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PRECISE COMPARISON OF ^{14}C AGES FROM CHOUKAI JINDAI CEDAR WITH INTCAL04 RAW DATA

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ABSTRACT. We measured the radiocarbon ages of 165 single-year tree rings from a Japanese Choukai Jindai cedar using accelerator mass spectrometry (AMS). By wiggle-matching the Choukai_AMS data set to the IntCal04 calibration data using OxCal v 3.10 and using the variation of the correlation coefficients between the Choukai_AMS and IntCal04 data sets, we precisely re-estimated that the 321 Choukai Jindai cedar tree rings range from 780 to 460 cal BC with an accuracy of 8 yr. The Choukai_AMS data set is older than the 3 raw data sets of European tree rings that comprise IntCal04. The Belfast and Seattle data sets are younger by –21.3 ± 5.5 and –22.7 ± 5.6 ^{14}C yr, respectively. The Choukai Jindai cedar is ~22 ^{14}C yr older than the European tree rings, which is equivalent to an offset of –2.8‰ in ^{14}C. In addition, the Choukai_AMS data set correlates well with the Belfast and Seattle data sets, with correlation coefficients of 0.89 and 0.68, respectively, between the temporal profiles. Hence, the temporal profile of the Choukai ^{14}C ages shows a global variation.

INTRODUCTION

The radiocarbon ages for 800–400 BC have an approximately constant value, indicating that they compose a characteristic era over the past 10,000 yr as shown in the IntCal04 calibration curve (Reimer et al. 2004). This profile corresponds to the ^{14}C concentrations in the vicinity of the period, which vary on a large scale as evidenced by the IntCal04 data set. Since ^{14}C concentrations in the atmosphere are basically determined by production rates related to cosmic rays and geomagnetic fields as well as the global environmental conditions in terms of the carbon circulation between the atmosphere and ocean, ^{14}C ages should decrease at a constant rate when these conditions are kept in equilibrium. Therefore, the ^{14}C ages for 800–400 BC, with their unlikely equability, bring valuable information about the solar modulation of cosmic rays and the climate due to the Sun-Earth connection during that period.

The Choukai Jindai cedar was buried in clay at Mt Choukai (39º05′N, 140º03′E) in Japan. The burial is considered to be the result of a sector collapse of the mountain 2700–2400 yr ago (Inokuchi 1988). The wood sample has more than 320 tree rings. Measuring 21 discrete samples over a range of 230 tree rings, the calendar age of the outer ring was assigned to 477.5 cal BC (±12.5 yr) by a wiggle-matching method employing the IntCal04 curve (Sakurai et al. 2006). As the Choukai Jindai cedar is located between 797 and 477 cal BC according to the calibration, the tree rings are adequate for investigating the variation of the ^{14}C concentrations for the characteristic era.

The ^{14}C ages of 165 single-year tree rings of the Choukai Jindai cedar were measured at 741–462 cal BC using accelerator mass spectrometry (AMS). Although the measurement was intended to observe the variation in ^{14}C concentrations as it relates to the 11-yr solar cycle for the era, for this paper we focused on a precise comparison of the ^{14}C ages between Japanese and European tree rings using the data set of the Choukai Jindai cedar. It is important to investigate the uniformity in the Northern Hemisphere of the variation in ^{14}C ages appearing on the IntCal04 data set, which was compiled using the raw data sets of European tree rings; this is because regional differences, if they are positive, can be used to connect estimations of climate change compared to the present time.

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Moreover, for Japanese archaeological research, fine-tuning the ^{14}C ages in this era is a key point for evaluating Yayoi and pre-Yayoi cultures on the historical timescale (Ozaki et al. 2007).

MEASUREMENTS

The 321 tree rings of the Choukai Jindai cedar used this study are numbered starting from a base tree ring of Y0. The tree rings are numbered from Y1 to Y235 toward the outer edge and from MY1 to MY85 toward the center portion. In total, using AMS we measured 165 tree rings consisting of 53 consecutive tree rings from MY46 to Y9 and 112 alternate tree rings from Y11 to Y233. Although there are 56 tree rings in the interval of MY46–Y9, for the 3 tree rings MY31, MY30, and MY27, we could not obtain ^{14}C data because the rings were too thin to peel.

The tree rings were taken from a block of boiled wood using tweezers. Each sample was milled prior to chemical treatment. We chemically extracted α-cellulose from the cell walls because it is the most reliable chemical component for measuring the annual concentration of ^{14}C. The α-cellulose yield from the wood was ~20% by weight. Graphite samples were produced by burning 3 mg of α-cellulose. The weight ratios of the graphite to 1 mg of iron powder were kept between 0.6 and 1.2 (Gandou et al. 2004).

Measurements of ^{14}C in the graphite samples were carried out using a 5MV tandem accelerator at the University of Tokyo (Micro Analysis Laboratory, Tandem Accelerator the University of Tokyo, MALT). The ^{14}C dates for the graphite samples were calculated using the measured values of the $^{14}C/^{12}C$ ratio for the sample and 4990C oxalic acid, which is a standard sample. The measured $^{14}C/^{12}C$ ratio for a blank sample using IAEA-C1 was typically 1.0×10^{-15}.

