

RADIOCARBON ANALYSIS OF TREE RINGS FROM A 15.5-CAL KYR BP PYROCLASTICALLY BURIED FOREST: A PILOT STUDY

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ABSTRACT. We have determined the radiocarbon ages for 40-yr-interval tree rings in 2 fossil trees of the Towada Hachinohe buried forest, northeastern Honshu Island, Japan. The ¹⁴C ages range from 13.0 to 13.3 kyr BP (about 15.5 cal kyr BP). The weighted average of the ¹⁴C age of the outermost 5 rings is 13,133 ± 33 BP, which can be calibrated to 15,363–15,679 cal BP by using the IntCal04 standard curve (Reimer et al. 2004). The estimated Δ¹⁴C values range between 265 and 300‰ and show approximately sinusoidal fluctuation of an indicated ~200-yr cycle, perhaps reflecting contemporary solar activity change. Comparison between the tree ¹⁴C profile and the Cariaco Basin ¹⁴C record provides further information on the accurate date of the Towada Hachinohe buried forest and the eruption that produced it. ¹⁴C analysis of tree rings from the buried forest may contribute to the construction of a better ¹⁴C calibration curve and the elucidation of solar activity change during the last glacial period, as well as possible global and regional impacts of the huge eruption from Towada Volcano.

INTRODUCTION

Accurate and precise knowledge of past radiocarbon concentration in the atmosphere is crucial not only for ¹⁴C dating but also for solar and Earth system science. Long-term records of atmospheric ¹⁴C concentration have been reconstructed by using evidence derived from tree rings (e.g. Stuiver et al. 1986), coral (e.g. Bard et al. 1998), speleothems (e.g. Beck et al. 2001), terrestrial fossils in varved sediments (e.g. Kitagawa and van der Plicht 2000), and biotic or abiotic carbonates in marine and lacustrine sediments (e.g. Voelker et al. 2000; Hughen et al. 2004a,b; van der Borg et al. 2004). These records have been used, for example, to determine past changes in solar activity (e.g. Stuiver and Quay 1980; Solanki et al. 2004) and for evaluating ocean circulation changes (e.g. Broecker et al. 2004; Robinson et al. 2005).

Tree growth rings are evidently the best archives of past ¹⁴C concentration. Unlike the other archives, tree rings directly record information on atmospheric ¹⁴C, and we do not need to consider any reservoir ages. The single-year resolution that is possible from the rings is superior to the time resolution derived from other records (e.g. Stuiver and Braziunas 1998; Miyahara et al. 2006). Nevertheless, tree-ring ¹⁴C age determinations have been until now restricted in their applicability to within the last 12.4 cal kyr BP (Reimer et al. 2004), except for a few scattered results (e.g. Gandou et al. 2004; Kromer et al. 2004). The reasons are the questionable precision of the ¹⁴C measurement, the scarcity of samples, and the weak framework of calendar age for the last glacial phase. However, the precision of accelerator mass spectrometry (AMS) ¹⁴C measurements is now progressively increasing (e.g. Fifield 2000; Bronk Ramsey et al. 2004). Tree-ring samples from the last glacial age are often excavated from middle- to low-latitude volcanic islands such as those of Japan. With the help of local dendrochronology, the ¹⁴C wiggle-matching technique applied to tree rings of the last glacial period (Kromer et al. 2004) and certain varved sediments (Kitagawa and van der Plicht 1998; Hughen et al. 2000, 2004b) may solve the problem of calendar age. Studies on former ¹⁴C atmospheric concentration based on tree rings of the last glacial epoch are thus now becoming feasible.

We have been exploring the possibility of high-resolution AMS ¹⁴C analysis of tree rings by using 2 stumps of coniferous trees obtained from a 15.5-cal kyr BP forest buried beneath volcanic ash. The

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Microscope observations show both trees to be of the genus *Picea*, and burnt bark is retained on the wood. A dendrochronological investigation of the Towada Hachinohe buried forest (Terada et al. 1994) has revealed that almost all such erect *Picea* stumps with remaining bark were killed at the same time. This characteristic provides us with a unique situation in which we can cross-check the ¹⁴C data of the last glacial period using the ages derived from the 2 stumps. It is also an important consideration that the appearance of the stump tree rings is fresh, resembling that of a living tree (Figure 1). Postdepositional alteration is likely to be minimal when compared to that ordinarily encountered in buried trees and plant remains.

