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RADIOCARBON IN 9TH TO 5TH CENTURY BC TREE-RING SAMPLES FROM THE OUBAN 1 ARCHAEOLOGICAL SITE, HIROSHIMA, JAPAN

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ABSTRACT. In order to investigate the regional atmospheric radiocarbon offset, accelerator mass spectrometry (AMS) ^{14}C measurements were made on 5-yr increments of a Japanese wood sample dendrochronologically dated to 820–436 BC. The ^{14}C data from the Japanese tree-ring samples were compared with the IntCal04 calibration curve (Reimer et al. 2004). In most parts, the differences between IntCal04 and ^{14}C dates in the Japanese tree-ring samples were within experimental statistical errors. At around 680 BC, however, significant differences of up to 100 ^{14}C yr were observed. These differences may indicate either regional offsets in Japan or the short-term fluctuation of a subdecadal timescale in atmospheric ^{14}C variations.

INTRODUCTION

It is generally assumed that radiocarbon dates can be calibrated precisely within assigned limitations using the international calibration data sets, the latest version of which is IntCal04 (Reimer et al. 2004). Although these data are based on the compilation of extensive observations of atmospheric ^{14}C variations over time from several ^{14}C laboratories, the samples used for the compilation have been limited to a rather small number of areas in the world. Tree-ring samples, which provide the most precise calibration data, have typically been obtained from Europe and North America.

Regional offsets, the differences in ^{14}C ages between the international calibration data set and individual sites, have been discussed by several researchers (e.g. Stuiver and Braziunas 1998; Hua et al. 2004). For example, offsets between the 2 hemispheres were identified by McCormac et al. (1998) and Hogg et al. (2002). Based on ^{14}C measurements of contemporaneous wood samples for the Northern and Southern hemispheres between AD 950 and 1950, differences in the ^{14}C concentration between the 2 hemispheres and their time dependence became apparent. This led to the development of a Southern Hemisphere-specific calibration curve (SHCal04, McCormac et al. 2004).

Regional offsets have been observed even within a hemisphere. Kromer et al. (2001) suggested that ^{14}C concentrations in Turkish wood during the period 800 to 750 BC differed from IntCal98. Sakamoto et al. (2003) also suggested possible regional ^{14}C offsets for Japan during the 2nd century AD by measuring ^{14}C in Japanese cedar tree rings. These examples indicate that more attention should be paid to regional ^{14}C offsets. In order to avoid biasing in calibration, it is important to investigate regional ^{14}C offsets for the region of interest and its time variation.

In this study, we performed accelerator mass spectrometry (AMS) ^{14}C measurements for a dendrochronologically dated Japanese cypress for the period 820–436 BC. The 1st millennium BC has been of great interest in archaeological research in Japan because a major cultural change took place during this period. The Jomon culture, characterized by a hunter-gatherer style of life, gave way to the Yayoi culture, characterized by paddy-rice agriculture and the subsequent use of iron and bronze. According to our recent investigations (Fujio et al. 2005), the transition started at around the 9th to 10th century BC in the northern part of the Kyushu Islands, then gradually spread eastward

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and reached northeastern Honshu in the 3rd to 4th century BC. In the past few years, we have obtained a number of ^{14}C measurements in the Yayoi period to investigate this period further and to better understand the cultural interactions between eastern Asia and the Japanese archipelago.

The possibility of a regional ^{14}C offset for Japan has been of great concern because the calibration curve is rather flat in the period 750–400 BC, and a regional offset would greatly affect the calibrations of ^{14}C dates and their interpretations. This study is also a part of our effort to construct a regional ^{14}C calibration curve for the Japanese archipelago and its neighbors, since it is expected that calibration data sets for individual regions should be more important in the future, in light of recent improvements in AMS precision and accuracy.

