

^{14}C MEASUREMENTS OF TREE RINGS OF A JAPANESE CEDAR DURING 1945 TO 2000 AND CORE SAMPLING FOR ENVIRONMENTAL STUDIES

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ABSTRACT. Japanese cedar rings sampled from Aomori Prefecture, Japan, were measured to obtain tree-ring radiocarbon data covering a period of the latter half of the 20th century in the northernmost area of Honshu (mainland Japan), to obtain records of variation in atmospheric ^{14}C concentration caused by past atmospheric nuclear testing, and to study any possible local effects. This work, carried out on a partial disk, was also intended to provide a reference for data obtained by core sampling of live, standing Japanese black pines as a part of marine environmental studies.

INTRODUCTION

The relationship between tree-ring radiocarbon data and air ^{14}C concentrations is used in archaeological chronologies and in environmental radioactivity studies (Cain and Suess 1976). As a result of large-scale atmospheric nuclear tests in the 1950s, air ^{14}C concentration increased considerably and, after the Partial Test Ban Treaty went into effect, exponentially decreased in the Northern and Southern hemispheres (UNSCEAR 2000). Tree-ring ^{14}C data covering a period of the latter half of the 20th century in Japan, however, are relatively scarce and limited to the central areas. Also, local influences on tree ^{14}C concentrations on the earth and among regions still seem to be a matter of interest (Yamada et al. 2005).

In this study, ^{14}C concentrations of Japanese cedar tree rings were measured without cross-dating of rings (partly because a local master chronology in the Aomori area does not seem to be fully established yet), a) to obtain tree-ring ^{14}C data covering a period of the latter half of the 20th century in Aomori Prefecture, Japan, the northernmost area of Honshu (mainland); b) to obtain records in this area of variation in the atmospheric ^{14}C concentration caused by the past nuclear testing; and c) to study any possible locality in tree-ring ^{14}C concentrations.

Results of this work, carried out on a partial disk, were also intended for use as reference data for environmental studies (Isogai et al. 2002) with respect to possible interactions between the air and the surface seawater. We used tree-ring ^{14}C data by core sampling (Otlet et al. 1983; Rakowski et al. 2004) Japanese black pines in windbreak forests on the east coast of northern Sanriku (facing the northwestern Pacific Ocean).

MATERIALS AND METHODS

Sampling

A block of Sugi or Japanese cedar (*Cryptomeria japonica*), from ~5 m above ground, was obtained from a newly fallen specimen in Kobashi, Aomori, Japan (40.92°N, 140.63°E; see Figure 1), from which 1.5-cm-thick partial disks were prepared (Figure 2). Core samples were obtained from Kuromatsu or Japanese black pines (*Pinus thunbergii*) by a Haglov Model CO 812 increment borer (1.2 cm inner diameter, 1.5 cm outer diameter, 45 cm long) in Rokkasho, Aomori. A pair of cores was bored in 2 directions from each tree. Following boring, regular protective measures were taken at each boring point. Technical training was provided by the Forest Tree Breeding Center (Hitachi, Ibaraki) and its Tohoku Regional Breeding Office (Takizawa, Iwate).

