

## **<sup>14</sup>C AGE MEASUREMENTS OF SINGLE-YEAR TREE RINGS OF OLD WOOD SAMPLES 22,000 <sup>14</sup>C YEARS BP**

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**ABSTRACT.** Radiocarbon ages of single-year tree rings were measured for Kaminoyama wood samples using accelerator mass spectrometry (AMS) in 2 Japanese facilities, MALT and JAEA, in order to investigate the periodic variation of <sup>14</sup>C concentrations relating to the 11-yr solar cycle near 26,000 yr BP. Eight sequential measurements of <sup>14</sup>C ages were carried out for a set of 13 alternate single-year tree rings covering 26 yr. Averages of the 5 data sets in MALT and 3 data sets in JAEA were  $22,146 \pm 50$  and  $22,407 \pm 58$  <sup>14</sup>C yr BP, respectively, indicating an offset of  $260 \pm 77$  <sup>14</sup>C yr. Multiple sequential measurements are advantageous for evaluating offsets. The standard deviation of the residuals of <sup>14</sup>C ages from the averages in each data set was 118 <sup>14</sup>C yr, in contrast to that of 234 <sup>14</sup>C yr for the combined data sets due to an elimination effect in the offsets. The profiles of weighted mean values for the residuals of <sup>14</sup>C ages showed similar enhancements with a width of ~12 yr for measurements in the 2 facilities. This indicates the reproducibility of the multiple sequential measurements. In the profile for the combined 8 data sets, the <sup>14</sup>C enhancement was  $73 \pm 36$  <sup>14</sup>C yr from the average.

### **INTRODUCTION**

Around 26,000 yr BP, radiocarbon concentrations in the atmosphere were about 40–60% higher than those of the present day, and approach 0% at 45,000 yr BP (Chiu et al. 2007). On the other hand, the temporal profile of the Earth's geomagnetic dipole was roughly in antiphase to that of <sup>14</sup>C concentrations over the past 45,000 yr. Near 26,000 yr BP, geomagnetic intensity was ~25% lower compared with the present (Laj et al. 1996). Since <sup>14</sup>C is produced in the atmosphere by cosmic rays coming to the Earth through geomagnetic fields, <sup>14</sup>C concentrations vary according to the geomagnetic intensity. It is inferred, therefore, that the higher <sup>14</sup>C concentrations near 26,000 yr BP are mainly caused by the higher intensity of cosmic rays reaching the Earth compared to the present.

It is necessary to evaluate the extent to which geomagnetic intensity affected cosmic rays coming to the Earth in that time period, as we do not know if the energy spectrum of cosmic rays is for the last 50,000 yr is the same as the present. Since cosmic rays are modulated with solar activities, such as the 11-yr solar cycle appearing as a periodic variation in sun spot numbers, <sup>14</sup>C concentrations should also be modulated with a periodicity of 11-yr (Stuiver and Braziunas 1998; Suzuki et al. 2007). Therefore, it is expected that the change in the amplitude of <sup>14</sup>C concentrations related to the 11-yr variation is considerable ~26,000 yr BP due to the weaker geomagnetic intensity at the time.

In order to investigate a periodic variation of <sup>14</sup>C concentrations relating to the 11-yr solar cycle around 26,000 yr BP, we measured the <sup>14</sup>C ages in single-year tree rings of an old wood sample, named Kaminoyama, whose <sup>14</sup>C date is ~22,500 <sup>14</sup>C yr BP, indicating a calendar year of 26,000 cal BP (Fairbanks et al. 2005). However, it is difficult to find solar modulation by measuring <sup>14</sup>C ages in a set of tree rings using accelerator mass spectrometry (AMS) since the statistical accuracy of an AMS measurement is ~1.2% for old wood samples, compared to the <0.5% amplitude expected for an 11-yr modulation in <sup>14</sup>C concentrations.

We thus attempted to statistically improve the precision for determining <sup>14</sup>C ages by employing multiple sequential measurements. Using AMS machines in 2 facilities, 8 sequential measurements

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were carried out for a set of 13 single-year tree rings of Kaminoyama wood samples. We describe the reproducibility of the multiple sequential measurements and a profile of variable components in the  $^{14}\text{C}$  ages.

## MEASUREMENTS

The Kaminoyama tree-ring samples used in this study were dug out of Kaminoyama ( $38^{\circ}07'\text{N}$ ,  $140^{\circ}17'\text{E}$ ) in northern Japan (Figure 1). There were approximately 100 tree rings sampled. In a previous study, a set of alternate consecutive 13 single-year tree rings covering 26 yr was chosen from the wood sample to evaluate the characteristics of multiple sequential measurement (Gandou et al. 2004). The set of  $\alpha$ -cellulose samples was chemically extracted for each single-year tree ring and graphite samples were prepared to measure the annual concentrations of  $^{14}\text{C}$ .