DESCRIPTIVE BACKGROUND

The sample specimen used in this study was cut from a wooden board before preservation treatment. The board was one of many wooden artifacts excavated from the Ouban 1 archaeological site (Hiroshima Prefecture, Japan), which dates from the middle of the Yayoi period. The wood was Japanese cypress, *hinoki* (*Chamaecyparis obtuse*), and had about 400 well-preserved tree rings. Dendrochronological dating was performed with a yearly precision by comparison with a standard ring pattern for Japanese cypress developed by the National Research Institute for Cultural Properties, Nara (1990). As a result, the specimen included tree rings ranging between 820–436 BC.

The sample specimen was divided into 5-yr blocks. Since the ring widths varied from layer to layer, the weight of each block varied from 20 to 80 mg. Each tree-ring block was pulverized into fine pieces and treated with the conventional acid-alkali-acid (AAA) method using an automatic apparatus (Sakamoto et al. 2004). These AAA-treated samples were further processed with $NaClO_2$ and HCl to bleach out lignin with Cl_2, and finally with 17.5% NaOH solution to remove most β- and γ-cellulose. The purified sample, mainly consisting of α-cellulose, was neutralized with HCl, washed with Milli-Q® water, filtered, and dried at 110 °C overnight. Two samples from 445–436 BC failed to yield enough α-cellulose for AMS measurements.

Several mg of extracted α-cellulose were weighed and sealed in a quartz tube, together with CuO (elementary analysis grade: Merck, Ltd.) and Sulfix® (elementary analysis grade: Wako Pure Chemical Industries, Ltd.), the latter being a mixture of cobalt and silver oxides used for removing sulfur oxide and halogens. The sample tube was heated to 850 °C for 3 hr to completely oxidize the α-cellulose. The gases obtained, including CO_2, were transferred to the high-vacuum CO_2 purification system at the National Museum of Japanese History, and CO_2 was purified cryogenically. The purified CO_2 was then reduced to graphite at 600 °C in the presence of hydrogen and an Fe catalyst. The graphite was pressed into targets for AMS measurement, which was performed at MALT (Micro Analysis Laboratory, Tandem Accelerator), the University of Tokyo (Pelletron 5UD, NEC) (Matsuzaki et al. 2004).

^{14}C dates of the samples were calculated from the measured $^{14}C/^{12}C$ ratios, after correcting for isotope effects by normalizing $^{13}C/^{12}C$ ratios to $\delta^{13}C = -25‰$, and the ^{14}C half-life of 5568 yr (Stuiver and Polach 1977; Mook and van der Plicht 1999). The $^{14}C/^{12}C$ ratios were normalized to the average of HOxII standards, which were prepared by using the same CO_2 purification processes as in the samples.

Table 1 Results of ^{14}C measurements for Japanese tree-ring samples.