ANALYTICAL METHODS

Both the A and B stumps were brought to the paleoenvironmental and cosmogenic nuclide laboratory of Hirosaki University, where they were dried on a clean bench for 4 weeks. A tree-ring count showed the total number of rings in A and B as 144 and 172, respectively. These rings are numbered inward, starting with #1 assigned to the outermost ring. For this preliminary study, thin samples of well-preserved wood (<1 mm), each of which covered a 40-ring interval, were taken from the stumps using a razor. The typical weight of the samples was 0.1 g. These were homogenized in an agate mill, and the outermost 5 rings of each stump were additionally subsampled. All samples were subjected to acid-alkali-acid (AAA) pretreatment.

Combustion of the samples, CO₂ purification, and graphitization were performed in the AMS facility of the University of Tokyo (MALT; Matsuzaki et al. 2004). Duplicate graphite targets were produced for each CO₂ gas sample (although a few graphite targets failed to produce sufficient mass during graphitization). ¹⁴C analysis was performed by using the MALT AMS (Matsuzaki et al. 2004). The analytical precision was from 50–70 yr.

RADIOCARBON AGES AND Δ¹⁴C

The results of the ¹⁴C analysis are shown in Table 1. The ¹⁴C ages range between 13.0 and 13.3 kyr BP. The ages of the same ring number agree well between stumps A and B, supporting the high reproducibility of our analysis, and there is no evidence in these internally consistent results that they were contaminated in situ with younger or older carbon. The ¹⁴C ages of 13.0 and 13.3 kyr BP are within the range of the previous radiometric and AMS ¹⁴C ages (about 12–14 kyr BP) of the buried forest and volcanics, which concentrate around 13 kyr BP (see Machida and Arai 2003). The ages correspond to ~15.5 cal kyr BP in the IntCal04 curve (Reimer et al. 2004), which is consistent with δ¹⁸O-stratigraphic evidence from the marine sediment core LM-8 (Aoki and Arai 2000), suggesting the stratigraphic position of the ToH tephra to be around 15 cal kyr BP (Aoki and Arai 2000; Machida and Arai 2003).

Sakurai et al. (2004) and Gandou et al. (2004) have analyzed the ¹⁴C of 2.5-kyr BP and 22-kyr BP tree rings using the MALT AMS. They have confirmed that the ¹⁴C ages of multiple graphite targets prepared from the same sample under the same conditions show a Gaussian distribution with a standard deviation equivalent to the average of the statistical (Poisson) error of each ¹⁴C measurement. This suggests that the greater part of AMS precision is determined by the counting statistics, and they thus employed the weighted average ¹⁴C ages of the multiple graphite targets to improve the ¹⁴C age accuracy for each sample.

Because we have also used the MALT system, the same methodology can be applied to our results. The weighted averages of the ¹⁴C ages for the same ring numbers of stumps A and B (Table 1) show excellent agreement within a small error value (~40 yr). This again confirms the high reproducibility of our analysis and the remarkably good sample preservation.

Table 1 ^{14}C data on 2 coniferous trees from the Towada Hachinohe buried forest.

Sample	Ring #	Turn #	Cathode #	^{14}C age (yr BP)	Average (yr BP)	Lab#
A-1	1-5	1	1	$13,113 \pm 54$	$13,135 \pm 40$	MTC-03732
			2	$13,161 \pm 58$		MTC-03733
B-1	1-5	1	1	$13,129 \pm 58$	$13,140 \pm 41$	MTC-03736
			1	$13,148 \pm 77$		MTC-04126
			2	$13,154 \pm 84$		MTC-04127
A-2	1-40	1	1	$13,203 \pm 59$	$13,188 \pm 42$	MTC-03734
			2	$13,172 \pm 60$		MTC-03735
B-2	1-40	1	1	$13,159 \pm 56$	$13,167 \pm 30$	MTC-03738
			2	$13,127 \pm 56$		MTC-03739
		2	1	$13,227 \pm 64$		MTC-04128
			2	$13,171 \pm 66$		MTC-04129
B-3	41-80	1	1	$13,039 \pm 63$	$13,155 \pm 33$	MTC-03740
			2	$13,160 \pm 58$		MTC-03741
		2	1	$13,205 \pm 75$		MTC-04130
			2	$13,272 \pm 77$		MTC-04131
A-4	81-120	2	1	$13,137 \pm 71$	$13,055 \pm 50$	MTC-04122
			2	$12,976 \pm 70$		MTC-04123
B-4	81-120	1	1	$13,027 \pm 56$	$13,047 \pm 34$	MTC-03742
			2	$13,005 \pm 55$		MTC-03743
		2	1	$13,147 \pm 71$		MTC-04133
A-5	121-140	2	1	$13,117 \pm 73$	$13,130 \pm 49$	MTC-04124
			2	$13,140 \pm 67$		MTC-04125
B-5	121-160	1	1	$13,147 \pm 61$	$13,176 \pm 35$	MTC-03744
			2	$13,165 \pm 57$		MTC-03745
			1	$13,224 \pm 66$		MTC-04134