The ^{14}C ages for 165 single-year tree rings between MY46 and Y233, known as the Choukai_AMS data set, are shown in Figure 1 as a function of the tree-ring number; the counting error was typically 30 ^{14}C yr.

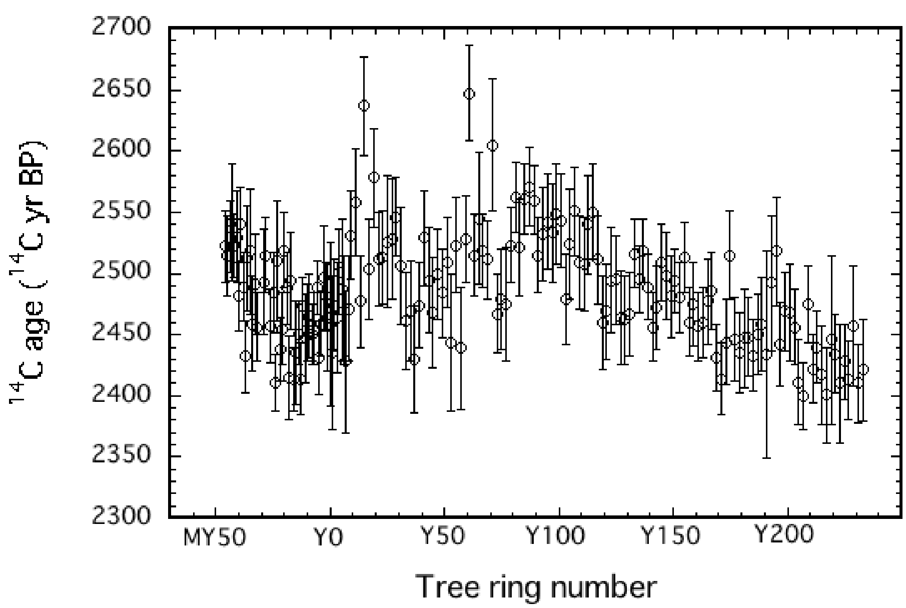

Figure 1 ^{14}C ages of 165 single-year tree rings in the Choukai Jindai cedar measured by AMS

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RESULTS AND DISCUSSION

Fine ^{14}C Dating of the Choukai Jindai Cedar

First, we performed a precise re-estimation of the calendar age for the Choukai Jindai cedar using the Choukai_AMS data set of 165 single-year ^{14}C ages over the span of 280 yr. The ^{14}C ages of the Choukai_AMS data set were calibrated using OxCal v 3.10 (Bronk Ramsey 1995, 2001; Bronk Ramsey et al. 2001) with the wiggle-matching for the IntCal04 calibration data (Reimer et al. 2004). The calculated calendar age of Y233 is 470–455 cal BC (2419–2404 cal BP) with a 95.4% confidence level, as shown in Figure 2. The previous age estimate of the Choukai Jindai cedar for Y233 was 492–467 cal BC, which was calculated using the 21 ^{14}C ages between Y0 and Y230 and measured by the liquid scintillation counting (LSC) system (Sakurai et al. 2006). Although both ages partially overlap, the width of this new age estimation was narrowed to 16 yr from 26 yr because the number of data points and the range covered by the Choukai_AMS data set are greater than those for the previous estimation.

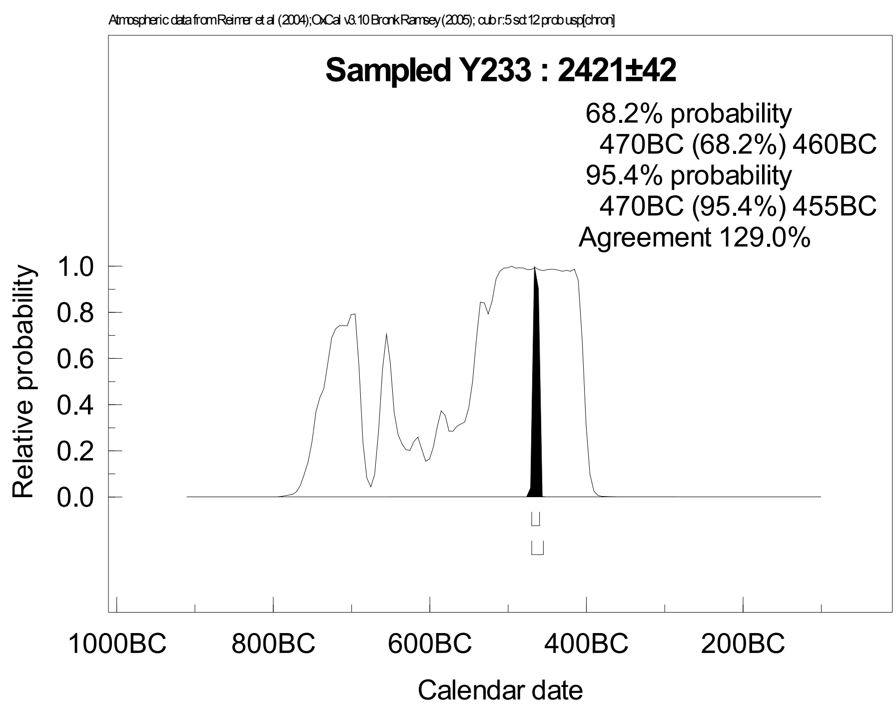

Figure 2 Most probable calendar date of tree-ring number Y233 calculated by OxCal v 3.10 for the IntCal04 calibration curve; 470–455 cal BC is equivalent to 2419–2404 cal BP.

