Delta^{14}$ C in trees prior to 1968, published values for a white spruce (68°N, 130°W) and spruce (27°N, 100°E) are also included in Figure 1.

Burchuladze *et al.* (1989) reported  $\Delta^{14}$ C values of 18 Georgian wine samples from 1970 to 1987. Because the growth season of wine grapes is different from that of a tree, these results are not included in Figure 1.

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Year $\Delta^{14}C$	1961 251	1962 392	1963 770	1964 909	1965 843	1966 764	1967 682	1968	1969	1970 549	1971 523	1972 436
	1973 431	1974 398	1975 393	1976 384	1977 367	1978 355	1979 348	1980 340	1981 310	1982 287	1983 238	

TABLE 1.  $\Delta^{14}$ C in 1961-1983 Tree Rings of a Dailing Spruce (in  $\% \pm 5\%$ )

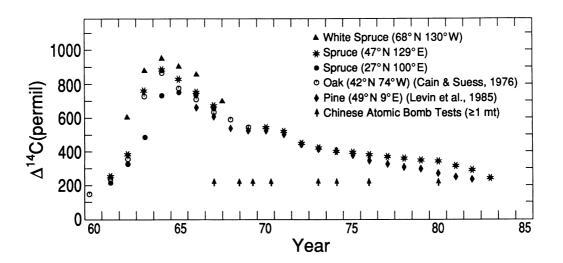


Fig. 1.  $\Delta^{14}$ C in tree rings from different locations

# DISCUSSION AND CONCLUSION

The striking difference between the  $\Delta^{14}$ C values of the Dailing spruce and those of the Obrigheim pine (Fig. 1) lies in the excess of <sup>14</sup>C in the Dailing spruce in 1976–1982 rings. Otherwise, the trees grown in the same latitude zone but separated widely in longitude responded closely to the bomb-produced <sup>14</sup>C, indicating rapid longitudinal mixing of atmospheric CO<sub>2</sub>. The arrows in Figure 1 represent the dates of the Chinese bomb tests yielding  $\geq 1$  Mt. We suggest that the excess in <sup>14</sup>C in the Dailing spruce results from the 1976 and 1980 tests; the 1976 test is the largest of the series (~4 Mt). The  $\Delta^{14}$ C values in the rings of a Japanese cypress grown at 35°6'N, 137°5'E reported by Nakamura, Nakai and Ohishi (1987) show two conspicuous maxima at 1976 and 1981–1982, respectively. These may also be due to the two Chinese bomb tests.

In the Obrigheim pine, the <sup>14</sup>C content in the 1970 ring is higher than in the 1969 rings. Levin *et al.* (1985) suggested that the nearby power plant caused this effect. The  $\Delta^{14}$ C value of the Obrigheim pine is identical to that of Dailing spruce. In the wine samples studied by Burchuladze *et al.* (1989), <sup>14</sup>C contents in 1970, 1971 and 1972 wine are also exceptionally high. Therefore, we believe that the excess in <sup>14</sup>C is a global effect.

Manning *et al.* (1990) measured  $\Delta^{14}$ C in the atmosphere collected at Wellington, New Zealand and several other sites in the South Pacific from 1954 to 1987. They then used smooth spline curves plotted in a semilog scale to fit all available  $\Delta^{14}$ C data, one for the northern and the other for the southern hemisphere (Fig. 2 of their paper). We reproduce their curves in Figure 2, with two additions: 1) an extrapolation of stratospheric  $\Delta^{14}$ C, assuming that the half-residence-time of excess  $\Delta^{14}$ C in the stratosphere is nine yr, according to Sowl *et al.* (1976); 2)  $\Delta^{14}$ C in the Dailing spruce from 1961 to 1983 (with two missing years, mentioned above). From Figure 2, the  $\Delta^{14}$ C excess found in 1976–1982 rings again becomes apparent. Otherwise,  $\Delta^{14}$ C in tree rings and atmospheric  $\Delta^{14}$ C generally agree. This indicated that, for any given year, the organic carbon assimilated by the tree from atmospheric CO<sub>2</sub> was used directly to form tree-ring cellulose in that year.

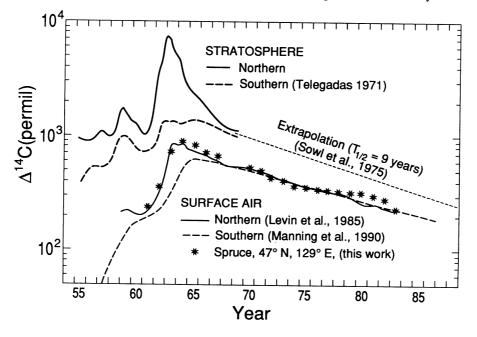


Fig. 2. Comparison of atmospheric  $\Delta^{14}$ C with  $\Delta^{14}$ C in tree rings of a Dailing spruce

# 756 Dai Kaimei, Qian Youneng and C. Y. Fan

Excluding the  $\Delta^{14}$ C values in 1976–1982 rings from the data set, we determined that tree-ring  $\Delta^{14}$ C decreased by half over 17 yr, the same as Manning *et al.* (1990) suggested. We note that this is not the residence time of CO<sub>2</sub> in the atmosphere, because there is constant exchange of tropospheric CO<sub>2</sub> with that in the stratosphere, the ocean and the biosphere; the  $\Delta^{14}$ C in the latter three "boxes" differ from the atmospheric value.

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