COMPARISON OF ^{14}C AGES BETWEEN LSC AND AMS MEASUREMENTS OF CHOUKAI JINDAI CEDAR TREE RINGS AT 2600 CAL BP

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ABSTRACT. Radiocarbon ages of Choukai Jindai cedar tree rings growing in the excess era of ^{14}C concentrations during 2757–2437 cal BP were measured using 2 types of ^{14}C measurement methods, i.e. liquid scintillation counting (LSC) and accelerator mass spectrometry (AMS). The difference between the 2 methods is 3.7 ± 5.2 ^{14}C yr on average for 61 single-year tree rings, indicating good agreement between the methods. The Choukai data sets show a small sharp bump with an average ^{14}C age of 2497.1 ± 3.0 ^{14}C yr BP during 2650–2600 cal BP. Although the profile of the Choukai LSC data set compares well with that of IntCal04, having a ^{14}C age difference of 4.6 ± 5.3 ^{14}C yr on average, the Choukai LSC ^{14}C ages indicate variability against the smoothed profile of IntCal04.

INTRODUCTION

The history of solar activity can be traced through the cosmogenic nuclide ^{14}C embedded in tree rings (Stuiver and Quay 1980; Stuiver and Braziunas 1989). Reconstructions of sun spot numbers from the ^{14}C records indicate that the activity of the sun was not constant but variable, with various periods for minima and maxima for the last 10,000 yr (Solanki et al. 2004; Usoskin et al. 2004; Miyahara et al. 2008). Stuiver and Braziunas (1989) found remarkable events of atmospheric ^{14}C excess in a 9600-yr record, categorizing them into 2 types of oscillations, Maunder and Spörer, by the duration of their enhancements. The Δ^{14}C oscillations are slightly different in length; the Maunder-type oscillations have a period of ~180 yr and the Spörer-type oscillations last ~40 yr longer. They showed that there were 9 Maunder-type and 8 Spörer-type events in the record, implying that the Δ^{14}C oscillations are due to variations in solar-wind forcing (Stuiver and Braziunas 1989).

A Choukai Jindai cedar (*Cryptomeria japonica*) containing 320 tree rings was dug out of Japan's northern region (39°05′N, 140°03′E; see Figure 1a). The calendar age of the cedar was wiggle-matched to 2426.5 ± 12.5 cal BP for the outer tree ring (Sakurai et al. 2006). Although cross-dating could not be done because dendrochronology for old tree rings in Japan is confined to trees with a good pattern, the calendar age of the cedar is refined by adding the ^{14}C ages of other tree rings. As seen in Figure 1b, this wood sample is located in a Maunder-type periods in the IntCal04 curve of the last 3000 yr (Reimer et al. 2004).

Although the era of the Choukai Jindai cedar, which we term the "Choukai excess," is classified as Spörer-type based on its duration, the profile of Choukai excess shows an important feature comprising a rapid change with a hump after the grand excess, as shown Figure 1c. In the IntCal04 calibration curve, there is a small sharp hump that corresponds to a rapid change in ^{14}C concentrations over 50 yr from 2650 to 2600 BP.

In order to investigate the variation in ^{14}C concentrations of the Choukai excess in detail, we measured the ^{14}C ages of single-yr tree rings of the Choukai Jindai cedar using 2 types of ^{14}C dating methods, highly accurate liquid scintillation counting (LSC) and accelerator mass spectrometry