Using the advantage of the Choukai_AMS data set—which consists of single-year data compared to the 5-yr data for the IntCal04 data set—we attempted to narrow down the age width estimation by wiggle-matching. The Choukai_AMS data set allows weighted average data sequences for intervals of 5 yr and assigned the calendar age of Y233 to within 470–455 cal BC. Hence, the correlation coefficient between the Choukai_AMS and IntCal04 data sets when assigning Y233 became an indicator for the correlation between both data sets in general. Figure 3 shows the correlation coefficients as a function of the calendar age for Y233. As shown in the figure, the correlation coeffi-

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cients functionally vary with a maximal portion. A least-squares-fitted quadratic function indicates a maximum at 462 cal BC, and the correlation coefficients were greater than 0.8 between 465 and 458 cal BC. From these calculations, the most probable calendar age for Y233 was estimated to be 462 cal BC, and the 321 tree rings of the Choukai Jindai cedar range from 780 to 460 cal BC.

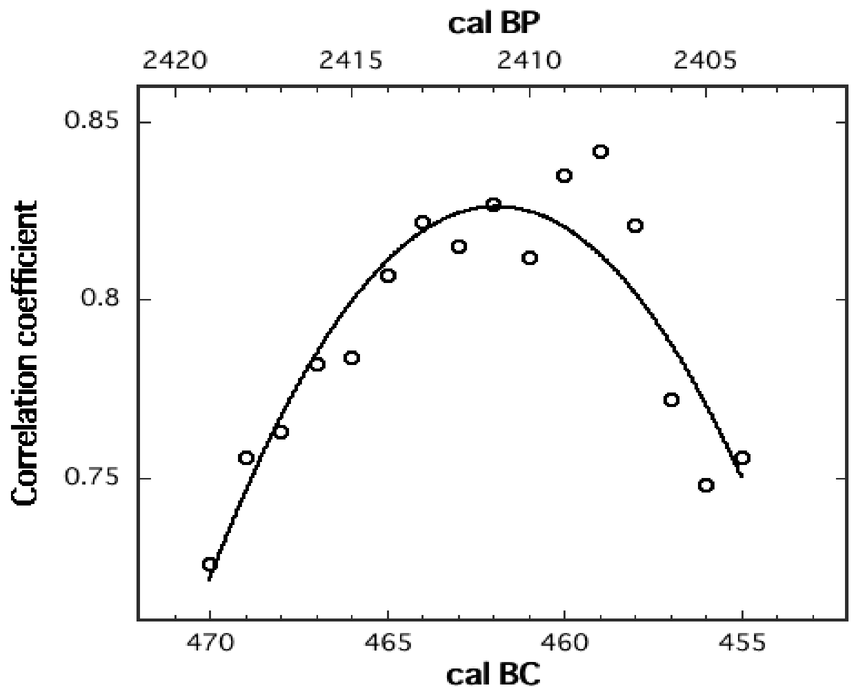

Figure 3 Most probable calendar date of Y233 calculated by the correlation coefficient (open circles) between the Choukai_AMS and IntCal04 data sets. A least-squares-fitted quadratic function (solid line) indicates a maximum at 462 cal BC (2411 cal BP) for Y233.

As mentioned previously (Mitsutani 2001), dendrochronology indicates that in 466 BC, Choukai Jindai cedars different from ours were felled by an eruption of Mt Choukai. For our Choukai Jindai cedar, the most probable calendar date for the outer tree-ring Y235 tangent to the bark is between 463 and 456 BC. If both Choukai Jindai cedars fell in the same year, our ^{14}C dating for this floating tree-ring sample is younger by 3–10 yr compared to the date according to dendrochronology.

Comparison of ^{14}C Ages between the Choukai Jindai Cedar and European Tree Rings

As the calendar age of the Choukai Jindai cedar was finely determined, it is possible to compare the ^{14}C ages between the Choukai_AMS and IntCal04 data sets (moreover, the IntCal04 data set covering the calendar ages of the Choukai_AMS data set). It is calculated using a random walk model for the raw data sets of European tree rings, and hence we are able to compare the raw data sets from Japanese and European wood. Three data sets of the European tree rings were available for comparison: the Belfast and Seattle data sets for 741–461 cal BC (2690–2410 cal BP); and the Heidelberg data set for 740–621 cal BC (2689–2570 cal BP).

