[14]C AGES OF 43 CONSECUTIVE SINGLE-YEAR TREE RINGS BETWEEN 2710 AND 2655 CAL BP USING ACCELERATOR MASS SPECTROMETRY

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ABSTRACT. We have measured the radiocarbon ages of 43 consecutive single-year tree rings using accelerator mass spectrometry (AMS) with a statistical accuracy of ~2.3‰. AMS ^{14}C ages of the 36 viable samples are between 2708 and 2666 cal BP, a period in which the $\Delta^{14}C$ of the IntCal04 curve (Reimer et al. 2004) shows an enhancement. The ^{14}C ages of the samples are scattered with a Gaussian distribution around the interpolated IntCal04 calibration curve. The time profile of the deviations of the 36 ^{14}C ages from the interpolated IntCal04 calibration curve indicates a linear trend and a characteristic variability rather than a random fluctuation around the curve. The trend indicates a higher gradient than that of the interpolated IntCal04 calibration curve. The profile implies a periodic variation of approximately 11 yr and an amplitude of roughly 18 ^{14}C yr.

INTRODUCTION

The number of sunspots is an indicator of solar activity. Observations of sunspot numbers since the beginning of the 17th century typically show a cyclical variation with a period of 11 yr (Hoyt and Schatten 1998). We expect that the 11-yr periodicity with an antiphase appears in radiocarbon concentrations in tree rings because ^{14}C is produced in the atmosphere by cosmic rays, which are affected by solar magnetic fields in the heliosphere, and hence by solar activities. In fact, during the Maunder minimum, when there were few sunspots, the atmospheric ^{14}C concentration at its peak was 20‰ greater than of modern carbon (Reimer et al. 2004). Moreover, the excess ^{14}C concentration in such eras as the Maunder minimum might result in the 11-yr cycles being prolonged to 13 to 15 yr (Miyahara et al. 2004). Since during the past 10,000 yr there have been several eras of excess ^{14}C concentration similar to the Maunder minimum, it is important to test the 11-yr periodicity with respect to durations of high amplitude in peak concentration in different time periods in order to study solar activity over long intervals.

To investigate the time variation of the ^{14}C concentration in an excess era with a resolution of 1 yr, we measured the ^{14}C ages of 43 consecutive single-year tree rings between 2710 and 2655 cal BP using accelerator mass spectrometry (AMS). The tree rings are taken from an old cedar in Japan (39°05′N, 140°03′E), the Choukai Jindai cedar, and range in age from 2757 to 2437 cal BP (Sakurai et al. 2006). As shown in Figure 1, the 43 single-year tree rings are located at the peak area of an excess stage in the IntCal04 $\Delta^{14}C$ curve (Reimer et al. 2004). We compare the ^{14}C ages of the 43 single-year data to the IntCal04 calibration data in 5-yr spans, as well as the time profile of the 43 single-year ^{14}C ages.

MEASUREMENTS

The 43 consecutive tree ring samples were taken from near the center of the Choukai Jindai cedar. The tree rings are labeled from the center (MY46) to the outside (MY4). They were carefully separated into 43 single-year tree ring samples by hand using tweezers. For each sample, α-cellulose was extracted. Approximately 50 mg of α-cellulose was extracted from 500 mg of wood sample for each single-year tree ring. As the target material of the accelerator, graphite samples weighing between

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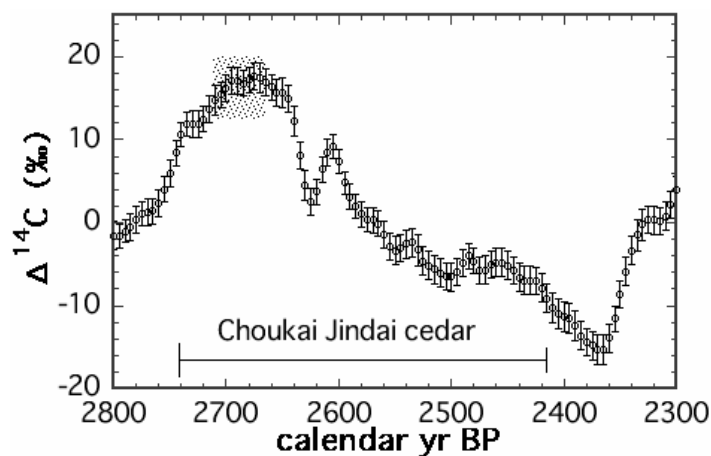[3]Research Center for Nuclear Science and Technology, University of Tokyo, Tokyo 113-0032, Japan.

Figure 1 Locations of the 43 single-year tree ring mesh and the Choukai Jindai
cedar in the IntCal04 curve.