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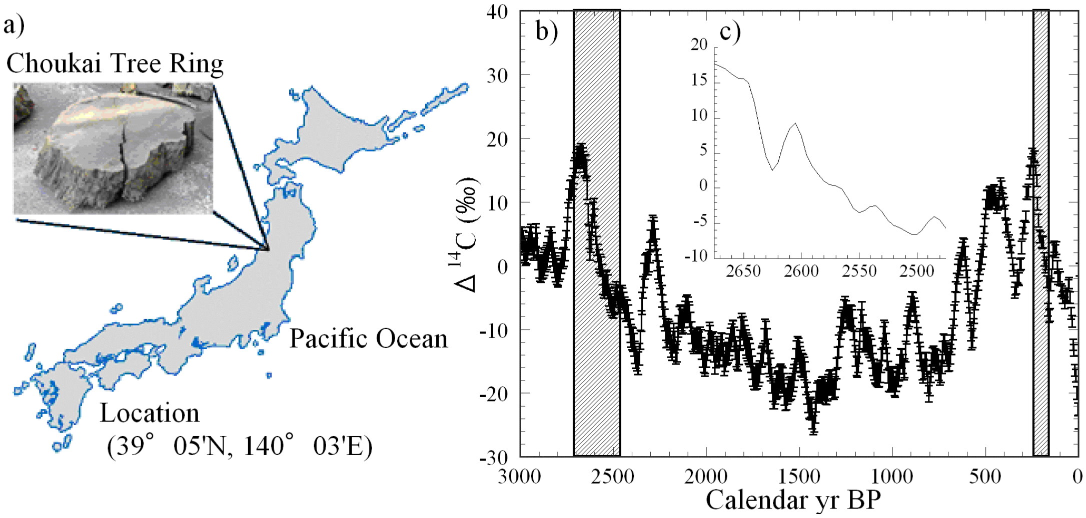

Figure 1 a) Location of the Choukai Jindai cedar; b) Period of the Choukai Jindai cedar and the Maunder period in the Δ^{14}C profile of IntCal04 (Reimer et al. 2004); c) Detail of the timeframe of the Choukai Jindai cedar.

(AMS) (Sakurai et al. 2006; Suzuki et al. 2007). A large number of samples constituting the entire structure of ^{14}C ages in the Choukai excess have been measured (Suzuki et al. 2010). Features of ^{14}C ages such as the small sharp hump have been precisely measured using the capability of a highly accurate LSC system. This paper demonstrates the consistency of both the methods, assuring accurate estimation of ^{14}C ages of the Choukai Jindai cedar tree rings. A precise profile of the small sharp hump is then compared with a data set of IntCal04, because there are few published data and few comparisons of ^{14}C ages between Japanese and European tree rings around the period of the bump.

MEASUREMENTS

The tree rings are numbered from Y0 to Y225 from the center toward the outer edge of the Choukai Jindai cedar wood sample. Y0 is assigned to a tree ring that is outside of ~85 tree rings from the pith. Using LSC, we measured the ^{14}C ages of 95 single-year tree rings from Y8 to Y145 and the ^{14}C ages of 8 decadal samples: D5, D20, D30, D40, D50, D60, D180, and D225. For instance, D5 is a 10-yr wood section from Y1 to Y10. Moreover, for comparison with IntCal04, the single-year data sets were averaged with 5-yr-span data to 27 data sets. It is advantageous to be able to compare a single-year data set with data sets spanning multiple years.

The wood sample was boiled and then separated using tweezers into single-year tree rings and 10-yr wood sections, which were subsequently milled for a series of chemical treatments. As the α cellulose in the cell walls of tree rings is the most reliable chemical component of the wood for measuring the annual concentration of ^{14}C, it was chemically extracted.

From the extracted cellulose, benzene was synthesized for LSC. The amounts of synthesized benzene were typically 5.3 or 10.5 g, produced from the single-year wood sample of 60 or 120 g, respectively. Benzene was poured in a high-purity Teflon®/copper counting vial (20 or 7 mL) or a Teflon/POM counting vial (7 mL).

Measurements of ^{14}C in the synthesized benzene were carried out with a typical statistical accuracy of 2‰ using an LSC system (Quantulus 1220$^{\text{TM}}$) with an ultra-low background level. Although the major factors of the systematic error in the LSC measurement are the amount of benzene and uniformity of quality for the vials, they are controlled to keep the error <1‰ (Endo et al. 2000). The counting rate of an old tree-ring sample is ~40 cpm for 5.3 g of benzene, and the background rate is typically 0.08 cpm for 1 g of dead benzene sample with a Teflon/POM counting vial. The ^{14}C age of a sample is calculated from the ratio of the concentrations of ^{14}C between the sample and a standard (Stuiver and Polach 1977). By following the same chemical process for the tree-ring samples, a standard benzene sample is synthesized from oxalic acid (SRM 4990-C) having a standard concentration of ^{14}C certified by the National Institute of Standards and Technology (NIST).