For the Belfast set, we used five 20-yr data for 2689–2590 cal BP and eighteen 10-yr data for 2589–2410 cal BP as shown in Figure 5a. For the Seattle set, we used three 20-yr and twenty-two 10-yr data for 2690–2410 cal BP as shown in Figure 5b. Two average values were used for duplicate samples in the Seattle.

Offsets

Figure 4 shows the time profiles of the Choukai_AMS and IntCal04 data sets between 741 and 462 cal BC. The Choukai_AMS data set has weighted averages of single-year data adjusted to the 5-yr spans of IntCal04. The Choukai_AMS data set is 16.4 ± 3.3 ^{14}C yr older than the IntCal04 data set in terms of the weighted average for the differences. We partially measured the 21 ^{14}C ages by single-year and decadal tree rings between Y0 and Y230 using a highly accurate LSC system, with 0.2% error (Sakurai et al. 2006). The weighted average of the difference between the Choukai_AMS and LSC data sets was 6.3 ± 4.7 ^{14}C yr. This consistency between both data sets, despite the different measuring methods, assured us that the Choukai_AMS data set is not biased towards an experimental offset. Estimation of interlaboratory offset between the MALT and the Seattle (or Belfast or Heidelberg) laboratories is difficult because we do not have direct cross-check data between the labs (Matsuzaki et al. 2004, 2007). However, considering that the precise LSC and the AMS ^{14}C ages for the Choukai tree rings indicate partly good consistency with the IntCal04 data, it is inferred that a laboratory offset is insignificant for the Choukai_AMS data set due to the MALT AMS (Takahashi et al. 2010).

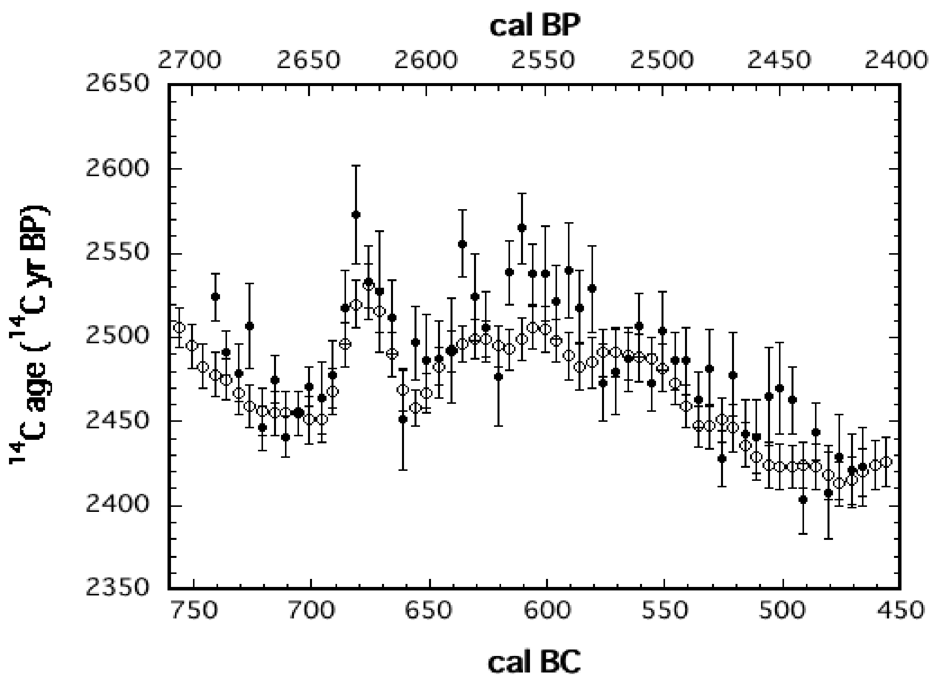

Figure 4 Time profiles of ^{14}C ages for the Choukai_AMS (closed circles) and IntCal04 (open circles) data sets. The Choukai_AMS data set consists of weighted averages for single-year data adjusted to the 5-yr spans of IntCal04, with Y233 defined as 462 cal BC.

Although the Belfast, Seattle, and Heidelberg data sets consist of decadal and/or bidecadal tree-ring samples, the Choukai_AMS data set can be compared to them by adjusting the data span as its data points are for single years. Most of the Choukai_AMS data, however, are weighted average values of alternate single-year ^{14}C ages, i.e. the ^{14}C age of a 10-yr span is averaged by 5 single-year ^{14}C

