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# High-precision age determination of Holocene samples by radiocarbon dating with accelerator mass spectrometry at Nagoya University

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ABSTRACT

Age determination with high accuracy as well as high resolution is a principal procedure in the research of Quaternary processes, such as volcanic eruptions, lacustrine and ocean sedimentation, and recent fault activation, for investigating past global environmental changes and predicting the realistic changes in the near future. Among several dating techniques applicable to late Quaternary samples, radiocarbon dating has been commonly used since 1950, although its application is limited to ages younger than ca. 50 cal ka BP. In particular, recent developments of accelerator mass spectrometry (AMS)-related techniques have opened a variety of applications. A Tandetron AMS system (Model 4130-AMS, HVE, B.V., the Netherlands) introduced in 1996/97 at Nagoya University, still has excellent performance, and this device has been used frequently for $^{14}C$ dating of late Quaternary samples. Here, we describe briefly the AMS $^{14}C$-measurement system of Nagoya University and then focus on the following four applications for Japanese Holocene samples: i) $^{14}C$ dating and dendrochronological analysis of wood samples excavated from archeological sites; ii) $^{14}C$ dating analysis of individual annual rings of a huge tree to compare the $^{14}C$ age variations with the IntCal13 dataset; iii) research on the carbon reservoir effects of marine samples around the Japanese Archipelago; iv) potential application of a new $^{14}C$ landmark for precise calendar age estimation of tree rings.



## 1. Introduction

Cosmogenic nuclides in the environment are commonly used as dating tools. In particular, application of accelerator mass spectrometry (AMS) has enabled investigations on precise age determination by high-accuracy measurements of cosmogenic nuclides in small samples. Age analysis by radiocarbon ($^{14}C$) dating of specimens such as lacustrine sediments, marine samples, cave speleothems, tephra layers, annual rings in buried woods, ancient paper documents, and cotton and silk cloth of ancient and medieval periods is commonly used for geological and archeological investigations. $^{14}C$ wiggle-matching analysis of tree rings and stably accumulated sediment layers is useful for estimating the precise age of historical, climatic, and environmental events (Skog and Regnell, 1995; Imamura et al., 2007). Studies of variations in the past marine $^{14}C$ reservoir and ventilation ages give us information on the changes in ocean circulation (e.g., upwelling and horizontal current movements), in addition to the precise calendar age of marine samples (Stuiver and Braziunas, 1993; Ikehara et al., 2011). Tephra layers in marine sediments provide excellent samples for estimating $^{14}C$ reservoir ages (Ikehara et al., 2011). Temporal variability in the dead carbon fraction of cave speleothems together with associated stable isotopes and trace elements also delivers useful information on past climate change and soil-to-speleothem carbon transfer dynamics (Southon et al., 2012). Understanding the temporal variations in $^{14}C$ reservoir effects can improve the $^{14}C$ dating method (Reimer et al., 2013).

The application of $^{14}C$ dating to late Quaternary samples has become quite popular for the following reasons: (1) only a very

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small amount of carbon, around 1 mg or less, is required for $^{14}C$ dating with AMS; (2) uncertainties of $^{14}C$ ages are normally from ±20 to ±40 $^{14}C$ years for samples younger than 10 ka BP; (3) highly reliable calendar dating of samples is now possible by calibrating $^{14}C$ age by selectively applying international calibration datasets, such as IntCal13, SHCal13, and Marine13 (Reimer et al., 2013), depending on the sample origin; (4) $^{14}C$ ages of marine samples in particular are reasonably corrected by using the Marine13 dataset, although a realistic correction for the local carbon reservoir effect remains an obstacle; and (5) statistical models of $^{14}C$ data analysis based on the Bayesian approach, such as wiggle matching, boundary, and sequence analyses, are now widely applicable (Bronk Ramsey, 2009). These advantages have popularized the application of AMS $^{14}C$ dating for late Quaternary samples worldwide, and the number of AMS laboratories for $^{14}C$ measurement has increased to more than 100 across the globe. Most of the application programs described above are not specific to AMS $^{14}C$ dating but applicable to $^{14}C$ ages measured by the conventional counting method, as well as other dating methods such as U-series, OSL, $^{10}Be$-based surface exposure age, etc. (Bronk Ramsey, 2009).

In this article, we first provide a brief summary of $^{14}C$ dating performances with AMS at Nagoya University and then discuss applications of $^{14}C$ dating analysis to domestic wood samples excavated at Japanese archeological sites and the $^{14}C$ calibration problems and carbon reservoir effect of marine samples, and finally we introduce newly-detected $^{14}C$ spikes recorded in tree rings by huge solar-flare events for the application of precise calendar age estimation of wood samples.

## 2. $^{14}C$ dating with the Nagoya University Tandetron AMS

In Japan, the following nine laboratories are performing $^{14}C$ dating studies in their own ways: Mutsu Establishment, Japan Atomic Energy Agency (JAEA) (Suzuki et al., 2004); Yamagata University (Tokanai et al., 2013); Institute of Accelerator Analysis, Ltd.; National Institute for Environmental Studies (Yoneda et al., 2004); Paleo-Lab, Ltd. (Kobayashi et al., 2007); MALT group (Matsuzaki et al., 2004) and the Atmosphere and Ocean Science Institute (Miyairi et al., 2013) of University of Tokyo; Tono Geoscience Center, JAEA (Saito-Kokubu et al., 2013); and Nagoya University (Nakamura et al., 2004).