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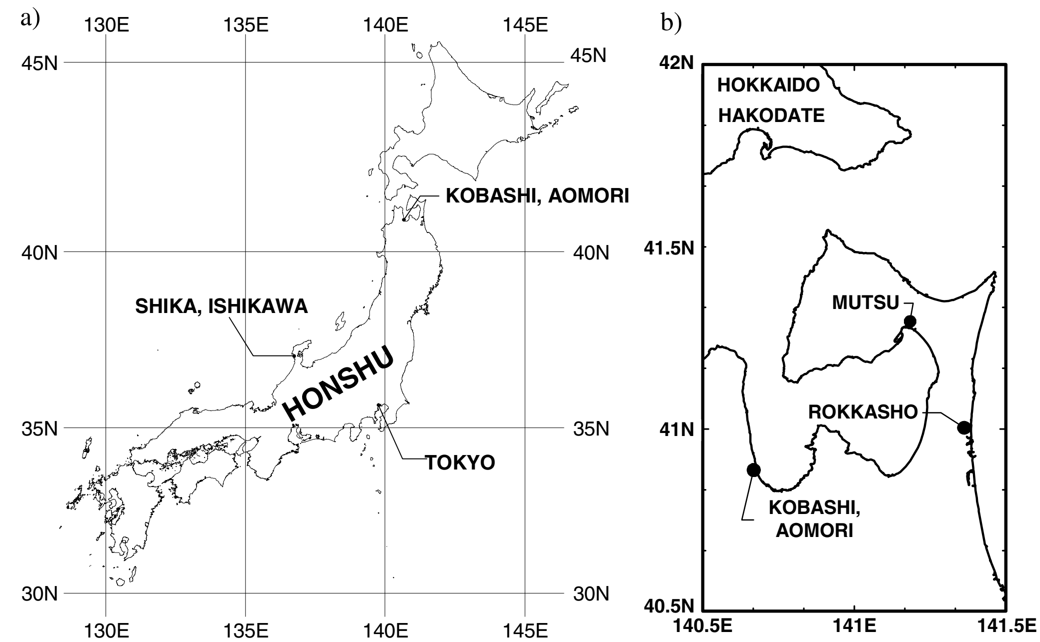

Figure 1 Sampling locations of the Japanese cedar, Kobashi (a); and Japanese black pines, Rokkasho, Aomori (b).

Figure 2 A 1.5-cm-thick partial disk from the Japanese cedar block sample obtained from Kobashi, Aomori, Japan. The bark, ~3-cm-thick originally at the left side, was lost in sawing. The right side is the inner wall of the hollow part of the trunk. Scale in cm.

Sample Preparation

Each ring was cut out as units of early wood and late wood. Rings weighing 0.25 to 1.4 g in the partial disk and ~0.05 g and up in core samples were sliced into very thin sections and subjected to repeated washing with purified water in an ultrasonic bath. The samples were then treated in turns with 1.2M hydrochloric acid, 1.2M sodium hydroxide, and 0.07M sodium chlorite on a hot plate. Alpha-cellulose fractions were recovered after further treatment with 17.5% sodium hydroxide.

An α-cellulose sample (~13 mg dry) was combusted with CuO using Ag catalyst in a sealed quartz tube at 850 °C for 2 hr, and the resultant gases were purified for CO_2 by using a glass vacuum line for $^{14}C/^{12}C$ measurement, $\delta^{13}C$ determination, and a reserve sample. Part of the purified CO_2 was reduced to graphite by pure H_2 using an Fe catalyst in a sealed Pyrex® tube at 650 °C for 6 hr (Kitagawa et al. 1993). Graphite samples together with iron powder were pressed into aluminum target holders and subjected to accelerator mass spectrometry (AMS) measurement for ^{14}C. NIST SRM 4990C, IAEA-C1 marble, and IAEA-C6 sucrose were used as the standard, blank, and quality control reference sample, respectively.

Measurement and Calculation of Results

Measurement of $^{14}C/^{12}C$ was carried out by the Mutsu Establishment, Japan Atomic Energy Agency (formerly Japan Atomic Energy Research Institute), using an HVEE Model 4130 accelerator mass spectrometer. The time of measurement per sample was 2700 s. The CO_2 samples were measured for $\delta^{13}C$ by Shoko Tsusho Co. Ltd., Tokyo, by using a Finnigan-MAT Delta^{PLUS} XL stable isotope mass spectrometer. Calculation of ^{14}C was carried out following Stuiver and Polach (1977).

RESULTS AND DISCUSSION

Analytical Results

$\Delta^{14}C$ data was obtained for the purpose of comparing the present results and literature data, while specific activity was also calculated to know absolute concentration of ^{14}C in the environment. The ^{14}C measurements as expressed in Bq per g of carbon (Bq/g C) obtained for the cedar rings between 1945 and 2000 are shown in Figure 3. The ^{14}C level seemed almost unchanged during the period 1945–1953. Then, it started to increase and reached a small peak in 1959. Following a sharp rise after 1961, it reached a maximum in 1964. After 1964, the Aomori cedar tree-ring ^{14}C showed decreasing concentrations until 2000 with small fluctuations in the 1970s and 1980s.