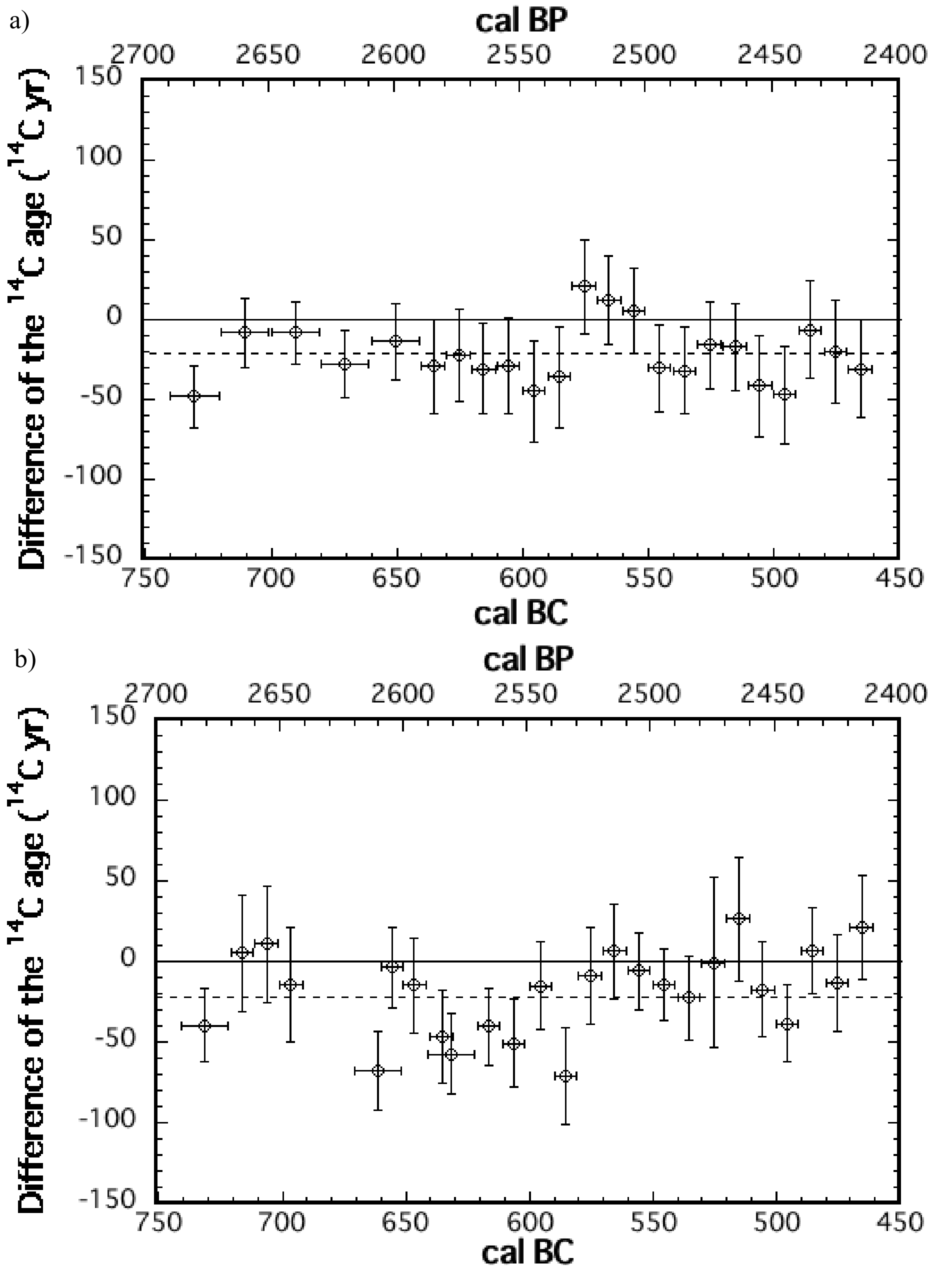

Figure 5 a) Time profiles of the ^{14}C age difference between the Belfast and Choukai_AMS data sets. The weighted average of the ^{14}C age difference was –21.3 ± 5.5 ^{14}C yr (dashed line). b) Time profiles of the ^{14}C age difference between the Seattle and Choukai_AMS data sets. The weighted average of the ^{14}C age difference was –22.7 ± 5.6 ^{14}C yr (dashed line).

ages in the span. Thus, it is necessary to evaluate the difference between average values of full and alternate single-year data for a span in comparison with 10-yr/20-yr span data of the Belfast, Seattle, and Heidelberg sets. The difference was calculated using a piece of consecutive 10 single-year

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Choukai_AMS ^{14}C ages. For 10- and 20-yr span data, the standard deviations (σ_2) for the average differences were 8.9 are 4.5 ^{14}C yr, respectively, when we estimate the averages using alternate 5 or 10 single-year data. Consequently, the error for an average value using alternate single-year data for 10- or 20-yr span is calculated by $\sigma^2 = \sigma_1{}^2 + \sigma_2{}^2$, where σ_1 is the estimated error for the average of alternate single-year data.

Figures 5a and b show the time profiles of the ^{14}C age difference for the Belfast and the Seattle data sets with respect to the Choukai_AMS data set, respectively. As shown in the figures, for 741–461 cal BC both data sets are younger than the Choukai_AMS data set; the weighted averages for the ^{14}C age differences were –21.3 ± 5.5 and –22.7 ± 5.6 ^{14}C yr for the Belfast and Seattle data sets, respectively. The difference for the Heidelberg data set was –9.2 ± 6.1 ^{14}C yr for 740–621 cal BC. Table 1 summarizes the differences of the 3 data sets compared to the Choukai_AMS data set, with the laboratory offsets of the Belfast and Heidelberg data sets referring to the Seattle data set for the long intervals. Considering that the Heidelberg data set has a laboratory offset of 15 ± 3 ^{14}C yr compared to the Seattle data set, the Choukai_AMS data set indicates an older offset of ~22 ^{14}C yr, which is equivalent to an offset of –2.8‰ in ^{14}C, compared to the 3 raw data sets of European woods.