Figure 2 shows the weighted averages of the ^{14}C ages plotted against the ring numbers. It is clear that the ^{14}C ages of the 2 stumps agree and also fluctuate concordantly throughout the total sample interval of about 150 yr. The fluctuation probably reflects a significant change in atmospheric ^{14}C concentration during this period, which may be discussed in the context of the $\Delta^{14}\text{C}$ profiles.

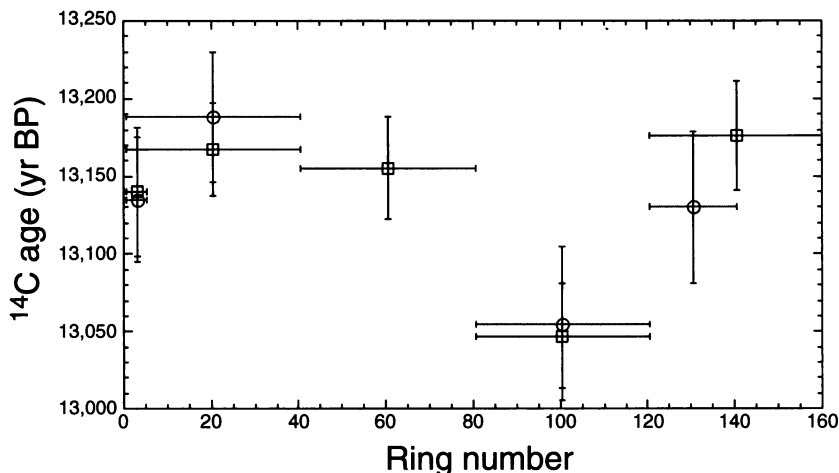


Figure 2 Weighted average ^{14}C ages plotted against ring number. The ^{14}C ages for each specimen agree and fluctuate concordantly throughout the total interval (circles: stump A; squares: stump B).

Since we have had no independent chronology for stumps A and B, a wiggle-matching technique is needed to obtain the $\Delta^{14}\text{C}$ profiles. However, high-resolution ^{14}C profiles for ~ 15.5 cal kyr BP, to serve as a target for the wiggle-matching, have not been published. Only a few varve and tree-ring data (Hughen et al. 2000, 2004b; Kromer et al. 2004) of decadal resolution reach 15 cal kyr BP. We therefore derived a calendar age for the outermost ring using the weighted average of all ^{14}C ages for the outermost 5 rings ($13,133 \pm 33$ ^{14}C yr BP) and the IntCal04 curve. The age obtained was 15,512 cal BP, which is the mean of the 1- σ range (15,363–15,679 cal BP) using CALIB v 5.0.1 (Stuiver and Reimer 1993). Although this is tentative, the estimation of $\Delta^{14}\text{C}$ values using this age supports our conclusions because only the extent of the fluctuation, not its values, is essential for the following discussion.

The estimated $\Delta^{14}\text{C}$ values vary from 265 to 300‰ and show an approximately sinusoidal fluctuation of a presumably ~ 200 -yr cycle (Figure 3). The possible causes of this fluctuation are changes in ocean circulation, geomagnetic field intensity, and solar activity. Ocean circulation variation could strongly influence atmospheric ^{14}C (e.g. Hughen et al. 2000), whereas periods of ^{14}C fluctuation resulting from geomagnetic field intensity are generally longer than century scale (e.g. Stuiver et al. 1991). Considering the nature of the fluctuation, we believe solar activity to be the most likely cause. A 30–40‰ change in $\Delta^{14}\text{C}$ is comparable to the change attributed to solar activity with a ~ 200 -yr cycle during the last millennium (Stuiver and Quay 1980; Reimer et al. 2004). Moreover, such 30–40‰ century-scale fluctuations taking place after 15 cal kyr BP can be detected in the last-glacial ^{14}C tree-ring record (Kromer et al. 2004) and in the varved sediment of Suigetsu Lake (Kitagawa and van der Plicht 2000) and the Cariaco Basin (Hughen et al. 2004b). The scattered nature of these profiles, which might be partly owing to the precision of the ^{14}C measurement and/or counting age, would be smoothed by our ~ 40 -yr sampling. This implies that the ^{14}C fluctuation around 15.5 cal kyr BP reflects a continuous ~ 200 -yr cycle of past solar activity change. However, this interpretation is based on a few data of coarse resolution, and further analysis would be necessary to really determine if the cycle is present.