The same 61 tree-ring samples (from a total 95) that were measured by LSC were measured using AMS at the Micro Analysis Laboratory, Tandem accelerator, University of Tokyo (MALT). The typical measurement error is 30 ^{14}C yr (Suzuki et al. 2007, 2010). ^{14}C ages were calculated using the measured value of the $^{14}C/^{12}C$ ratio for the sample and the NIST standard samples.

RESULTS AND DISCUSSION

All LSC measurements were carried out with a counting statistical accuracy of <2‰ for the 95 samples from Y8 to Y146. ^{14}C ages ranged between 2449 and 2539 ^{14}C yr BP with errors of 16–22 ^{14}C yr for the 95 samples. For the half-decadal data set, the errors are between about 6 and 14 ^{14}C yr as determined from the weighted average of the single-year data sets.

All LSC measurements were performed more than twice using the Quantulus system in order to ensure the stability of counting. The error multiplier $k = \sigma_2/\sigma_1$ is 1.3 for the 95 samples; where σ_1 is the average error in ^{14}C ages and σ_2 is the standard deviation calculated from the ^{14}C age differences between the measurements for each sample. The value of k confirms consistent LSC measurements for all samples because σ_2 is approximately $\sigma \sqrt{2}\, \sigma_1$ for a measurement with same counting statistics without systematic errors.

A comparison of 61 single-year data sets allows us to ensure accuracy for both LSC and AMS measurements. The consistency between the ^{14}C ages of the same tree rings obtained by these different methods indicates little offset, and hence accurate determination of ^{14}C ages by both methods. Figure 2 shows a distribution of the ^{14}C age differences between the 61 single-year data sets of LSC and AMS. The weighted average of the differences between the ^{14}C ages determined by the 2 methods is 3.7 ± 5.2 ^{14}C yr, indicating good consistency between the methods. Moreover, a least-square-fitted curve in the figure represents a Gaussian function with a standard deviation of 42.6 ^{14}C yr ($\chi^2/\nu = 1.0$). Considering this Gaussian distribution and that the errors in the ^{14}C ages determined from LSC and AMS measurements are typically 16 and 30 ^{14}C yr, respectively, the differences between ^{14}C ages approximately represent a statistical distribution.

In Figure 3, the ^{14}C ages measured using LSC are compared with AMS measurements of 5-yr spans as a function of the tree-ring number. ^{14}C dating of the tree rings was carried out by wiggle-matching to the IntCal04 calibration data using OxCal v 3.10 (Bronk Ramsey 1995, 2001). The calendar age of Y8 is 2645–2635 cal BP, estimated using the LSC data set of 95 ^{14}C ages with a 95.4% confidence level. Figure 3 depicts a peak of ^{14}C ages with a sharp enhancement between Y0 and Y50 for both measurements. The height of the hump is ~73 ^{14}C yr. As the calendar date of Y0 determined from the LSC data is 2648 cal BP, this hump thus corresponds to the small sharp peak in the IntCal04 calibration curve. Moreover, ^{14}C ages of the 10-yr sections are shown in the figure at the location corresponding to the enhancement, indicating the appearance of the small sharp peak even in the 10-yr profile.