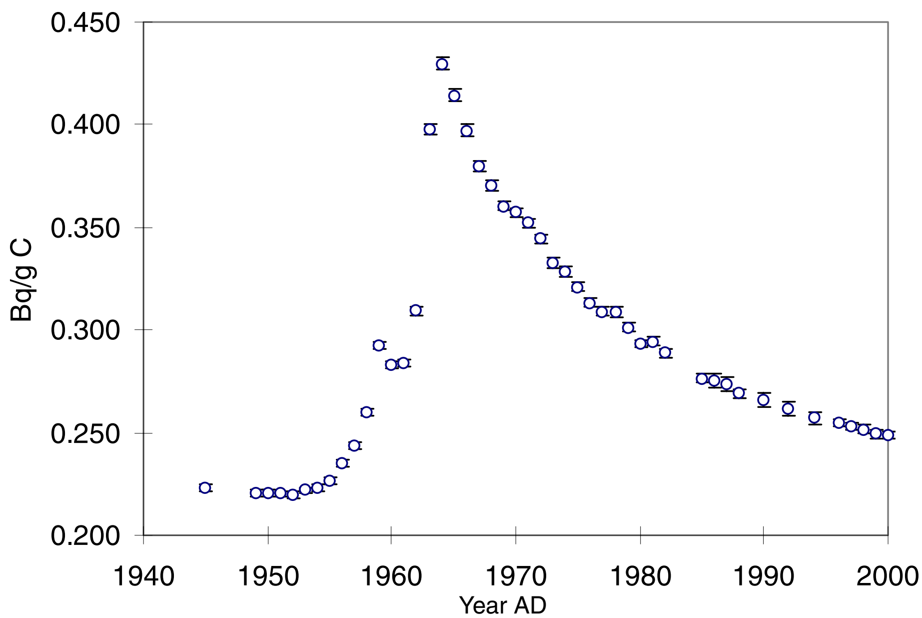

Figure 3 ^{14}C radioactivity concentrations in Japanese cedar in Aomori during the period 1945–2000 (in Bq/g C; error bar: ±1 σ).

These changes were roughly parallel with the reported yield of atmospheric nuclear tests (Table 1; UNSCEAR 2000:207). Data from UNSCEAR shows that the global release of ^{14}C produced and globally dispersed in the atmospheric nuclear testing was 213 PBq (UNSCEAR 2000:52). Time lags between injections of bomb-produced ^{14}C to Earth's atmosphere and incorporation by tree rings must be considered, as influenced by the atmospheric transport, growth of trees, etc. The changes during 1954 to 1964 were also confirmed to be due to large-scale atmospheric nuclear tests carried out by the USA, USSR, and UK until 1962. Fluctuations found in the decreasing curve of tree ^{14}C concentrations may have reflected a smaller number of atmospheric tests carried out by China in the Northern Hemisphere until 1980 and perhaps less by those carried out by France in the Southern Hemisphere until 1974.

Table 1 Annual fission and fusion yields of atmospheric nuclear tests (UNSCEAR 2000:207).

Year	Yield (Mt)	Year	Yield (Mt)	Year	Yield (Mt)
1945	0.057	1957	9.64	1969	3
1946	0.042	1958	56.8	1970	5.78
1948	0.10	1960	0.072	1971	1.46
1949	0.022	1961	86.5	1972	0.13
1951	0.59	1962	170.4	1973	2.52
1952	11.0	1964	0.02	1974	1.21
1953	0.71	1965	0.04	1976	4.12
1954	48.3	1966	1.14	1977	0.02
1955	2.06	1967	3.18	1978	0.04
1956	22.9	1968	7.60	1980	0.6

A possible effect of the ^{14}C release (total 44 PBq) from 26 April to 5 May 1986 in the Chernobyl accident was sought in 1986 and 1987 rings because excessive ^{14}C concentrations were reported in early woods of 1986 rings around Chernobyl (Buzziny et al. 1998). In the present data, such an effect seemed, however, to be insignificant within the errors of measurement.