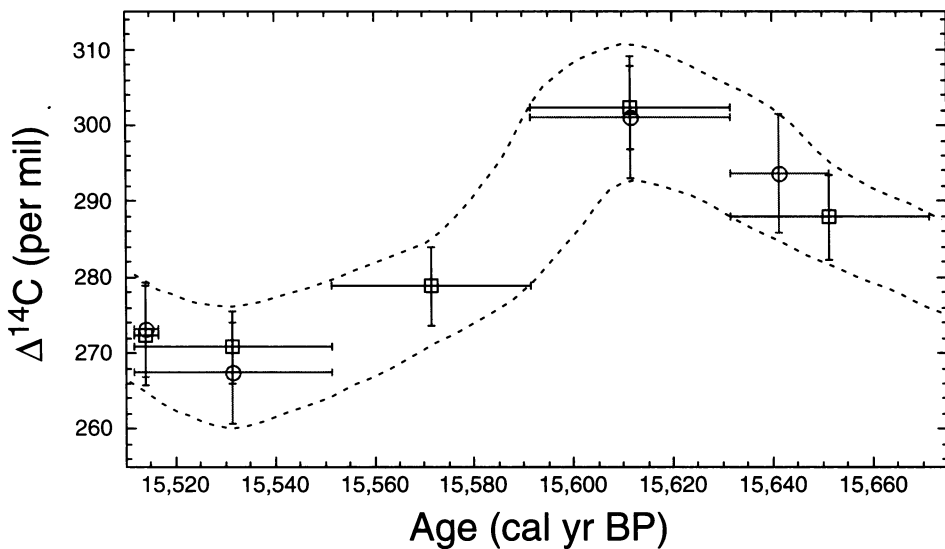


Figure 3 Estimated $\Delta^{14}\text{C}$ profile for ~ 15.5 cal kyr BP. The calendar age of the outermost rings of stumps A and B is assumed to be 15,512 cal BP (see text). The ^{14}C values show an approximately 30–40‰ amplitude sinusoidal curve and an indicated ~ 200 -yr cycle, possibly reflecting contemporary solar activity changes. Dotted lines show the approximate range of the sinusoidal curve (circles: stump A; squares: stump B).

IMPLICATIONS FOR ^{14}C CALIBRATION CURVE AND POSSIBLE IMPACT OF THE ERUPTION

Although we used the IntCal04 curve (Reimer et al. 2004) for our $\Delta^{14}\text{C}$ estimation, it is unclear whether our results agree with certain ^{14}C records of this period or not. Figure 4 shows the weighted averages of the ^{14}C ages of stumps A and B plotted against calendar age in relation to the IntCal04 curve. Our data is mostly in the $1\text{-}\sigma$ error range of IntCal04 and also close to the nearest calibration point presented by Bard et al. (1998). However, the detailed similarity and/or difference between our data and the atmospheric ^{14}C standard curve is unclear. That is partly because of the very coarse resolution of coral ^{14}C records used for constructing IntCal04 before 14.7 cal kyr BP; there are only 3 calibration points between 15 and 16.5 cal kyr BP. An alternative standard record other than the IntCal series may be the Cariaco Basin ^{14}C data (Hughen et al. 2004a). This record has higher time resolution during the target period than others such as the Lake Suigetsu ^{14}C record (Kitagawa and van der Plicht 1998, 2000). The Cariaco record is corrected with a marine reservoir age of 420 yr for constructing an atmospheric ^{14}C curve, and is directly connected to the annual-layer chronology of the GISP2 ice core from Greenland (Meese et al. 1994, 1997).