Table 1 Offsets between the Choukai_AMS data set and the 3 IntCal04 raw data sets. Asterisks show the laboratory offsets by Reimer et al. (2004:1035).

Data set	cal yr interval cal BP	*n*	Offsets
Belfast-Choukai	2689–2410 (740–461 cal BC)	23	21.3 ± 5.5
Seattle-Choukai	2690–2410 (741–461 cal BC)	25	–22.7 ± 5.6
Heidelberg-Choukai	2689–2570 (740–621 cal BC)	12	9.2 ± 6.1
Belfast-Seattle*	3470–5 (1521 cal BC–cal AD 1945)	124	4 ± 2
Heidelberg-Seattle*	11,655–2575 (9706–600 cal BC)	86	15 ± 3

The Belfast, Seattle, and Heidelberg data sets measure ^{14}C in an Irish oak and 2 German oaks, respectively; they correspond well to the Choukai_AMS data set. The ^{14}C production rate due to cosmic rays at higher latitudes—where the European trees originate—is greater than at the mid-latitude of the Choukai Jindai cedar because of geomagnetic cut-off rigidities: 1–3 GV in Ireland and 3–5 GV in Germany compared to 10 GV in Japan. However, as the ^{14}C is quickly distributed as $^{14}CO_2$ and/or ^{14}CO (Jöckel and Brenninkmeijer 2002) in the atmosphere, the latitude gradient of $^{14}CO_2$ is diluted. Using a three-dimensional global tracer transport model simulation, Braziunas et al. (1995) indicated that the preindustrial atmospheric ^{14}C latitudinal gradient was rather modest at latitudes lower than 60°N in the Northern Hemisphere. This is because regional atmosphere-ocean $^{14}CO_2$ fluxes are controlled by regional wind-dependent gas exchange, the partial pressures of CO_2 in the air and sea, and the oceanic ratio of $^{14}C/^{12}C$. However, the offset of –2.8‰ in the Choukai Jindai cedar compared to the European tree rings is beyond the simulated difference of less than –1‰.

The island of Japan has a rainy season in summer caused by to the East Asian summer monsoon (EASM), which is a subtropic monsoon. In climatology, development of the EASM is linked to a northeastward intrusion of the equatorial southwesterlies. Meanwhile, Hua and Barbetti (2007) reported that the southwest Asian summer monsoon acts as a carrier of an atmosphere of Southern Hemisphere (ASH), and the atmospheric ^{14}C in low latitudes is reduced by the monsoon flow and northward migration of the Intertropical Convergence Zone (ITCZ) in summer. Although it is unlikely that the summer ITCZ reaches Japan Island at ~39ºN, the development of the EASM can provide the ASH to Japan in summer (Kueh and Lin 2010). Moreover, for the mid- to late Holocene, the EASM variability was investigated using the $\delta^{18}O$ of a stalagmite from a Chinese cave (29º29′N, 109º32′E) (Cosford et al. 2008, 2009). It shows that the Choukai Jindai cedar was covered by a period when the EASM intensified a little. This implies that the atmospheric ^{14}C in growth of the Choukai Jindai cedar is influenced by the ASH.

We estimated the influence of the Southern atmosphere to the Choukai Jindai cedar using an equation as well as Hua and Barbetti (2007). Using the ^{14}C time profiles as shown in Figure 6, the influence at each data point was calculated and then the averaged contribution of the ASH was estimated. In the calculation, N_1, T, and S are the Choukai_AMS data set, the Belfast or Seattle data sets, and SHCal04 Southern Hemisphere calibration data set, respectively (McCormac et al. 2004):

$$s = \frac{(N_1 - T)}{(N_1 - S)}$$

The ^{14}C dates are age-corrected (Stuiver and Polach 1977). The Belfast and Seattle data sets were calculated using the ^{14}C age data at the mid-point of decadal/bidecadal samples, respectively. The SHCal04 data set was averaged adjusting to the data span of the Belfast and Seattle data sets.