0.3 and 1.3 mg were produced from each α-cellulose sample. Because it was difficult to obtain a sufficient quantity of graphite for 7 of the single-year tree rings, we only prepared 36 graphite samples for AMS ^{14}C measurement.

AMS measurements were carried out using the tandem accelerator at the Micro Analysis Laboratory, University of Tokyo (MALT) (Gandou et al. 2004). An injected current of ^{12}C was maintained between 40 and 80 µA. In ^{14}C measurements at MALT, the errors other than statistical errors are negligible (5‰) (Matsuzaki et al. 2004). The average ^{14}C count for samples was about 1.9×10^5 counts, and hence the statistical accuracy was ~2.3‰. The ^{14}C age of each graphite sample was calculated using the measured value of the ^{14}C/^{12}C ratio for the sample and the National Institute of Standards and Technology (NIST) standard samples.

The 36 samples were divided into 2 sets of tree rings labeled by odd and even numbers, and each set was separately measured using 2 different machine time schedules. As shown in Figure 2, the ^{14}C ages are distributed between 2411 and 2542 ^{14}C yr BP (with a statistical error of 22 ^{14}C yr BP) and show a linear trend. The ^{14}C ages and the errors are shown in Table 1 for each tree-ring sample. The average ^{14}C ages are 2477 ± 9 and 2470 ± 7 ^{14}C yr BP for the odd and even sets, respectively. By using a least-squares fitting with a linear function, the gradient is calculated as 1.7 ± 0.4 for the 36 samples with a correlation coefficient of 0.6; the gradients are 2.2 ± 0.6 and 1.4 ± 0.5 for the odd and even data sets, respectively. Because both the gradients and the averages for the odd and even data sets are comparable to each other within the error in the different machine times, it is presumed that both the measurements were carried out under the same conditions. Therefore, the results indicate that both the data sets can be reliably combined to form a data set we call the Ch data set.

RESULTS AND DISCUSSION

The Ch data set is compared with the IntCal04 calibration curve interpolated at 1-yr intervals because the IntCal04 data set is for 5-yr spans, although the Ch data set is for single years. Here, the interpolation was carried out using a cubic function as well as by using the program OxCal v 3.10 (Bronk Ramsey 1995, 2001). Using the difference $\Delta T = T_s - T_{int}$, where T_s and T_{int} are the ^{14}C ages of the Ch data set and the interpolated IntCal04 data set, respectively, the quality of fit to the IntCal04 data set was investigated. Since the calendar age of the Choukai Jindai cedar was deter-
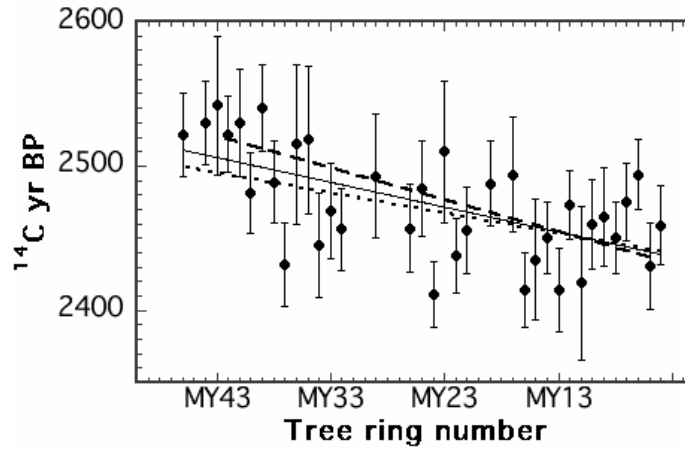
Figure 2 Least-squares fittings with a linear function for the 36 14C ages. The thin solid line is for the 36 samples. The dashed line and the dotted line are for the data sets of odd and even tree ring numbers, respectively.

Table 1 14C ages and the errors for 36 tree-ring samples.

Sample nr	14C yr BP	Error	Sample nr	14C yr BP	Error
MY4	2459	27	MY24	2411	23
MY5	2431	30	MY25	2484	33
MY6	2494	24	MY26	2457	30
MY7	2475	27	MY29	2493	43
MY8	2450	25	MY32	2456	28
MY9	2465	34	MY33	2469	33
MY10	2460	31	MY34	2445	36
MY11	2419	53	MY35	2518	51
MY12	2473	24	MY36	2515	55
MY13	2414	29	MY37	2432	29
MY14	2450	25	MY38	2489	28
MY15	2435	42	MY39	2540	30
MY16	2414	26	MY40	2481	28
MY17	2494	40	MY41	2530	37
MY19	2488	29	MY42	2522	26
MY21	2455	30	MY43	2542	48
MY22	2438	26	MY44	2530	29
MY23	2510	49	MY46	2522	29