During 1991–2000, the apparent yearly decrease (in ^{14}C Bq/g C) was estimated to be larger than 0.5%. The trend may reflect the atmospheric ^{14}C concentration as controlled by transfer to the biosphere, to Earth's surfaces, and to seawater. Dilution of ^{14}C caused by combustion of fossil fuels should also be considered (UNSCEAR 2000; Fuma et al. 2002; Rakowski et al. 2004) since Kobashi is near Aomori City (population 297,859 in the year 2000).

COMPARISON WITH DATA FROM PREVIOUS STUDIES

Tree-Ring Data from Other Regions

The ^{14}C values obtained in Kobashi, Aomori (40.9°N, 140.6°E) may be compared with tree-ring data reported from Germany (Schauinsland, 48°N, 8°E; and Obrigheim, 49°N, 9°E), Hungary (47.35°N), and China (Dailing, 47°N, 129°E) in the mid-latitude regions of the Northern Hemisphere as shown in Figure 4 (Levin et al. 1985; Kaimei et al. 1992; Levin and Kromer 1997). The trend is generally in agreement, but some differences were noticed.

For 1963–4, the Japanese Aomori data, obtained from an apparently lower latitude, were lower than the Hungarian and Chinese Dailing tree rings. However, in 1965–7, the Aomori data were lower than the Chinese but higher than the Hungarian data.

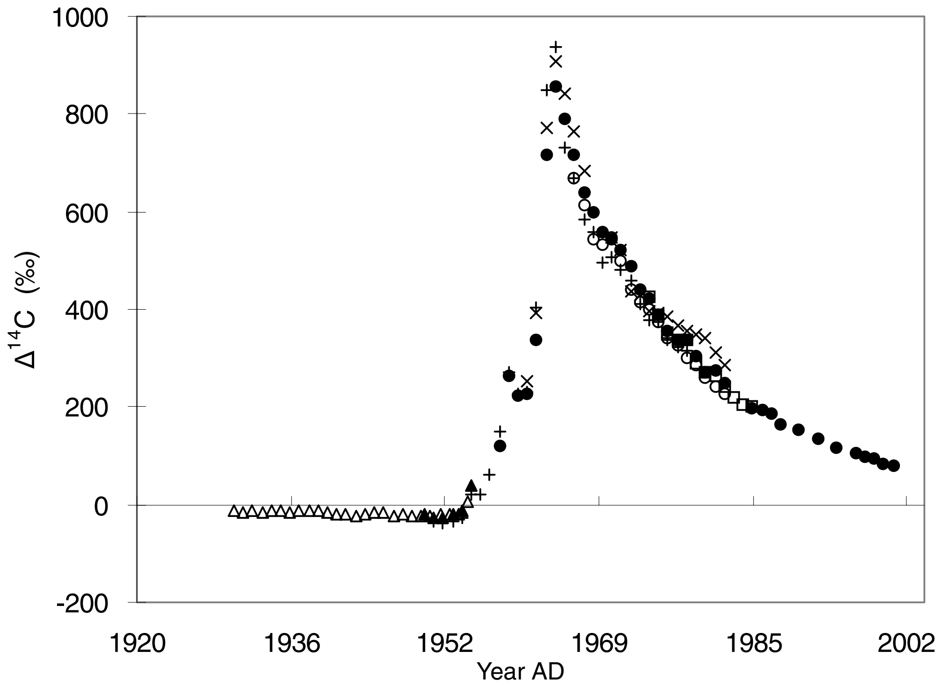

Figure 4 Comparison of Δ^{14}C of the Japanese cedar in Aomori with tree-ring data in previous studies (reconstructed from Levin et al. 1985; Kaimei et al. 1992; Levin and Kromer 1997). Symbols: • = Aomori, Japan (41°N, 141°E); empty squares = Schauinsland, Germany (48°N, 8°E); empty circles = Obrigheim, Germany (49°N, 9°E); empty triangles = Arizona, USA (32.26°N, 110.47°W); × = Dailing, China (47°N, 129°E); + = Hungary (47.35°N); filled triangles = New York, USA (41.3°N, 74.0°W).