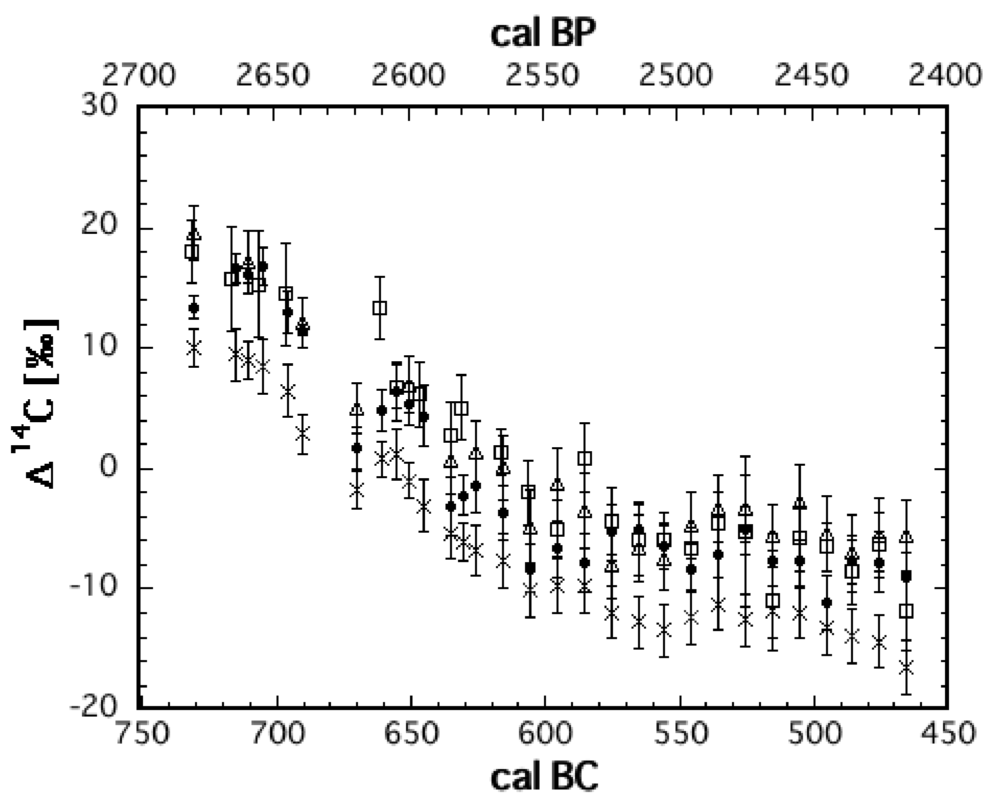

Figure 6 Time profiles of $\Delta^{14}C$ for the Choukai_AMS data (filled circles), the Belfast data (open triangles), the Seattle data (open squares), and SHCal04 Southern Hemisphere calibration data (crosses).

The contributions of the atmosphere of the Southern Hemisphere (ASH) to the Choukai Jindai cedar are estimated to be 31% and 26% for the Belfast and Seattle data sets, respectively. Meanwhile, the bomb profiles from nuclear detonations in AD 1964–1967 indicate that the influence of the ASH on the Japanese tree rings (Agematsu) as T is estimated at 35% using the average value between ~40ºN