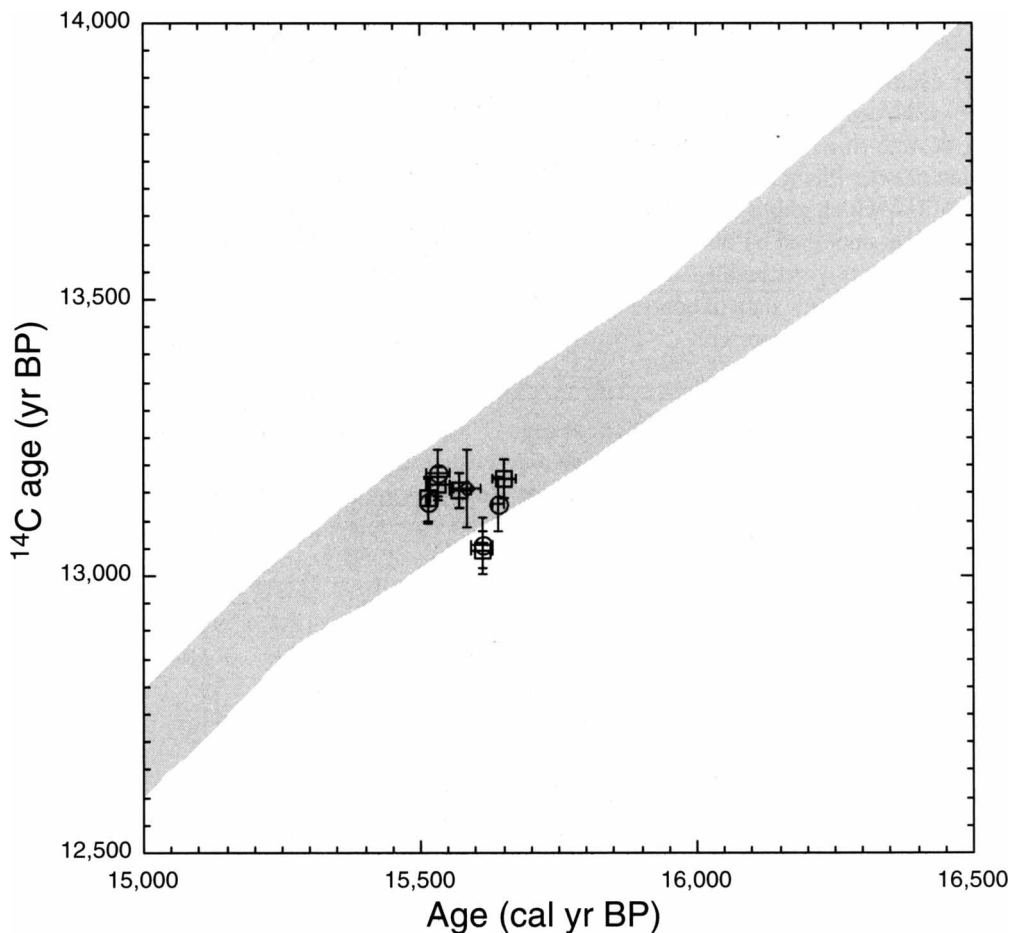


Figure 4 Comparison between our tree-ring data and the IntCal04 record (Reimer et al. 2004). The shaded area shows the $1\text{-}\sigma$ range of the IntCal04 curve. The age of the outermost ring is initially set to 15,512 cal BP (see text). The nearest calibration point provided by the Mururoa coral record (Bard et al. 1998) for IntCal04 is also shown here (diamond). Circles: stump A; squares: stump B.

