# <sup>14</sup>C DATING ANCIENT JAPANESE DOCUMENTS

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ABSTRACT. We measured radiocarbon ages of 11 pieces of ancient Japanese documents by accelerator mass spectrometry (AMS). The purpose of this study is to compare the relationship between the calibrated <sup>14</sup>C age and the historical age of Japanese paper samples. Calibrated ages of nine pieces agree with their historical ages, indicating that Japanese ancient documents can be used for <sup>14</sup>C dating in the recent historic period. On the other hand, the <sup>14</sup>C age of paper that was used for reinforcement of a sutra is ca. 300 yr older than the historical age of the sutra. This shows that the sutra was repaired with old paper.

#### Introduction

Radiocarbon dating has provided useful information for archaeological study because it gives an absolute age (after calibration) of the ancient artifact. For age determination of culturally valuable objects, the most advantageous characteristic of AMS is its ability to measure <sup>14</sup>C ages for ca. 1-mg carbon samples. Because of the historical value, small size, or low carbon content of most cultural objects, there is a limit to the amount of sample submitted for <sup>14</sup>C measurement. However, the AMS method has made it possible to date such samples, thus extending the application to valuable cultural objects produced in the recent historic period.

History is a reconstruction of past human activity. This reconstruction is accomplished, in part, by gathering evidence in the form of relics or documents. The historic period is largely clarified with the help of ancient documents. Therefore, the ages of ancient documents determined by scientific methods will provide useful information for studying the historic period. <sup>14</sup>C ages should be calibrated to obtain calendar ages, because the atmospheric <sup>14</sup>C concentration in the past is a function of time. But even the calibrated age, obtained by projecting the <sup>14</sup>C age on the calibration curve, indicates only a period that the sample has been kept in a system closed from the atmosphere. For this reason, the calibrated age can be different from the historical age in which the cultural object existed as a tool. A calibrated age is usually older than an historical age. For example, the calibrated age of a statue made of the central part of a log is older than historical age when the statue was made.

The difference between calibrated age and historical age becomes more serious for more recent samples, usually because a more accurate age is required. It is important to clarify the relation between calendar age and historical age for some sorts of the cultural properties made of wood, bone and paper. This investigation intends to clarify the relation between calibrated age and historical age by AMS <sup>14</sup>C measurements of Japanese ancient documents that have known historical ages.

#### **METHODS**

## Features of the Samples

We used AMS to measure the <sup>14</sup>C ages of 11 fragmentary pieces of Japanese ancient documents whose historic ages had been obtained by palaeography. The oldest sample was written during the last part of Heian period or the early part of Kamakura period (roughly corresponding to the 12th

century AD). The youngest sample was written in the Edo period (17th–19th century AD). The historical ages of the samples are summarized in Table 1. Six samples (Sample no. 1–6) were cut out of the Buddhist sutras, two (No. 7, 8) were cut from scattered pieces of sutras, one (No. 9) was cut from a scroll, one (No. 10) was cut out of paper that was attached to the back of a sutra for reinforcement, and one (No. 11) was cut out of paper that had been made in the later part of this century from the paper used to reinforce the sample No. 10. Four samples (No. 1,2,4,5) feature Chinese characters written with brush and Chinese ink; the other samples have no ink. Each sample with Chinese characters was cut from a part of the margin, avoiding the characters in order to preserve paleographical information, and avoiding contamination by carbon used for writing the characters.

TABLE 1. List of Ancient Documents

Sample		
no.	Historical period	Feature of original sample
1	Last Heian-early Kamakura period (12th century)	Buddhist sutra, with Chinese characters
2	First Kamakura period (13th century)	Buddhist sutra, with Chinese characters
3	First Kamakura period (not certain) (13th century)	Buddhist sutra, no Chinese characters
4	Early Kamakura period (13th century)	Buddhist sutra, with Chinese characters
5	Nanbokutyou period (latter 14th century)	Buddhist sutra, with Chinese characters
6	Last Nanbokutyou period (last 14th century)	Buddhist sutra, no Chinese characters
7	Last Heian-early Kamakura period (12th-13th century)	Scattered piece of a Buddhist sutra, no Chinese characters
8	Last Heian-early Kamakura period (12th-13th century)	Scattered piece of a Buddhist sutra, no Chinese characters
9	Last Heian-early Kamakura period (12th-13th century)	A scroll with axis, no Chinese characters
10	Edo period (17th-19th century)	Reinforcing paper of a sutra, no Chinese characters
11	Edo period (17th–19th century)	Recycled paper from No. 10 sample, no Chinese characters