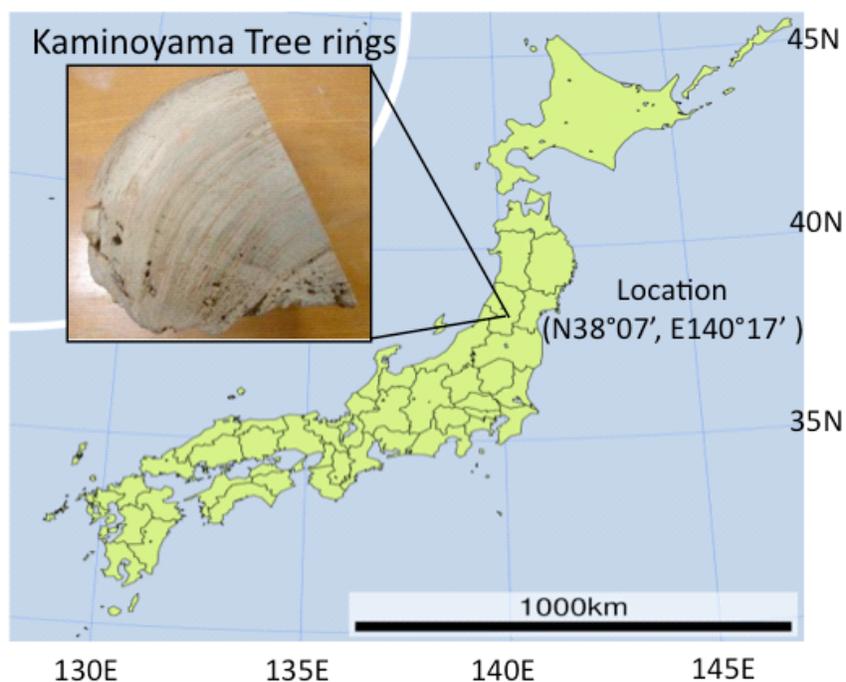


Figure 1 Photograph of Kaminoyama tree rings and the sample location in Japan

$^{14}\text{C}$  measurements for the set of graphite samples were carried out using the 2 AMS facilities: the 5MV Pelletron tandem accelerator at the Micro-Analysis Laboratory (MALT), University of Tokyo, and the 3MV Tandetron accelerator at the Japan Atomic Energy Agency (JAEA). Comparison between the measurements using different AMS machines is valuable trying to locate spurious signals in the profile of  $^{14}\text{C}$  ages for a same set of tree rings.

Since the typical statistical error of  $^{14}\text{C}$  measurement is  $\sim 100$   $^{14}\text{C}$  yr for the Kaminoyama wood sample, measuring just 1 set of single-year tree rings is not sufficient to evaluate a small variability like the 11-yr solar periodic variation since a single set of measurements can have an amplitude of  $\sim 40$   $^{14}\text{C}$  yr. However, multiple measurements of a tree-ring sample can improve the precision of the  $^{14}\text{C}$  age. For instance, the data set of 8 sequential measurements used here reduces the error to an average of 37  $^{14}\text{C}$  yr, comparable to the amplitude related to an 11-yr solar variation.

A set of 13 graphite samples was produced from each set of  $\alpha$ -cellulose samples; thus, 104 graphite samples were measured in total. Consequently, 8 data sets of  $^{14}\text{C}$  ages were obtained from the 13 single-year tree rings. Of the 8 total data sets, 5 sets were measured in MALT and 3 in JAEA. This multiple sequential measurement is advantageous to find out and eliminate any offsets between the data sets.

**RESULTS AND DISCUSSION**

Figure 2 shows the 8 data sets of  $^{14}\text{C}$  ages measured for the tree rings as a function of the tree-ring number. The average for all data sets was 22,238  $^{14}\text{C}$  yr BP with a standard deviation of 234  $^{14}\text{C}$  yr.  $^{14}\text{C}$  ages were scattered within an error of  $\sim 97$   $^{14}\text{C}$  yr estimated from the counting statistics. From the resulting average  $^{14}\text{C}$  age, the calendar age of the Kaminoyama tree rings is estimated to be  $26,740 \pm 292$  cal BP using the calibration curve of Fairbanks et al. (2005).