Comparing available data between 1976 and 1982, the Chinese Dailing Δ^{14}C (Kaimei et al. 1992) was clearly higher than the Japanese Aomori, Hungarian, and German data, especially in 1980. The Aomori and Schauinsland data seem almost identical during the period.

Major atmospheric nuclear tests with fusion yields of 1.1 to 1.8 Mt were carried out in Lob Nor in June 1967, December 1968, September 1969, October 1970, July 1973, and November 1976, with smaller tests in 1964–5, 1966–7, 1971–2, 1974, 1976, 1978, and 1980. A major atmospheric test with a fusion yield of 1.3 Mt in the Southern Hemisphere was carried out in August 1968 (UNSCEAR 2000:195). The higher Dailing Δ^{14}C values compared to others observed during 1976–1982 may be due to Dailing's closer proximity to the test site (Kaimei et al. 1992).

Tree-Ring Data from Elsewhere in Japan

Recently, tree-ring ^{14}C data for a Japanese black pine was reported from central Shika-machi, Ishikawa, in an area facing the Japan Sea (37.0°N, 136.8°E; Yamada et al. 2005). Other data reported on tree-ring ^{14}C values from Hinoki (Japanese cypress, *Chamaecyparis obtuse*), obtained from Kiso, Gifu (35.6°N, 137.5°E) in central mainland Japan (Nakamura et al. 1987), were not plotted in the figure.

To compare peak values among the above 3 data sets, Kobashi data was higher than Shika and Kiso data during 1964–6. In comparison with Dailing, China (47°N), and Hungary (47.35°N), however, Aomori (41°N) showed lower values as shown in Figure 4. There apparently exists a fluctuation or slight increase in Δ^{14}C during the period 1968–1971 in the Kobashi data that may be attributable to Chinese nuclear tests (Kaimei et al. 1992).

It should be pointed out, though, in this and further comparisons with existing data, that the possibility of any very small errors in sample preparation and measurement cannot be excluded, nor errors incurred when counting rings. Some regional factors including the so-called latitudinal effect, however, can still be inferred from the data relevant to that period. These data suggest a need for further study, including cross-dating of rings.

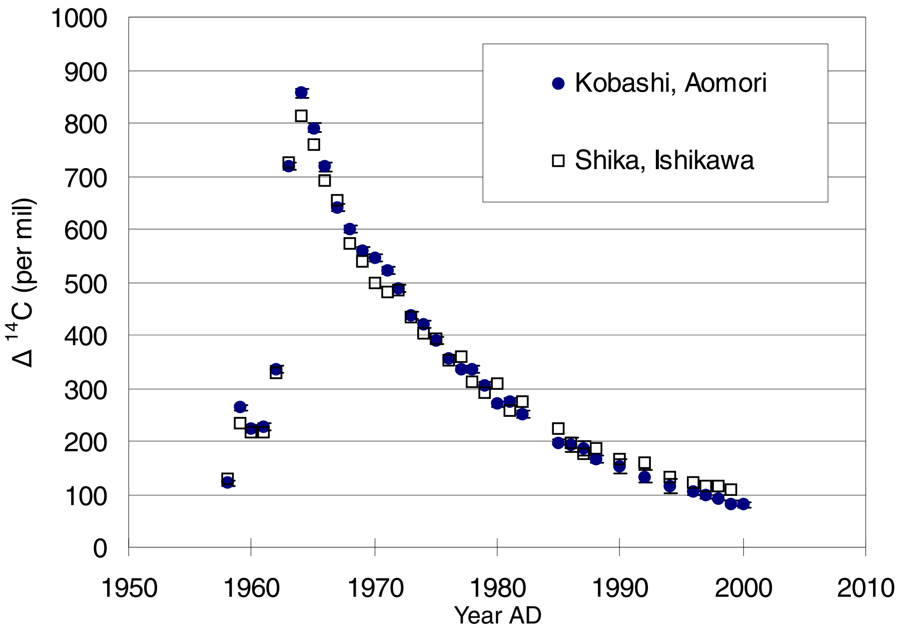

Figure 5 Comparison of the present data (Kobashi, Aomori) with the reported tree-ring ^{14}C from central Japan, facing the Japan Sea (Shika, Ishikawa). Shika plots reconstructed from data in Yamada et al. (2005).