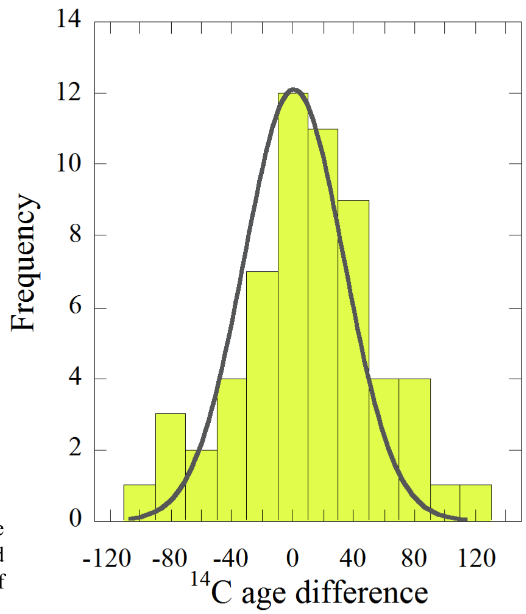

Figure 2 Distribution of ^{14}C age differences between the LSC and the AMS measurements. A least-square-fitted Gaussian function represents the standard deviation of 42.6 ^{14}C yr ($\chi^2/\nu = 1.0$).

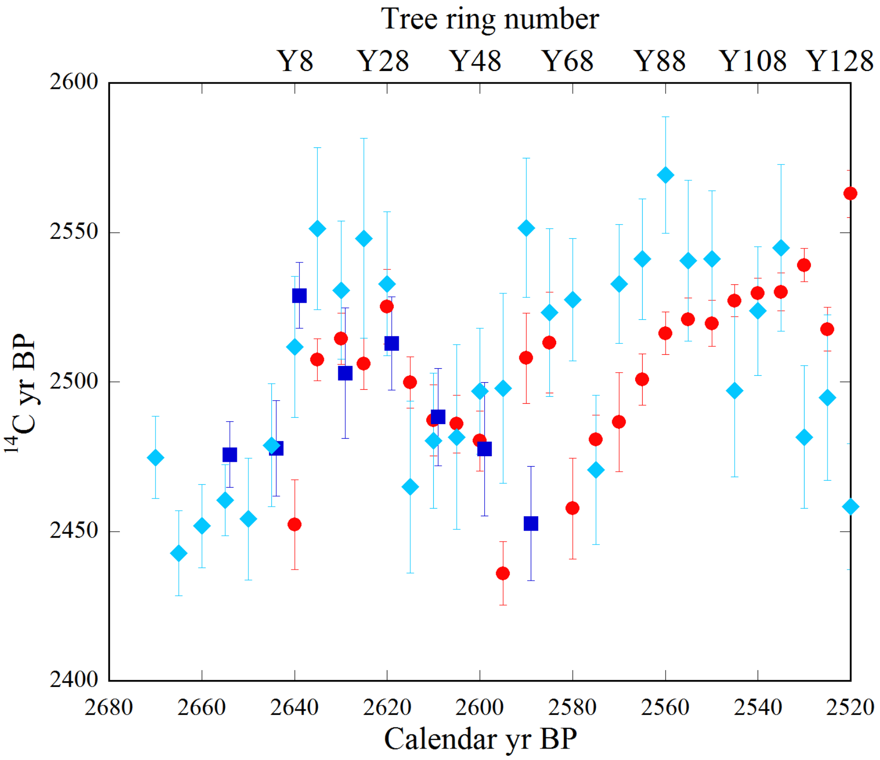

Figure 3 Comparison of ^{14}C ages between LSC (filled circles) and AMS (filled diamonds) measurements of Choukai 5-yr data sets. Filled squares represent the Choukai LSC 10-yr data set.

Over the 48 yr from Y8–Y55, corresponding to the small sharp hump of the IntCal04 calibration curve, the averages of ^{14}C ages are 2497.1 ± 3.0, 2505.0 ± 6.5, and 2510.0 ± 8.4 ^{14}C yr BP for the LSC single-year, LSC 10-yr, and AMS single-year wood sections, respectively. These values are mutually consistent within the errors, assuring high accuracy in the measurements.

There are 2 published data around the small sharp hump for an east Japanese cypress and German oak tree ring. The average ^{14}C age of the Japanese tree rings in Ouban 1 archaeological site (34°41′N, 132°70′E) is 2500.0 ± 8.8 ^{14}C yr BP between 2600 and 2635 cal BP from AMS measurements of its 5-yr wood sections. A few data points around the sharp hump, however, are coarsely dispersed between 2461 and 2610 ^{14}C yr, with an error of 27 ^{14}C yr (Ozaki et al. 2007). Moreover, the average ^{14}C age of the German oak is 2497.2 ± 7.2 ^{14}C yr BP from its 10-yr wood sections, with a height of 55 ^{14}C yr for the hump (Kromer 2004). Average ages of the east Japanese and European tree rings agree well with the Choukai Jindai cedar data around the period of the small sharp hump, indicating a global event as the cause for the enhancement.