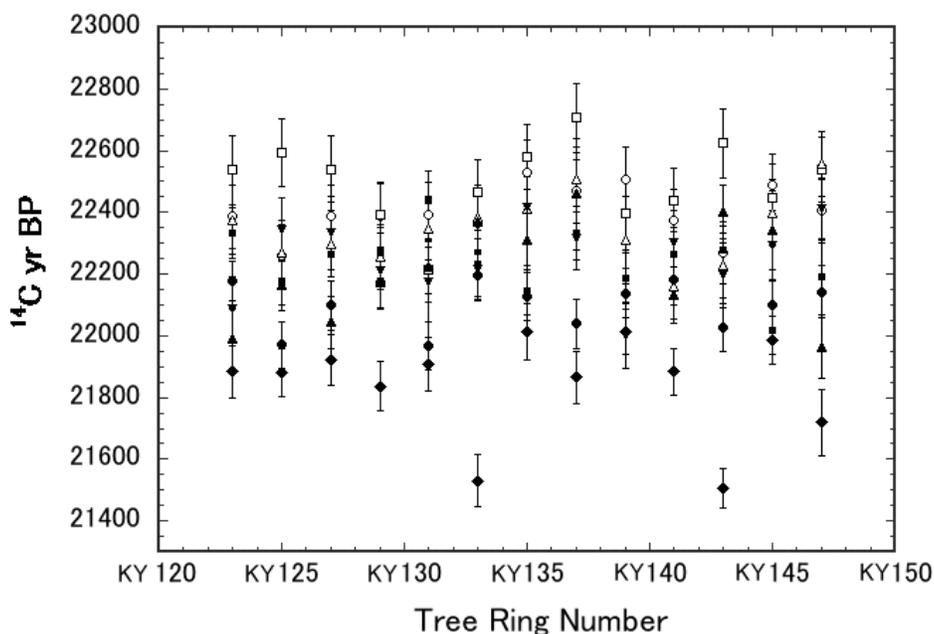


Figure 2 Eight data sets of  $^{14}\text{C}$  ages measured for the set of 13 single-year tree rings

Average  $^{14}\text{C}$  ages for each data set are shown in Figure 3 as a function of the data set number. The averages of the 5 data sets in MALT (sets 1 to 5) and the 3 data sets in JAEA (sets 6 to 8) were  $22,146 \pm 50$   $^{14}\text{C}$  yr BP and  $22,407 \pm 58$   $^{14}\text{C}$  yr BP, respectively. Although the average of data set 1 seems to deviate from the other 4 averages at MALT, a  $\chi^2$  test for the set of 5 averages did not indicate a significant deviation at the 5% level. The difference between the averages at the facilities was  $260 \pm 77$   $^{14}\text{C}$  yr. The offset, presumably, implies an uncertainty of the machine background for significant old samples.

Ratios for the 8 data sets of the standard deviations to the counting errors were between 0.8 and 2.0. Except for the 2 data sets at MALT, the ratios of the remaining 6 data sets were between 0.8 and 1.2. These values indicate that the variations in  $^{14}\text{C}$  ages are similar between the 8 data sets despite having different averages.