Air ^{14}C Data

The data seem to be largely in accordance with air ^{14}C concentrations reported until the 1990s, except during 1962–4 (Figure 6). A decrease in ^{14}C during the period 1979–2000 was fitted with a single exponential function with a half-life of 11.0 yr and an apparent lifetime of 15.9 yr (yr = 299.9 $e^{-0.06276t}$).

In red pine (*Pinus densiflora*) in Korea, the steady decrease of $\Delta^{14}C$ from 1965–2000 was described with a single exponential function with a lifetime 15.99 ± 0.43 yr, which was similar to that of the high-latitude region in Europe (Park et al. 2002). For measurement of Japanese wine during 1984–2000, the decrease in the $\Delta^{14}C$ lifetime was reported to be 14.86 yr (Fuma et al. 2002).

^{14}C Concentrations in Live Japanese Black Pines as Studied by Core Sampling

Distribution of $^{14}C/^{12}C$ in a Single-Year Ring

Two pairs of core ring samples, taken from the west (open side) and east (hill) sides of a trunk of a Japanese black pine, were analyzed separately for ^{14}C concentration. The results were similar: 110.4 ± 0.60 and 110.6 ± 0.67 pMC for the 1997 ring; and 109.3 ± 0.61 and 109.5 ± 0.63 pMC for the 1999 ring, respectively. The result, although from only 1 tree, may indicate that differences in ^{14}C due to direction of boring are within errors of measurement and suggest that a ring sample from a core may represent the whole ring.