### **Sample Preparation**

The paper samples were covered with dust and stained with water from the surroundings. We extracted the alpha-cellulose fraction from the samples. First, the samples were washed with distilled water in a supersonic washer to exclude any possible contamination from the dust. Next we removed adsorbed components by alternating washings with 1.2 M HCl and 1.2 M NaOH solutions, each washing taking ca. 2–3 hr. Each treatment was repeated several times, depending on the degree of stain. Cellulose was prepared with a 0.07 M NaClO<sub>2</sub> solution under acidic conditions (adjusted with HCl). We repeated this treatment 4 or 5 times. Finally, alpha-cellulose was extracted by soaking

at room temperature for ca. 30 min in a 17.5% NaOH solution. The alpha-cellulose was combusted to  $CO_2$  at 950°C (using CuO). The  $CO_2$  was purified by successive vacuum trap with liquid  $N_2$ , frozen ethyl alcohol and frozen n-pentane. The graphite (to be used for AMS) was prepared by reducing  $CO_2$  with  $H_2$  and Fe as catalyst at 650°C in a sealed Vycor® glass tube (Kitagawa et al. 1993).

#### <sup>14</sup>C Measurement

The instrument used for this study was the Tandetron AMS at the Dating and Materials Research Center, Nagoya University (Nakamura et al. 1985; Nakamura and Nakai 1988). The conventional  $^{14}$ C ages are calculated using measured  $^{14}$ C/ $^{13}$ C ratios, using standard procedures (Oda 1994). The 1- $\sigma$  error we used is the square root of unbiased variance of  $^{14}$ C/ $^{13}$ C ratios obtained by alternative measurements on sample and standard. The  $\delta^{13}$ C value used for isotopic fractionation correction was measured by a Finnigan MAT-252 mass spectrometer on the CO<sub>2</sub> gas before graphitization. The  $^{14}$ C age for each sample of the ancient documents was measured 2 to 4 times, in order to improve statistics. The 1- $\sigma$  error of the averaged  $^{14}$ C ages used here includes a full statistical treatment.

### RESULTS AND DISCUSSION

Table 2 shows the  $^{14}$ C ages and  $\delta^{13}$ C results for the ancient documents. The  $^{14}$ C ages are calibrated using the curves from Stuiver and Pearson (Stuiver and Pearson 1993; Pearson and Stuiver 1993). The calibrated age ranges are shown in Table 2. The calibrated ages of the averaged results are summarized in Figure 1. The calibrated age of the  $^{14}$ C age is plotted as a black circle; the confidence intervals are shown as intermittent error bars. The known historical ages of the documents are shown in Figure 1 as dotted rectangles.

As can be seen in Figure 1, the calibrated ages of ancient documents are in good agreement with their corresponding historical ages, except for samples 2, 10 and 11. Samples 1–9 have different sources, i.e., from a Buddhist sutra, a scattered piece of sutra or a scroll. But their use has been for writing historical information. In Japan, paper has been made mainly from Kouzo (Broussonetia Kazinoki Sieb.), a type of deciduous shrub. The trunk grows to ca. 4 m in height and ca. 2 cm in diameter. Japanese people normally harvested branches of Kouzo that were 3–15 yr old. In addition, new twigs grown under a year old were harvested selectively for manufacturing Japanese paper. (Older twigs yield paper of poor quality.) Because of this selection, there is no "old wood" effect. Such an effect indeed is not apparent in Figure 1, except for sample 2. The calibrated age of this sample is older than the historical age. Although Japanese paper has been normally made from twigs of Kouzo, it is known that there are some kinds of paper manufactured from waste paper. Such recycled paper is called Syukusi or Usuzumigami. Usuzumigami is usually slightly blackish due to incomplete bleaching. It would be possible to consider sample 2 to be recycled paper.

The measured age of sample No. 10 is ca. 300 yr too old. This sample had been used to reinforce a sutra by attachment to the back side. If brand-new paper is used for repairing a damaged ancient document, the document will become torn or creased due to the shrinking of the new paper. Therefore, old paper is commonly used for repair to prevent distortion. For this reason, the calibrated <sup>14</sup>C age of the reinforcing paper is older than the historical age when the sutra was written.

The calibrated age of sample No. 11 is a little younger than that of sample No. 10. Sample No. 11 was manufactured from sample No. 10 in the later part of this century. The difference in calibrated ages can, therefore, be caused by contamination of modern carbon in manufacturing.