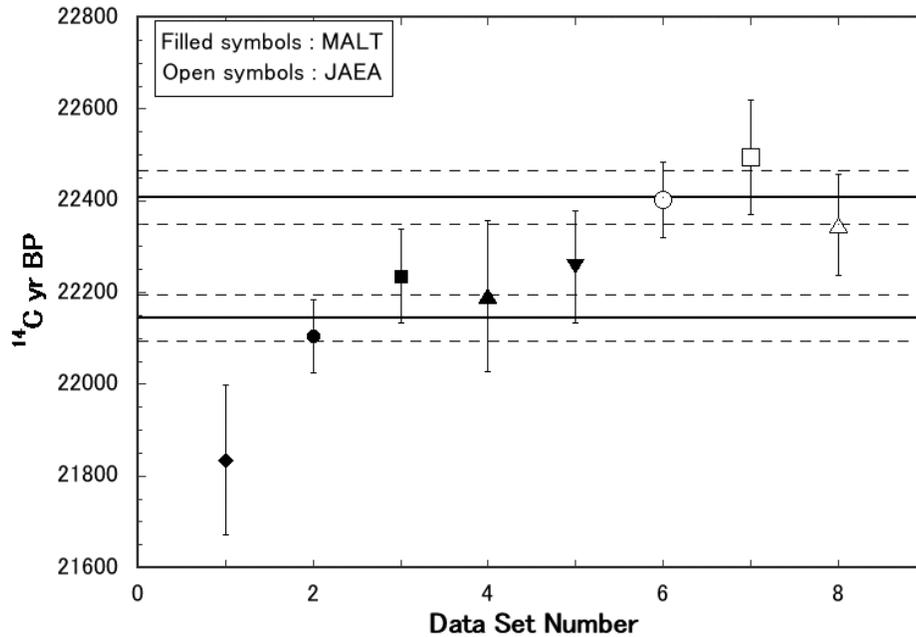


Figure 3 Averages of the  $^{14}\text{C}$  ages for data sets. The difference between the averages at MALT and JAEA is  $260 \pm 77$   $^{14}\text{C}$  yr. Symbols in this figure correspond to those of Figures 2 and 4 for each data set.

Figure 4 shows the residuals subtracted from the averages of  $^{14}\text{C}$  ages in each data set. The standard deviation was 118  $^{14}\text{C}$  yr for the residuals, in contrast to 234  $^{14}\text{C}$  yr for all the  $^{14}\text{C}$  ages. Moreover, for the residuals, the ratios of the standard deviations to the counting errors for the 8 data set are between 0.6 and 1.8 for the each tree ring, compared to ratios between 1.8 and 3.6 for all  $^{14}\text{C}$  ages. These values indicate an elimination effect of offsets between the data sets and illustrate the advantage of multiple sequential measurements for a set of single-year tree rings.

Although our objective is to pick out variable components of  $^{14}\text{C}$  ages for a set of single-year tree rings via multiple sequential measurements, the 8 data sets for the residuals are available for a preliminary analysis. We calculated the weighted mean values of the  $^{14}\text{C}$  residuals for each tree ring. For instance, they are the weighted mean values of 5, 3, and 8  $^{14}\text{C}$  residuals at each tree ring for the MALT, JAEA, and combined data sets, respectively. Figure 5 plots 3 profiles of the weighted mean values as a function of the tree-ring number for the data sets. Those values have approximate errors of 40, 61, and 33  $^{14}\text{C}$  yr for the MALT, JAEA, and combined data sets, respectively. A similar variability appeared as a hump with a width of roughly 12 yr between KY131 and KY143 in all 3 profiles.

In the profile for the combined data sets, the enhancement of the hump was  $73 \pm 36$   $^{14}\text{C}$  yr against the average  $-4 \pm 9$   $^{14}\text{C}$  yr of the 13 points. Assuming that the profile is uniform with a constant value, the  $\chi^2$  value was 16.6, which is equivalent to a significance level of 17% at a degree of freedom of 12. Hence, presumably we can check the statistical significance of the hump at a level of 5% by adding 3 more data sets to the multiple sequential measurements.

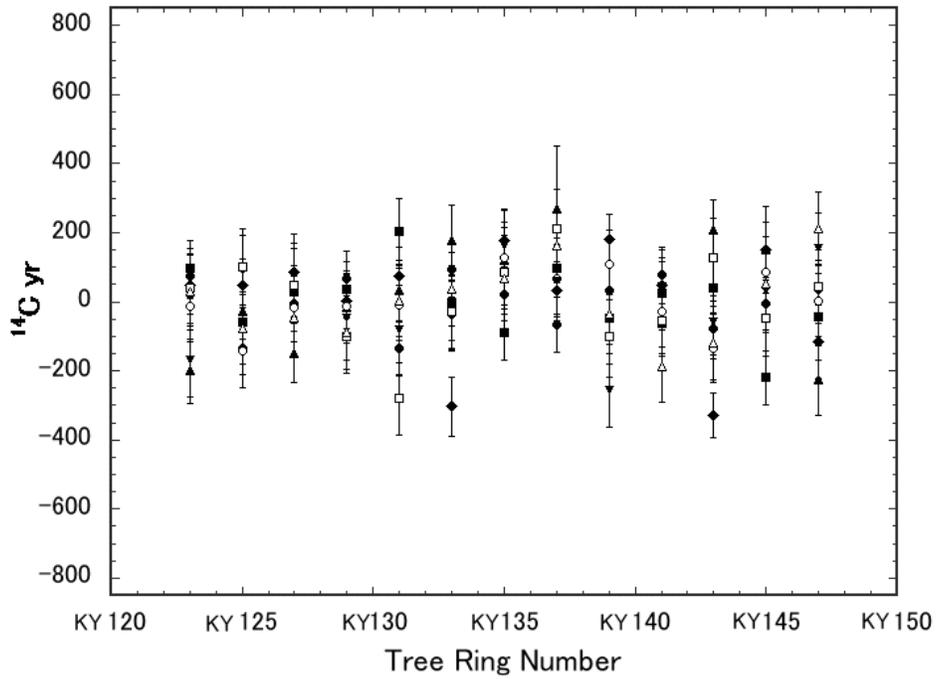


Figure 4 Residuals of  $^{14}\text{C}$  ages from the averages for the 8 data sets. Symbols as in Figure 3.