Figure 5a shows the Cariaco Basin record and our tree-ring data, in which the age of the outermost ring is set at the initial value of 15,512 cal BP. Because of the large uncertainty of the calendar age of the Cariaco record due to multiplying the errors of both the correlation procedure and the GISP2 chronology, our data are clearly within the error range of the Cariaco record. On the other hand, using the raw Cariaco/GISP2 age model without any errors on the calendar age estimation, we may have the following different “best match” between our tree-ring and the Cariaco records. The simplest solution is probably that shown in Figure 5b: the calendar age of the outermost ring is shifted about +50 yr from the initial value, and thus the age of the outermost ring becomes 15,560 GISP2 yr BP. In this case, we can follow the original 420-yr reservoir correction presented in Hughen et al. (2000) and there is only small discrepancy between the 2 profiles. Another somewhat drastic but better “best match” solution may be the case shown in Figure 5c: the calendar age of the outermost ring is shifted about +680 yr from the initial value, and thus the age of the outermost ring becomes 16,190 GISP2 yr BP. In this case, we have to use a 650-yr reservoir age for the Cariaco Basin record. Kromer et al. (2004) correlated their floating tree-ring ¹⁴C data with the high-resolution, varve-chronology-based Cariaco ¹⁴C record (Hughen et al. 2000, 2004b) of 12.8–14.2 cal kyr BP and showed that the best match appears in an estimation of a 650-yr reservoir age before the Younger Dryas period, instead of a 420-yr reservoir age since that time. If the latter case of our best match is correct, our data suggest that a 650-yr reservoir age of the Cariaco Basin is applicable at least from 16 cal kyr BP. Further high-resolution analyses of both our tree rings and the Cariaco Basin record are crucial for answering this interesting question and for precise dating of the Towada Hachinohe buried forest by using the wiggle-matching technique.

The eruption of ToH (and ToHP), which formed the Towada Hachinohe buried forest, is the largest volcanic event known of the last deglaciation stage (11.5–19 kyr BP) in Japan (Machida and Arai 2003). The Volcanic Explosivity Index (VEI) of this eruption is estimated at 6.7 (Hayakawa 1985). The precise chronology of this eruption is therefore quite important for studies of regional archaeology and Quaternary geology and perhaps for knowing the global impact of the huge eruption. Zielinski et al. (1996) showed the record of explosive volcanism from the GISP2 ice core using volcanic SO₄²⁻ signals (EOF-5 values >>75 ppb) with the Meese et al. (1997) age model. From 15,400 to 16,400 GISP2 yr BP, 9 signals are found in their SO₄²⁻ profile. Around the horizons (±100 yr) corresponding to the estimated GISP2 ages of the outermost ring (15,560 and 16,190 BP), they extracted 4 peaks: a 103-ppb peak at 15,603 BP, a 111-ppb peak at 15,611 BP, a 75-ppb peak at 16,131 BP, and a 314-ppb peak at 16,191 BP. At present, we have no additional data for a detailed discussion on the origins of those signals. However, further ¹⁴C analysis of tree rings from the Towada Hachinohe buried forest and comprehensive consideration of the atmospheric transportation and the preservation process of volcanic SO₄²⁻ signals in ice cores would help to make clear the question of whether a ToH volcanic signal is really recorded in the Greenland ice core, as well as the global impact of this eruption.

CONCLUSION

The ¹⁴C profiles for ~15.5 cal kyr BP obtained from the study of 2 fossil coniferous trees, both probably killed at the same time, show excellent agreement. This cross-check of ¹⁴C data confirms the high reproducibility of our analysis and negligible postburial carbon contamination. The Δ¹⁴C values estimated by using an IntCal04 calibrated age (15,512 cal BP) of the outermost ring vary from 265 to 300‰ and illustrate an approximately sinusoidal curve of 30–40‰ amplitude and an indicated ~200-yr cycle. This ¹⁴C fluctuation can be interpreted as reflecting past solar activity change. Comparison of the obtained ¹⁴C profiles and the Cariaco Basin ¹⁴C record suggests that the “best match” would be found in the case that the age of the outermost ring is 15,560 or 16,190 GISP2 yr