In Figure 4, the ^{14}C age profile of the Choukai Jindai cedar determined by LSC is compared to that of IntCal04 in the vicinity of the small sharp hump. For the period 2640–2600 cal BP, the weighted average ^{14}C age difference between the Choukai LSC and the IntCal04 data sets is 4.6 ± 5.3 ^{14}C yr. Although both the profiles are mutually well consistent, the Choukai LSC profile exhibits variability in the ^{14}C ages in contrast to the IntCal04 profile, which indicates the smoothing effect caused by the data constructed using a random walk model for the data sets of 10- and/or 20-yr spans.

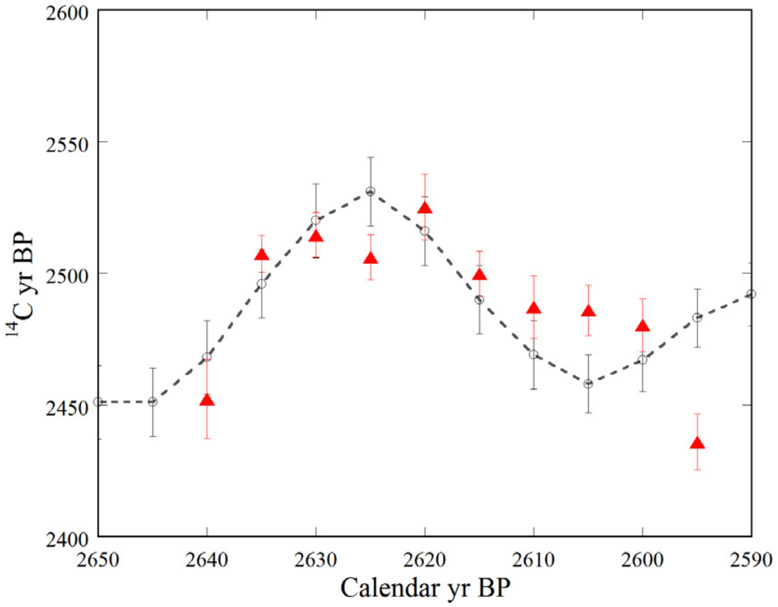

Figure 4 Comparison of ^{14}C age profiles between the Choukai LSC 5-yr data set (filled triangles) and the IntCal04 data set (open circles) in the period of the small sharp hump. The dotted line is an interpolation of IntCal04.

CONCLUSION

^{14}C ages of 61 single-year Choukai Jindai cedar tree rings were measured using LSC and AMS. The weighted average of ^{14}C age differences between the LSC and AMS measurements is 3.7 ± 5.2 ^{14}C yr, indicating good consistency between the 2 methods. This result demonstrates that both the measurements are accurate for determining ^{14}C ages. Precise measurements showed that the Choukai tree rings have a sharp hump of 73 ^{14}C yr during 2650–2600 cal BP, similar to that found in IntCal04.

The LSC data set can be compared with published data sets on 5- or 10-yr sections from east Japanese and European tree rings using single-year measurements. Around the period of the small sharp hump, the average ^{14}C age of 2497.1 ± 3.0 ^{14}C yr BP for the Choukai LSC data set agreed well with the averages of 2500.0 ± 8.8 and 2497.2 ± 7.2 ^{14}C yr BP for the east Japanese and the German oak tree rings, respectively, indicating that the enhancement is not limited to one region but is instead a global event.

For the period 2640–2600 cal BP, the weighted average ^{14}C age difference between the Choukai LSC and IntCal04 data sets is 4.6 ± 5.3 ^{14}C yr. Although both profiles are mutually well consistent, the Choukai LSC data indicates variability in the profile when compared to the smoothed profile of IntCal04.

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