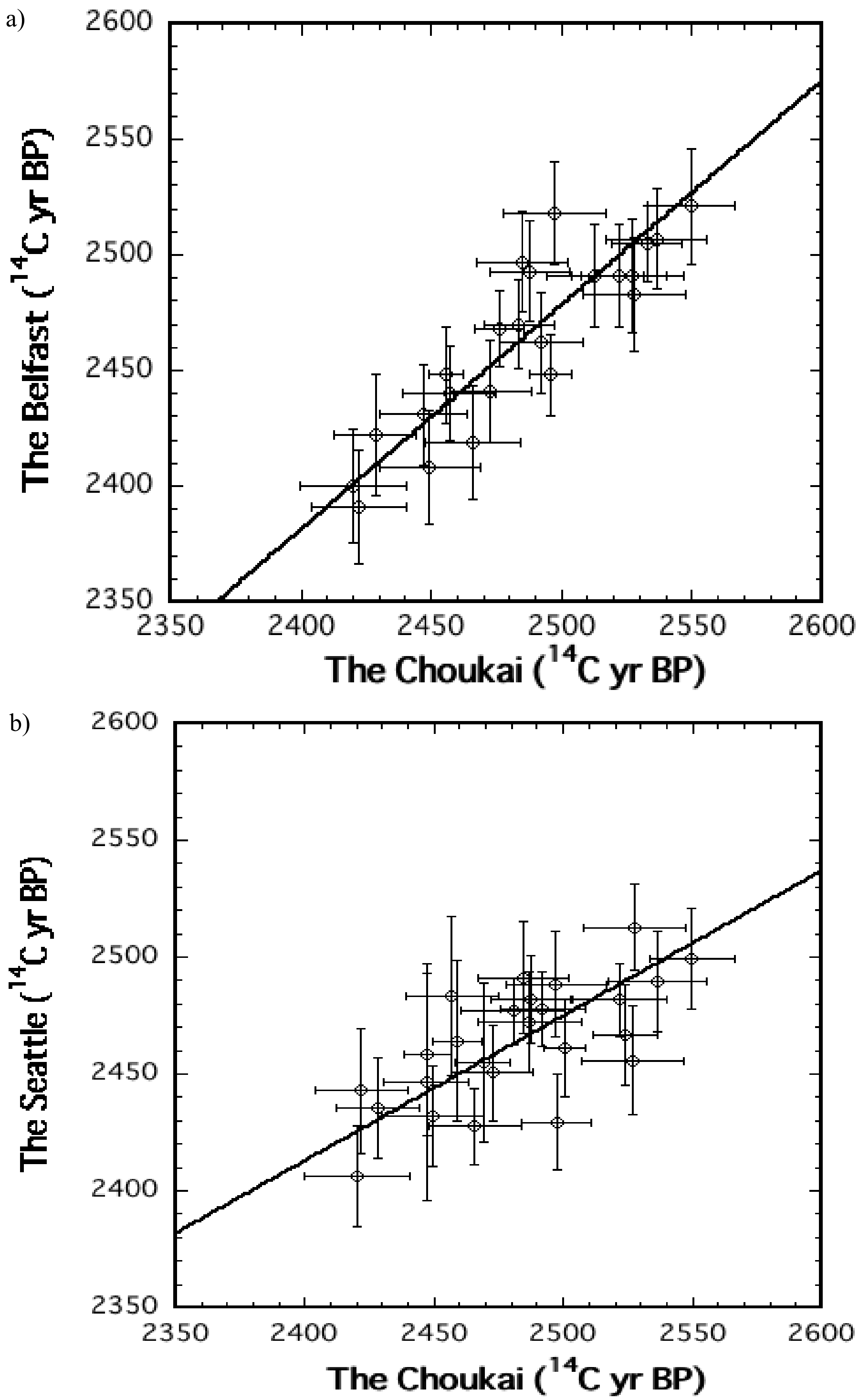

Figure 7 a) Scatter plot for the Choukai_AMS and Belfast data sets. The gradient of the least-squares-fitted line is 0.97 ± 0.16. b) Scatter plot of the Choukai_AMS and Seattle data sets. The gradient of the least-squares-fitted line is 0.62 ± 0.14.

to the North Pole (N_1) and the Southern Hemisphere winter data as *S* (Hua and Barbetti 2004). As the estimations are similar to each other, it is considered that the ASH flow through the EASM affects an offset of the Choukai Jindai cedar from the European tree rings.

Correlation of Time Variation

It is important to investigate not only the offset but also the relationship of time variations in ^{14}C ages between European and Japanese tree rings; this is because the correlation indicates a uniformity in the ^{14}C concentration in the atmosphere on a global scale. The correlations of the Belfast and Seattle data sets to the Choukai_AMS data set are shown as scatter plots in Figures 7a and b, respectively. As the figures show, the Choukai_AMS data set correlates well with the Belfast and Seattle data sets, with correlation coefficients of 0.89 and 0.68, respectively. Moreover, the gradients of least-squares-fitted lines (York 1968) are 0.97 ± 0.16 and 0.62 ± 0.14 for the Belfast and Seattle data sets, respectively. Similarly, the correlation coefficient and gradient of the IntCal04 data set to the Choukai_AMS data are 0.83 and 0.76 ± 0.07, respectively. The gradients indicate that the variability of the Belfast and Seattle data sets is comparable to and less than that of the Choukai_AMS data set, respectively.

CONCLUSION

We measured the ^{14}C ages of 165 single-year tree rings of the Japanese Choukai Jindai cedar using the AMS of the 5MV tandem accelerator at the University of Tokyo; the ^{14}C ages during 800–400 BC captured in the Choukai tree rings vary on a large scale, as evidenced by the IntCal04 data set. In order to precisely compare the ^{14}C ages with those of the European tree rings, we re-estimated the calendar age of the Choukai Jindai cedar precisely using 165 ^{14}C ages over a span of 280 yr. As a result, the new estimate for the Choukai Jindai cedar was that the 321 tree rings range from 780 to 460 cal BC (2729–2409 cal BP); this was accomplished by wiggle-matching for the IntCal04 calibration data using OxCal v 3.10 and evaluating the correlation coefficients between the Choukai_AMS and IntCal04 data sets. The uncertainty is 8 yr.

A comparison between the Choukai_AMS data set and the 3 IntCal04 raw data sets from European tree rings indicated that the Choukai_AMS data set has offsets compared to the 3 European data sets. The offsets were –21.3 ± 5.5 and –22.7 ± 5.6 ^{14}C yr for the Belfast and Seattle data sets, respectively. For 741–461 cal BC, the Belfast, Seattle, and Heidelberg raw data sets were ~22 ^{14}C yr younger than the Choukai_AMS data set, which is equivalent to 2.8‰ higher than the Choukai_AMS data set in terms of ^{14}C. Flows from the ASH into the Northern Hemisphere due to southeasterlies caused by the development of the EASM have probably produced the offsets.

The temporal profile of the Choukai_AMS data set was compared to the Belfast and Seattle data sets. The Choukai_AMS data set correlates well with the Belfast and Seattle data sets, with correlation coefficients of 0.89 and 0.68, respectively. Moreover, the gradients of least-squares-fitted lines in the scatter plot between the Choukai_AMS and the Belfast and Seattle data sets were 0.97 ± 0.16 and 0.62 ± 0.14, respectively, indicating that the variability of the Choukai_AMS data set is comparable to and greater than the former and latter, respectively. These results for the temporal profile of the Choukai ^{14}C ages show a global variation.

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