mined with a statistical accuracy of ±12.5 cal BP (95.4% confidence level) by wiggle-matching (Sakurai et al. 2006), the estimated calendar age of sample MY4 is 2666 ± 12.5 cal BP. As shown in Figure 3, we checked the calendar age of 2666 cal BP for MY4, compared with the weighted average of each $|\Delta T|$ of the Ch data set when the calendar age of MY4 varies around 2666 cal BP. Although at 2661 cal BP the average value of $|<\Delta T>|$ was at its minimum, it was comparable to the value at 2666 cal BP within the statistical error. Hence, we define the calendar age to be 2666 cal BP for MY4, and thus the Ch data set ranges from 2708 to 2666 cal BP.
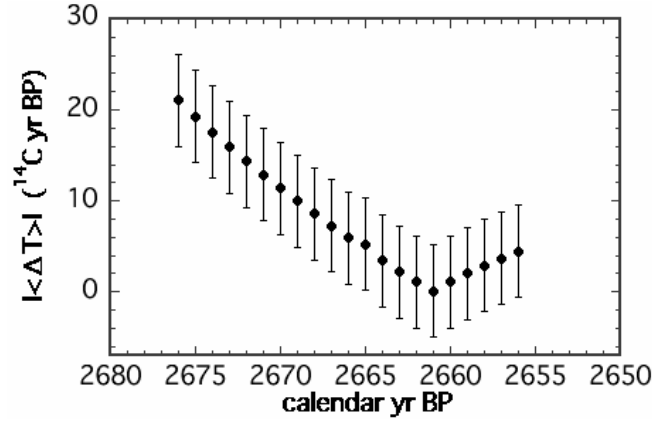
Figure 3 Absolute value of the average of each Δ*T* for the Ch data set when
the calendar age of the MY4 varies between 2656 and 2676 cal BP (see text).

Figure 4 shows a time profile of the Ch data set with the IntCal04 data and the interpolation as a
function of the calendar year. The figure shows that the 36 data points are scattered around the
IntCal04 interpolated curve. The distribution of

$$\Delta Z = \frac{\Delta T}{\sigma_s}$$

for the 36 samples, where σ_s is the statistical error of T_s, is shown using a histogram and a best-fitted
Gaussian curve in Figure 5. The average and the standard deviation of ΔZ are –0.01 and 1.08,
respectively. From the 0.43 value (degrees of freedom [dof] = 4) resulting from a reduced χ^2 test,
which evaluates the quality of fit of the Gaussian distribution to the histogram, the 36 ^{14}C ages of the
Ch data set are approximately distributed around the interpolated IntCal04 calibration curve with a
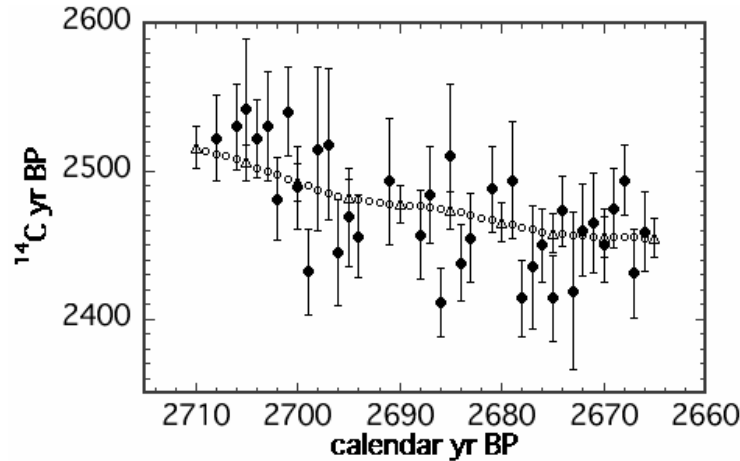Gaussian distribution.

Figure 4 Time profile of the Ch data set (closed circles) with the IntCal04 data (open
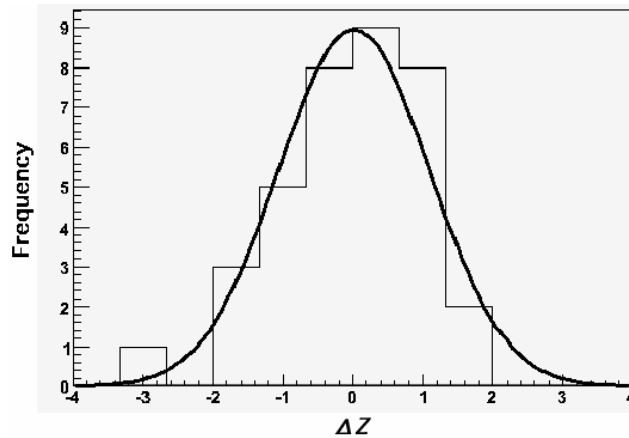triangles) and the interpolation curve (open circles).