Sample name	Dendro-age (BC)	^{14}C age (BP)	$\delta^{13}C$ (‰)	Lab code
HRHH-C261-1	820–816	2664 ± 37	–23.1 ± 1.6	MTC-07000
HRHH-C261-2	815–811	2670 ± 37	–22.5 ± 1.7	MTC-07001
HRHH-C261-3	810–806	2659 ± 40	–21.2 ± 2.1	MTC-07002
HRHH-C261-4	805–801	2649 ± 38	–22.7 ± 1.8	MTC-07003
HRHH-C261-5	800–796	2614 ± 38	–22.6 ± 1.7	MTC-07004
HRHH-C261-6	795–791	2569 ± 37	–22.4 ± 1.8	MTC-07005
HRHH-C261-7	790–786	2603 ± 37	–21.1 ± 1.8	MTC-07006
HRHH-C261-8	785–781	2549 ± 38	–22.3 ± 1.9	MTC-07007
HRHH-C261-9	780–776	2578 ± 38	–21.6 ± 1.9	MTC-07008
HRHH-C261-10	775–771	2555 ± 38	–22.0 ± 1.9	MTC-07009
HRHH-C261-11	770–766	2520 ± 38	–22.2 ± 1.9	MTC-07010
HRHH-C261-12	765–761	2494 ± 63	–22.9 ± 2.0	MTC-07011
HRHH-C261-13	760–756	2486 ± 66	–28.8 ± 5.0	MTC-07012
HRHH-C261-14	755–751	2485 ± 38	–22.2 ± 2.0	MTC-07013
HRHH-C261-15	750–746	2519 ± 37	–21.5 ± 1.8	MTC-07014
HRHH-C261-16	745–741	2467 ± 36	–18.6 ± 1.7	MTC-07015
HRHH-C261-17	740–736	2445 ± 38	–21.7 ± 2.2	MTC-07016
HRHH-C261-18	735–731	2417 ± 39	–21.2 ± 2.1	MTC-07017
HRHH-C261-19	730–726	2488 ± 36	–22.7 ± 1.7	MTC-07018
HRHH-C261-20	725–721	2477 ± 40	–21.1 ± 2.3	MTC-07019
HRHH-C261-21	720–716	2448 ± 38	–21.5 ± 1.6	MTC-07020
HRHH-C261-22	715–711	2475 ± 37	–23.1 ± 1.5	MTC-07021
HRHH-C261-23	710–706	2430 ± 37	–23.1 ± 1.7	MTC-07022
HRHH-C261-24	705–701	2406 ± 38	–22.1 ± 1.7	MTC-07023
HRHH-C261-25	700–696	2388 ± 37	–23.3 ± 1.7	MTC-07024
HRHH-C261-26	695–691	2554 ± 26	–23.1 ± 0.6	MTC-07184
HRHH-C261-27	690–686	2516 ± 26	–22.5 ± 0.5	MTC-07185
HRHH-C261-28	685–681	2461 ± 27	–22.5 ± 0.8	MTC-07186
HRHH-C261-29	680–676	2610 ± 27	–21.3 ± 0.8	MTC-07187
HRHH-C261-30	675–671	2523 ± 26	–22.8 ± 0.4	MTC-07188
HRHH-C261-31	670–666	2492 ± 26	–23.0 ± 0.4	MTC-07189
HRHH-C261-32	665–661	2473 ± 26	–21.9 ± 0.6	MTC-07190
HRHH-C261-33	660–656	2459 ± 27	–21.8 ± 0.8	MTC-07191
HRHH-C261-34	655–651	2467 ± 26	–21.5 ± 0.6	MTC-07192
HRHH-C261-35	650–646	2501 ± 26	–23.0 ± 0.4	MTC-07193
HRHH-C261-36	645–641	2569 ± 27	–24.0 ± 0.6	MTC-07194
HRHH-C261-37	640–636	2481 ± 26	–22.9 ± 0.4	MTC-07195
HRHH-C261-38	635–631	2547 ± 26	–22.1 ± 0.5	MTC-07196
HRHH-C261-39	630–626	2539 ± 25	–21.7 ± 0.4	MTC-07197
HRHH-C261-40	625–621	2503 ± 26	–22.3 ± 0.5	MTC-07198
HRHH-C261-41	620–616	2501 ± 26	–23.1 ± 0.3	MTC-07199
HRHH-C261-42	615–611	2481 ± 26	–21.9 ± 0.5	MTC-07200
HRHH-C261-43	610–606	2539 ± 26	–22.6 ± 0.6	MTC-07201
HRHH-C261-44	605–601	2523 ± 48	–22.3 ± 0.6	MTC-07202
HRHH-C261-45	600–596	2525 ± 26	–22.3 ± 0.5	MTC-07203
HRHH-C261-46	595–591	2509 ± 26	–21.2 ± 0.6	MTC-07204
HRHH-C261-47	590–586	2521 ± 26	–23.3 ± 0.5	MTC-07205
HRHH-C261-48	585–581	2486 ± 27	–22.2 ± 0.9	MTC-07206
HRHH-C261-49	580–576	2516 ± 27	–22.0 ± 0.7	MTC-07207
HRHH-C261-50	575–571	2498 ± 41	–22.1 ± 0.8	MTC-07208
HRHH-C261-51	570–566	2489 ± 28	–22.6 ± 0.7	MTC-07210
HRHH-C261-52	565–561	2445 ± 27	–22.5 ± 0.7	MTC-07211
HRHH-C261-53	560–556	2446 ± 28	–22.5 ± 0.7	MTC-07212
HRHH-C261-54	555–551	2495 ± 27	–23.2 ± 0.5	MTC-07213
HRHH-C261-55	550–546	2427 ± 28	–18.3 ± 0.8	MTC-07214
HRHH-C261-56	545–541	2486 ± 27	–22.5 ± 0.5	MTC-07215