TABLE 2. Results of <sup>14</sup>C Age Measurements on Ancient Japanese Documents

	-0	δ <sup>13</sup> C	Calibrated age
Sample no.	<sup>14</sup> C age (BP)	(‰)	(cal AD)*
1	1000 ± 38 916 ± 41 Average 958 ± 28	-26.3 -26.3	1011(1022)1036 1038(1069,1071,1129,1131,1160)1186 1026(1038)1055,1083()1122,1138()1157
2	1018 ± 42 912 ± 58 Average 965 ± 36	-26.0 -26.0	996(1017)1030 1032(1161)1216 1022(1034)1055,1083( )1122,1138( )1157
3	958 ± 49 869 ± 38 879 ± 43 Average 902 ± 25	-26.1 -26.0 -26.1	1020(1038)1163 1162(1198)1226 1060()1079,1125()1135,1158(1177)1224 1054()1084,1122()1138,1156(1164)1184
4	744 ± 56 743 ± 44 687 ± 38 664 ± 68 Average 709 ± 26	-26.6 -26.6 -26.5 -26.5	1252(1283)1296 1265(1283)1294
5	571 ± 64 484 ± 56 Average 527 ± 43	-25.5 -25.5	1308( )1358,1381(1402)1428 1410(1435)1450 1402(1417)1435
6	728 ± 73 681 ± 46 446 ± 40 434 ± 83 Average 571 ± 32	-25.5 -25.5 -26.0 -26.0	1252(1286)1304,1370( )1370 1286(1297)1309,1356( )1383 1434(1445)1470 1422(1448)1515,1591( )1621 1325( )1336,1394(1402)1411
7	807 ± 35 779 ± 39 Average 793 ± 26	-26.1 -26.1	1219(1248)1278 1228(1276)1285 1229(1261)1279
8	$740 \pm 76$ $732 \pm 30$ $656 \pm 51$ $620 \pm 65$ Average $687 \pm 29$	-26.4 -26.5 -26.5 -26.4	1231(1284)1302 1279(1286)1293 1291(1303)1325,1335()1394 1296(1315,1346,1391)1407 1289(1296)1303
9	904 ± 21 842 ± 22 Average 873 ± 15	-26.0 -26.0	1057()1081,1123()1137,1157(1164)1173 1206(1221)1232 1167(1189)1216
10	551 ± 38 540 ± 66 534 ± 43 530 ± 42 Average 539 ± 24	-25.3 -25.0 -25.0 -25.3	1398(1407)1425 1323()1338,1394(1410)1437 1401(1413)1433 1402(1415)1434 1404(1411)1425
11	453 ± 48 403 ± 39 Average 428 ± 31	-25.6 -25.6	1430(1443)1471 1445(1471)1509,1602( )1615 1441(1449)1475

<sup>\*</sup>Values in parentheses are the calibrated ages of the mean <sup>14</sup>C ages. Values outside parentheses are error ranges.

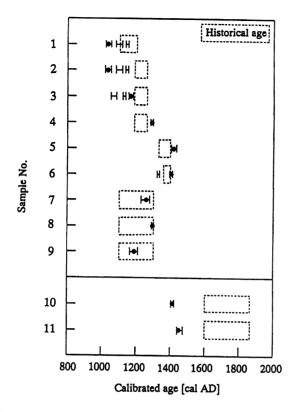


Fig. 1. Comparison of calibrated <sup>14</sup>C age and historical age of ancient Japanese documents

#### CONCLUSION

We used AMS to measure the <sup>14</sup>C ages of ancient Japanese documents and compared the results with known historical ages. We conclude that such documents are suited for <sup>14</sup>C age measurement for the recent historic period because of the negligible difference between the calibrated and historical ages.

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### REFERENCES

Kitagawa, H., Masuzawa, T., Nakamura, T. and Matsumoto, E. 1993 A batch preparation method of graphite targets with low background for AMS <sup>14</sup>C measurements. *Radiocarbon* 35(2): 295-300.

Nakamura, T., Nakai, N., Sakase., T., Kimura, M., Ohishi, S., Taniguchi, M. and Yoshioka, S. 1985 Direct detection of radiocarbon using accelerator techniques and its application to age measurement. *Japanese Journal of Applied Physics* 24: 1716–1723.

Nakamura, T. and Nakai, N. 1988 Fundamentals of radiocarbon dating. *Memoirs of the Geological Society of Japan* 29: 83-106.

Oda, H. 1994 A study of correction for carbon isotopic

fractionation in <sup>14</sup>C/<sup>13</sup>C ratio measurement with AMS. Summaries of Research Using AMS at Nagoya University 5: 244–251.

Stuiver, M. and Pearson, G. W. 1993 High-precision bidecadal calibration of the radiocarbon time scale, AD 1950-500 BC and 2500-6000 BC. In Stuiver, M., Long, A. and Kra, R. S., eds., Calibration 1993. Radiocarbon 35(1): 1-23.

Pearson, G. W. and Stuiver, M. 1993 High-precision bidecadal calibration of the radiocarbon time scale, 500-2500 BC. In Stuiver, M., Long, A. and Kra, R. S., eds., Calibration 1993. Radiocarbon 35(1): 25-33.