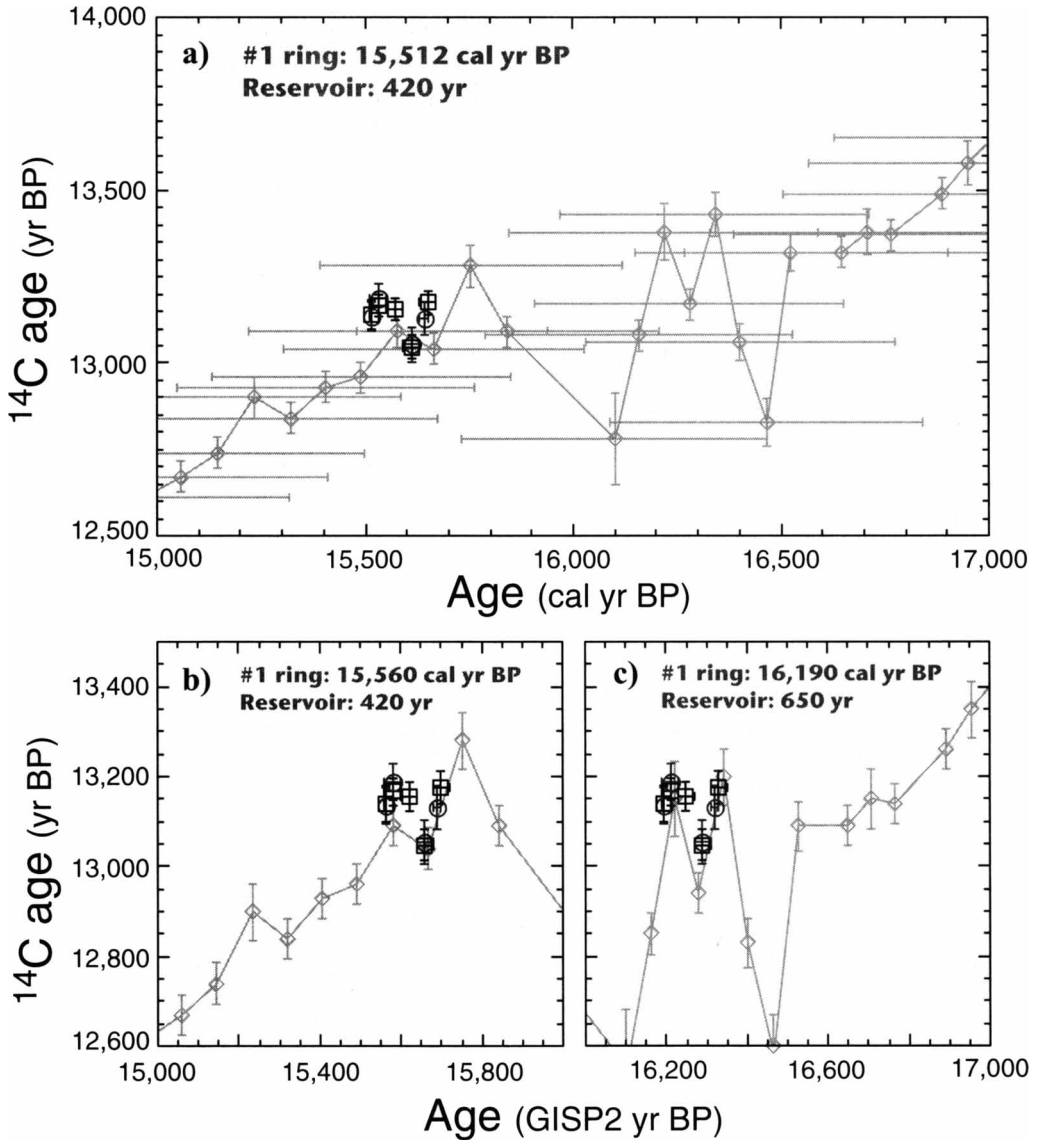


Figure 5 Comparison between our tree-ring data and the Cariaco Basin ¹⁴C record (Hughen et al. 2004a): circles: stump A; squares: stump B; diamonds: Cariaco Basin. a) The calendar age of the outermost ring is set at 15,512 cal BP and the reservoir age of the Cariaco record is 420 yr (the original value). The uncertainty of the calendar age of the Cariaco Basin record is estimated by multiplication of the errors of both the correlation procedure and GISP2 chronology (Hughen et al. 2004a). b) The age of the outermost ring is 15,560 cal BP and the reservoir age of the Cariaco record is 420 yr. c) The age of the outermost ring is 16,190 cal BP and the reservoir age of the Cariaco record is 650 yr. An estimated error of ±180 yr for the correlation procedure between Cariaco and GISP2 (Hughen et al. 2004a) is not shown in (b) and (c).

BP; for the latter case, a 650-yr reservoir correction of the Cariaco Basin record is necessary. Such an attempt, with the help of ice-core records of volcanic signals and further ¹⁴C analysis of the tree rings, would make clear the accurate age of the Towada Hachinohe buried forest and the global impact produced by the eruption.

Although this study has obtained a reasonable ¹⁴C profile, the analytical method may allow improvement. We are now investigating alternative pretreatment procedures such as Soxhlet extraction and/or delignification. This is an attempt to determine the single-year ¹⁴C atmospheric concentration based on 2 coniferous tree samples from the last glacial period. Our work may help in the study of ice age solar activity using cosmogenic nuclides from both tree rings (¹⁴C) and ice cores (¹⁰Be and ³⁶Cl).

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