Table 1 Results of ^{14}C measurements for Japanese tree-ring samples. *(Continued)*

Sample name	Dendro-age (BC)	^{14}C age (BP)	$\delta^{13}C$ (‰)	Lab code
HRHH-C261-57	540–536	2408 ± 26	−21.6 ± 0.5	MTC-07216
HRHH-C261-58	535–531	2392 ± 27	−23.4 ± 0.4	MTC-07217
HRHH-C261-59	530–526	2447 ± 27	−20.6 ± 0.5	MTC-07218
HRHH-C261-60	525–521	2452 ± 27	−18.2 ± 0.6	MTC-07219
HRHH-C261-61	520–516	2450 ± 55	−21.6 ± 0.6	MTC-07220
HRHH-C261-62	515–511	2374 ± 30	−21.7 ± 0.5	MTC-07221
HRHH-C261-63	510–506	2423 ± 30	−21.4 ± 0.5	MTC-07222
HRHH-C261-64	505–501	2464 ± 48	−21.1 ± 0.6	MTC-07223
HRHH-C261-65	500–496	2486 ± 56	−22.9 ± 0.3	MTC-07224
HRHH-C261-66	495–491	2463 ± 27	−20.9 ± 0.5	MTC-07225
HRHH-C261-67	490–486	2420 ± 27	−22.4 ± 0.5	MTC-07226
HRHH-C261-68	485–481	2440 ± 28	−21.9 ± 0.6	MTC-07227
HRHH-C261-69	480–476	2481 ± 28	−21.1 ± 0.8	MTC-07228
HRHH-C261-70	475–471	2392 ± 27	−22.1 ± 0.5	MTC-07229
HRHH-C261-71	470–466	2444 ± 27	−21.7 ± 0.5	MTC-07230
HRHH-C261-72	465–461	2429 ± 26	−21.0 ± 0.5	MTC-07231
HRHH-C261-73	460–456	2395 ± 27	−21.4 ± 0.6	MTC-07232
HRHH-C261-74	455–451	2426 ± 28	−18.5 ± 0.5	MTC-07233
HRHH-C261-75	450–446	2353 ± 27	−23.0 ± 0.6	MTC-07234