Figure 5 Histogram and best-fitted Gaussian distribution curve for ΔZ

Finally, we investigate the time profile of ΔT of the Ch data set by a 5-point moving average, as shown in Figure 6. In the figure, the points with fewer than 5 points for calculation of the moving average are averaged by the smaller number of points, and the error bars show the errors for the moving average. The profile indicates a trend and a characteristic variability rather than a random fluctuation around the interpolated IntCal04 calibration curve corresponding to the line $\Delta T = 0$ in the figure. The quadratic curve in the figure with a reduced χ^2 value of 0.82 (dof = 33) is calculated by a least-squares method for the data of ΔT, not for the moving average data. The curve shows the trend of the residual of the Ch data set from the interpolated IntCal04 calibration curve. It indicates that the left, middle, and right parts are older, younger, and of equal age, respectively, compared with IntCal04. Hence, the trend of the Ch data set more rapidly decreases to the middle point and then increases slowly, compared with the IntCal04 curve. Although the minimum point at 2683 cal BP is consistent with the center of the excess peak of the $\Delta^{14}C$ curve in IntCal04, the ^{14}C age is ~13 ^{14}C yr younger than that of the IntCal04 data. Moreover, the connected line of the moving average points indicates a periodic variation of ~11 yr and an amplitude of ~18 ^{14}C yr.
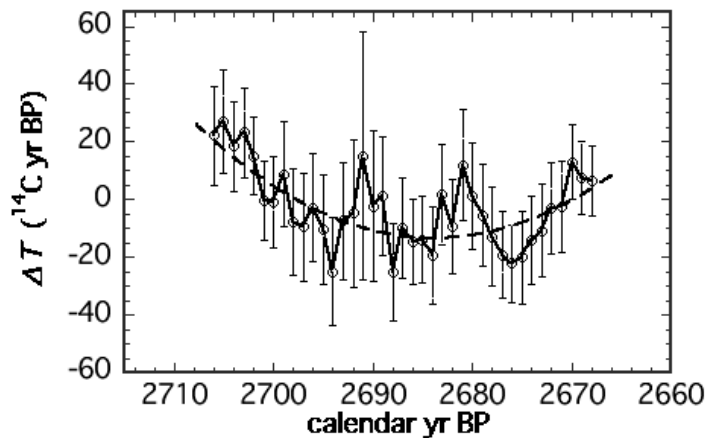

Figure 6 Time profile of ΔT of the Ch data set by 5-point moving average and the quadratic curve of ΔT obtained by a least-squares fitting.

The variation of cosmic rays caused by the 11-yr solar cycle is strongly suppressed by global carbon circulation (Siegenthaler et al. 1980). The neutron variation of secondary cosmic rays has been reported since 1952 as the observational data of cosmic rays (http://ulysses.sr.unh.edu/NeutronMonitor/neutron_mon.html), which indicates an 11-yr variation of about 20–30% and is consequently reflected in the ^{14}C concentration as a variation of only 2–3‰. The amplitude of 18 ^{14}C yr corresponds to 2.3‰, and this might be due to the variation of the flux of cosmic rays during the 11-yr solar cycle.

CONCLUSION

Forty-three consecutive tree rings were sampled from the center portion of the Choukai Jindai cedar and dates ranged from 2757 to 2437 cal BP. ^{14}C ages of the 36 single-year tree rings were measured by AMS with a statistical accuracy of ~2.3‰ to investigate short-term variations of ^{14}C in periods of excess ^{14}C production 2360 yr before the Maunder minimum. The ^{14}C ages are distributed between 2411 and 2542 ^{14}C yr BP (with a statistical error of 22 ^{14}C yr BP) and indicate a linear trend.

Comparison of the Ch data set of 36 ^{14}C ages with the interpolated IntCal04 calibration curve interpolated at 1-yr intervals indicates that the Ch data set is located between 2708 and 2666 cal BP. The Ch data set is distributed around the interpolated IntCal04 calibration curve with a Gaussian distribution. The time profile of the deviations of the Ch data from the interpolated IntCal04 calibration curve indicates a trend and a characteristic variability rather than a random fluctuation around the curve. The trend has a higher gradient than that of the interpolated IntCal04 calibration curve. Moreover, the profile implies a variation of ^{14}C with a period of ~11 yr and an amplitude of ~18 ^{14}C yr.

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