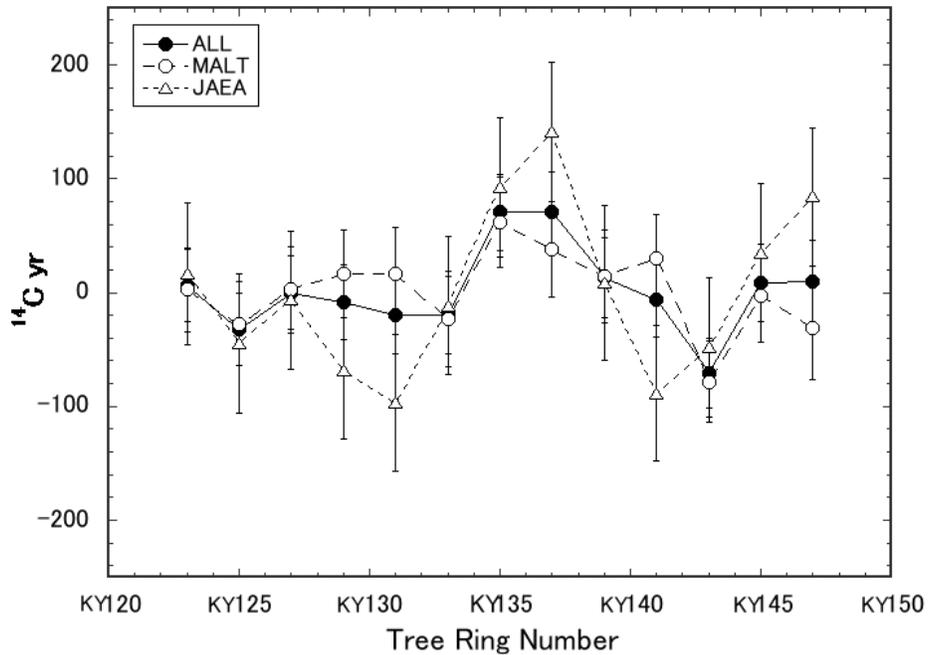


Figure 5 Three profiles of weighted mean values for the residuals of  $^{14}\text{C}$  ages for the 8 data sets. Symbols show the 5 data sets of MALT (open circles), 3 data sets of JAEA (triangles), and all 8 data sets (filled circles).

## CONCLUSION

$^{14}\text{C}$  ages of single-year tree rings were measured for the Kaminoyama wood sample using 2 AMS facilities, MALT and JAEA, in order to investigate a periodic variation of  $^{14}\text{C}$  concentrations relating to the 11-yr solar cycle around 26,000 yr BP. In order to evaluate such a small variability of 11 yr, 8 sequential measurements of  $^{14}\text{C}$  ages were carried out for a set of 13 alternate single-year tree rings, producing 104 graphite samples in total.

The average for all 8 data sets was 22,238  $^{14}\text{C}$  yr BP with a standard deviation of 234  $^{14}\text{C}$  yr. The averages of the 5 data sets in MALT and the 3 data sets in JAEA were  $22,147 \pm 50$  and  $22,407 \pm 58$   $^{14}\text{C}$  yr BP, respectively, indicating an offset of  $260 \pm 77$   $^{14}\text{C}$  yr between the 2 facilities. The standard deviation for the residuals of  $^{14}\text{C}$  yr ages from the average in each data set was 118  $^{14}\text{C}$  yr in contrast to that of 234  $^{14}\text{C}$  yr for all the combined data sets. This difference signals an elimination effect of the offsets between the data sets and illustrate the advantage of multiple sequential measurements for a set of single-year tree rings.

The profiles of weighted mean values for the residuals of  $^{14}\text{C}$  ages for each data set showed a similar variability appearing as a hump in the curve, with a width of roughly 12 yr, almost identical to the expected 11-yr variation due to the solar cycle. These results attest to the reproducibility of multiple sequential measurements.

In the profile for the 8 data sets, the enhancement in the curve was  $73 \pm 36$   $^{14}\text{C}$  yr from the average. Presumably, we can check the statistical significance of the hump at a level of 5% by adding 3 more data sets of multiple sequential measurements.

## ACKNOWLEDGMENTS

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