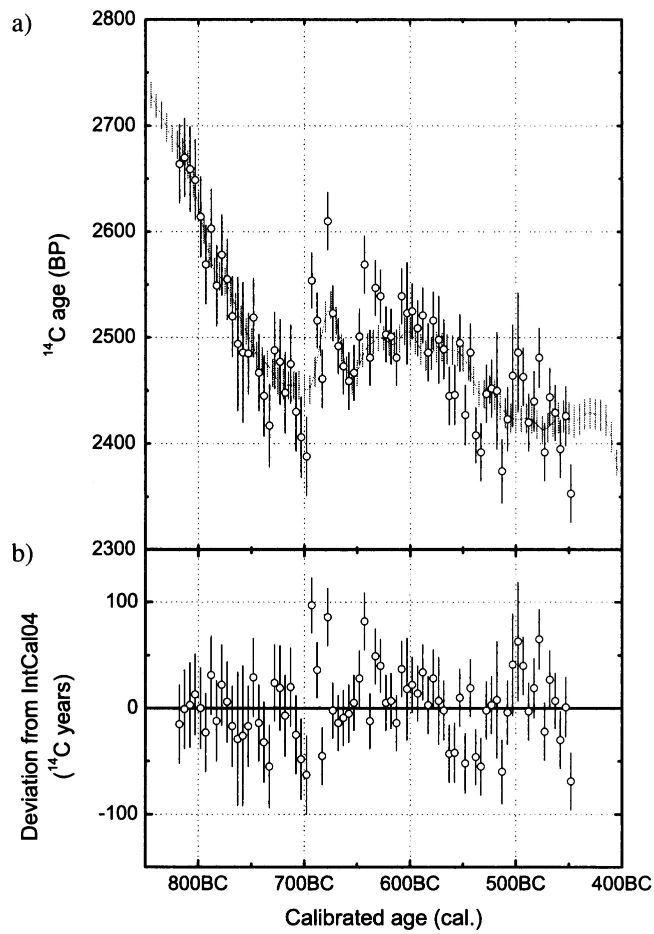

Figure 1 Comparison between IntCal04 (gray line) and ^{14}C ages of Japanese tree-ring samples (open circles). ^{14}C ages obtained in this study are plotted with IntCal04 (a). Deviations from IntCal04 are plotted against tree-ring ages (b). The 2 data sets agree well within the measurement errors (1 σ shown), with a few exceptions.

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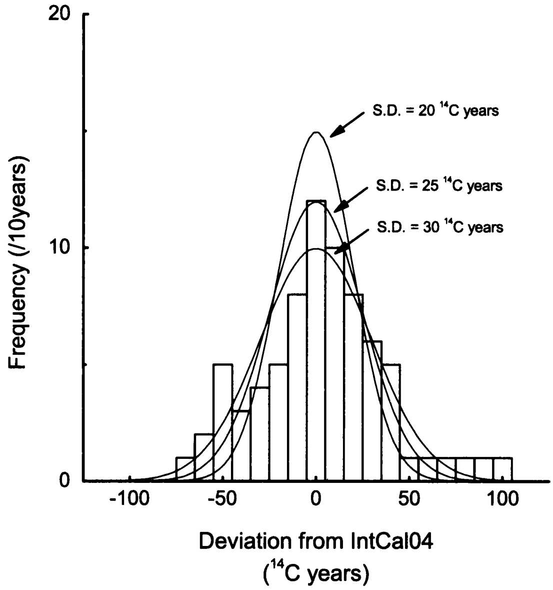

Figure 2 Distribution of ^{14}C age differences between Japanese cypress and IntCal04. Normal distribution curves with standard deviations of 20, 25, and 30 ^{14}C yr are also illustrated. The distribution of differences between Japanese wood and IntCal04 agrees well with a normal distribution with a standard deviation of 25 ^{14}C yr.

RESULTS AND DISCUSSION

The results of the ^{14}C measurements for each sample specimen are given in Table 1. The errors include the statistics of ^{14}C counts in tree-ring samples, uncertainties in the $^{13}C/^{12}C$ and $^{14}C/^{12}C$ ratios of the standards, and processed blanks. Uncertainty given for the certified ^{14}C value of HOxII standard was not included. Processed blank samples were prepared in the same manner from commercial graphite powder (99.9%, Soekawa Chemical Co. Ltd.), which was confirmed to have the same ^{14}C level as IAEA C1 standard. The extent of modern carbon contamination was ~0.1 pMC or less, practically negligible.

In Figure 1, the results obtained in this study are illustrated with the IntCal04 calibration curve (Reimer et al. 2004) for comparison. The deviations from IntCal04 are also plotted against calendar year (Figure 1, lower panel). Error bars illustrated in Figure 1 represent 1-σ uncertainty. The 2 data sets mostly agree well within 2-σ error. The overall distribution of those differences shows a normal distribution with a standard deviation of around 25 ^{14}C yr (Figure 2). The weighted average of these differences was calculated to be 3.5 ± 3.6 ^{14}C yr. Considering that the error given for each measurement mostly ranges from 25 to 40 ^{14}C yr, we conclude that our ^{14}C data from Japanese tree rings agree well with IntCal04, which was based on tree rings from North America and Europe.