In 1981/82, a Tandetron AMS system (Model 4130-AMS) manufactured by General Ionex, Corp., United States was installed at Nagoya University as one of five products delivered worldwide, and routine $^{14}C$ measurements began in 1983 (Nakai et al., 1984; Nakamura et al., 1985). An additional AMS system (Model 4130-AMS) dedicated to $^{14}C$ measurements, built by the High Voltage Engineering Europa (HVE), B.V., the Netherlands, was delivered to Nagoya University in 1996/97. Acceptance tests of its performance for carbon isotope measurements were completed in January 1999, and routine measurements began in mid-2000. The machine still shows the performance that the standard deviation (one sigma error) of the $^{14}C/^{12}C$ ratio is around ±0.2% to ±0.4% (estimated including reproducibility tests with more than six HOxII reference targets measured in the same runs, and a bit larger than the uncertainty of about ±0.2% calculated by $^{14}C$ counting statistics for individual targets) and that of the corresponding $^{13}C/^{12}C$ ratio is ±0.03% to ±0.07%, as tested for HOxII targets (Nakamura et al., 2004).

## 3. Applications of $^{14}C$ dating with the Nagoya University Tandetron AMS

Below, we describe four recent applications of $^{14}C$ dating for Holocene samples with the Tandetron AMS system at Nagoya University. All the samples described here are from Japan, as shown in Fig. 1.

### 3.1. *$^{14}C$ dating of trees and dendrochronology*

In some cases, excavation of archeological sites located in wetlands has revealed a huge number of wooden artifacts used by ancient peoples as materials for the construction of residential houses as well as wooden tools. To determine chronology, wood materials are normally analyzed by dendrochronology if disk samples can be easily collected from them. If the tree-ring age range covered by the wood samples does not correspond to the master sequence of annual ring width patterns for any particular wood species, precise age estimation of the woods is not possible. However, cross-dating analysis of wood samples excavated at the sites could be possible. More than 80 wooden poles excavated at the Aota site, Niigata Prefecture, Japan, were grouped temporally into two parts, and tree-ring age relations were established for each group by Kimura (2012) by comparing the ring-width data for almost all wood poles collected. Kimura (2012) estimated the age difference of the two groups to be 91 years by cross-dating the ring-width patterns of the two groups by connecting them to a ring-width pattern of one natural tree that seemed to cover the full age range extended by the two groups. Very recently, by applying $\delta\ ^{18}O$ patterns of the cellulose fraction from annual rings more effectively, instead of ring width, Nakatsuka et al. (2004) and Nakatsuka and Sano (2014) successfully determined the age of the two groups precisely. The age of cutting the older trees in group A was estimated to be about 530 BC, and that of the younger trees in group B was about 477 BC. These results will be described elsewhere in detail.

We also conducted cross-dating of wooden poles excavated at the Mawaki site, located in the middle of the Noto Peninsula. In the

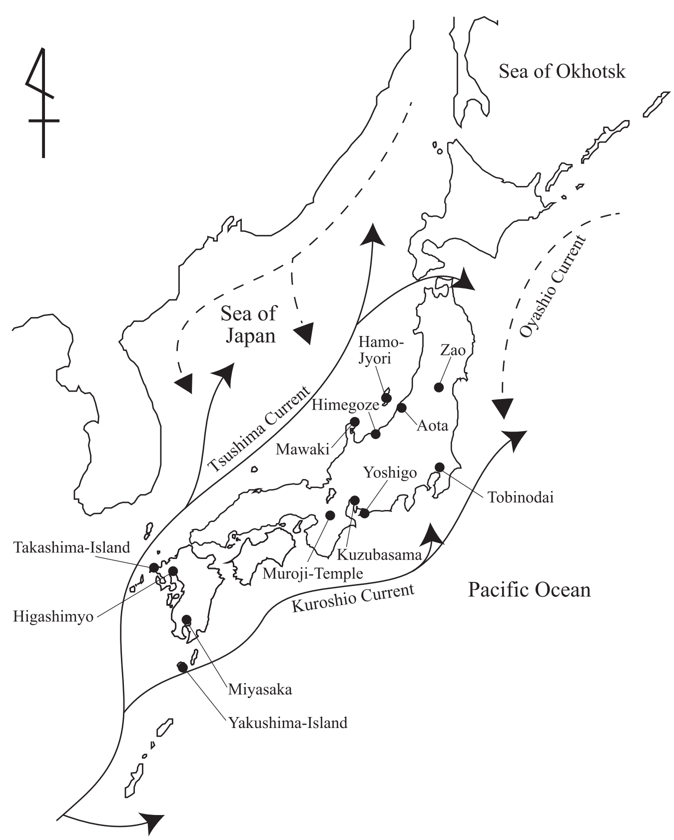


**Fig. 1.** Map showing locations of collecting the samples described in this text.