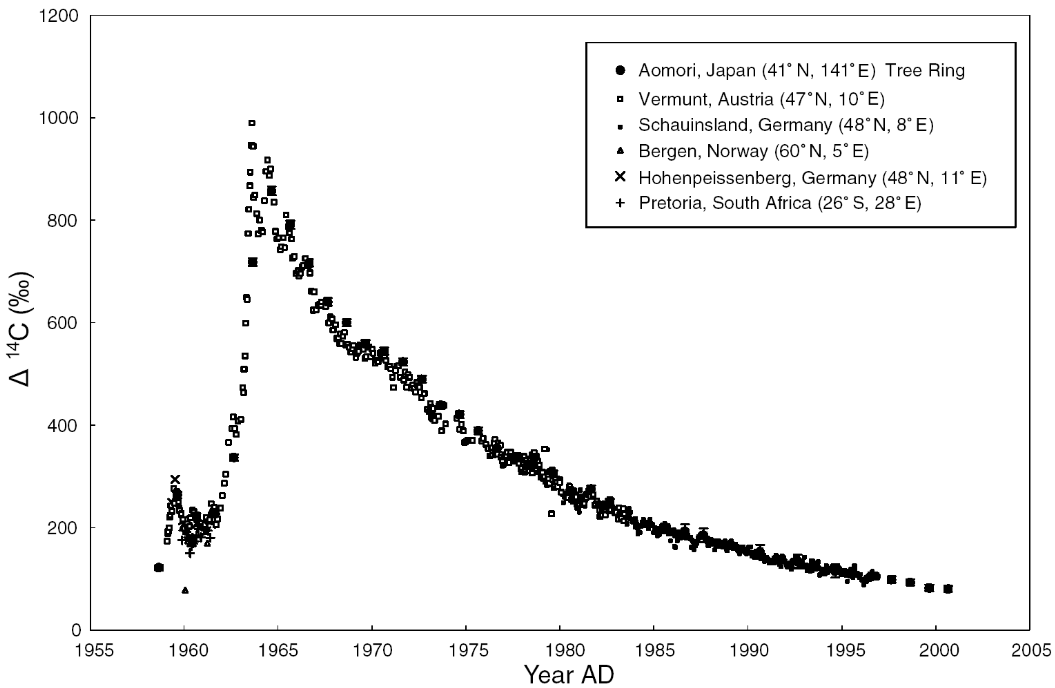

Figure 6 ^{14}C in the Japanese cedar in Aomori, Japan, and literature ^{14}C data for the atmospheric CO_2 (reconstructed from Levin et al. [1985] and Levin and Kromer [1997]; abscissa, reported date in the atmospheric data, and middle of year assumed in the tree data).

Trends in ^{14}C Concentrations in Coastal Japanese Black Pines

^{14}C concentrations in core ring samples obtained for 8 Japanese black pines were pooled (see Figure 7). The concentration levels were found to be close to those obtained for the Japanese cedar during the same period, and the pine ^{14}C concentration levels also generally decreased slowly from 1991 to 2001. During that period, linear correlation analysis was made for the approximation of the trend in ^{14}C activity and an average yearly decrease of ~0.5% was obtained.

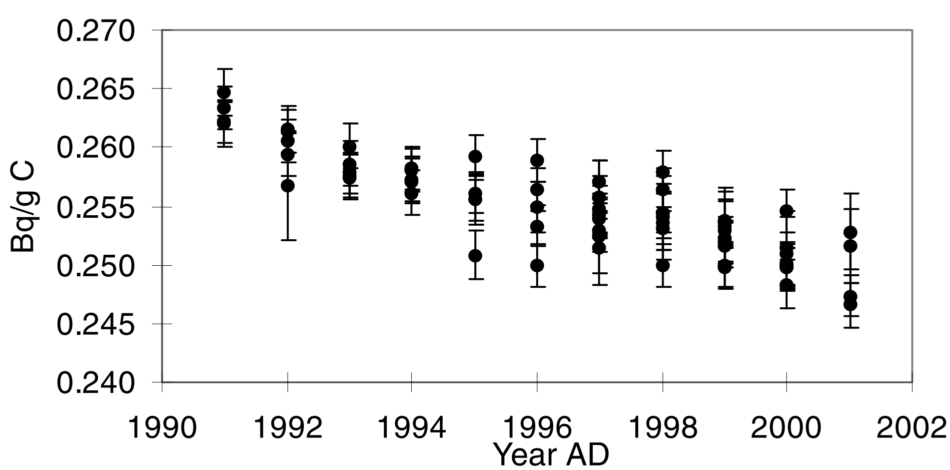

Figure 7 ^{14}C concentrations in Japanese black pines (filled circles represent data for each tree; error bars ±1 σ).

As for the solar cycle of 11 yr, a solar minimum probably existed around 1996–1997 according to solar luminosity data (Foukal 2003). When considering only a small change in the atmospheric ^{14}C concentration, influence of the solar minimum in these data may not be expected.

Merits and Demerits of the Core Sampling of Live, Standing Trees for ^{14}C

For the past 3 or 4 yr after the first core sampling, there has been no discernible effect on the growth of the trees under study. The results as a whole suggest that intermittent core sampling from the same group of trees may conveniently provide "average" yearly data on the atmospheric ^{14}C in an area of interest. However, one should also be aware of some limitations of the technique, e.g. the difficulty of identifying false rings.

CONCLUSION

The tree-ring ^{14}C data obtained for a Japanese cedar showed known influences of past atmospheric nuclear tests during the period 1945–2000 in Aomori, Japan. The data were largely in accordance with air ^{14}C concentrations reported until the 1990s as well as with tree-ring ^{14}C data from mid-latitude regions of the Northern Hemisphere. Slight differences between the present results and comparable data reported elsewhere may suggest a need for further study of spatial differences.

Core sampling on standing, live Japanese black pines demonstrated that the technique might, without destroying trees, provide possible continuous ^{14}C measurement in environmental studies in an area of interest.

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