The differences from IntCal04 are up to ~100 ^{14}C yr for a few data points at around 680 BC. Such a cluster of exceptional data is hard to explain by a purely statistical error. In Figure 3, 10-yr moving averages of results obtained in this study are illustrated in the same manner as Figure 1. The magnitude of differences from IntCal04 become smaller than the raw results for 5-yr samples. Since the data used for construction of IntCal04 in this period are decadal samples—while the present results

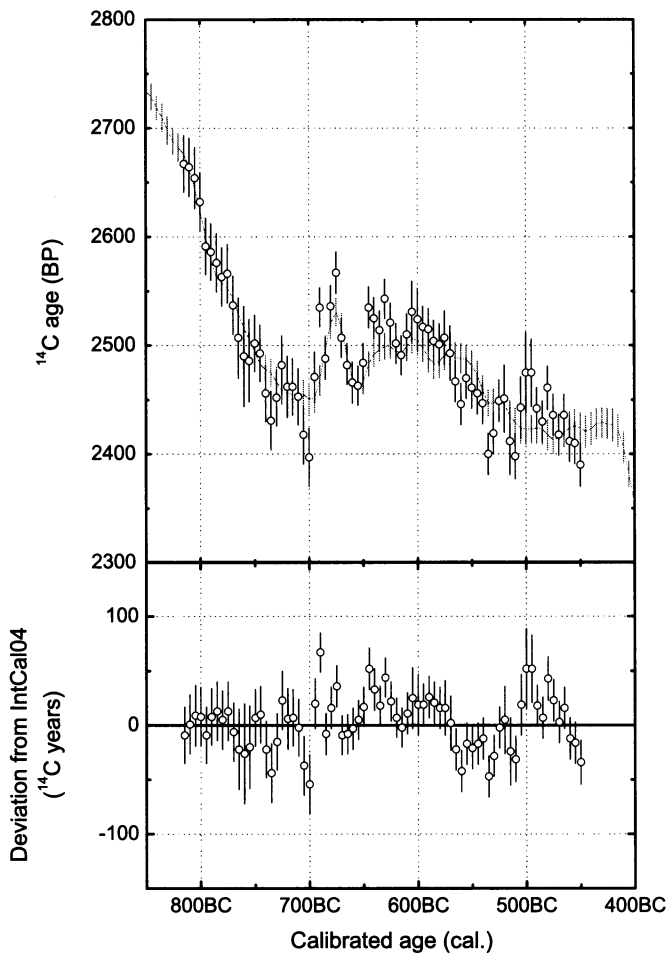

Figure 3 The 10-yr moving averages of ^{14}C ages of Japanese tree-ring samples (open circles) are plotted with IntCal04 (gray line). Compared to raw results for 5-yr samples (Figure 1), 10-yr moving averages agree much better with IntCal04.

are obtained for 5-yr samples—this might represent short-term fluctuation of ^{14}C variations, like an 11-yr solar cycle. To confirm this possibility, we plan to measure ^{14}C for single-year samples. However, we cannot exclude the possibility that these differences are caused by local effects in a limited area, rather like the influence volcanic gases can have in producing old ^{14}C ages. We also plan to measure other tree-ring samples of the same periods, taken from a different area of Japan and from other countries.

CONCLUSION

From the 9th to 5th century BC, the atmospheric ^{14}C variation in Japan is in acceptable agreement with the IntCal04 calibration curve, which was constructed with ^{14}C ages of tree rings from Europe and North America. However, significant differences were observed at around 680 BC. We plan to investigate these deviations in more detail by using single-year tree-ring samples and samples from other areas.

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