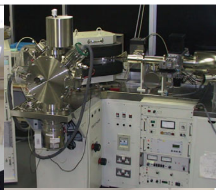
Division for Chronological Research


- Anthropogenic history and geochronology
- Accelerator mass spectrometry
- Electron probe microanalysis
- Paleoclimate reconstruction and future Earth
- Geosphere stability
- Isotope geoenvironmental chemistry
- CHIME dating
- Development of new analytical methods

Short- and long-term forecasts of global environmental changes and their countermeasures are issues of great urgency. Determining when an event occurred in the past, via “dating,” is of importance understanding present and predicting future states of the Earth. We promote chronological studies on a broad range of subjects from events in Earth’s history, spanning 4.6 billion years, to archaeological materials, cultural properties, and modern cultural assets. The Tandetron dating group conducts interdisciplinary research involving radiocarbon (^{14}C) dating using accelerator mass spectrometry to understand changes in the Earth’s environment and the cultural history of humankind from about 50,000 years ago to the present day. In addition, the group studies near-future forecasts of Earth and Space environments, focusing on spatiotemporal variations in cosmogenic nuclides, such as ^{14}C and ^{10}Be, and conducts research that integrates art and science through collaboration between researchers in archeology, historical science, and other fields. The microscale spatial dating group uses the chemical U-Th total Pb isochron method (CHIME), which was firstly developed at Nagoya University, to shed light on events in Earth’s history from its formation 4.6 billion years ago up to approximately 1 million years ago. An electron probe microanalyzer (EPMA) have been used to perform nondestructive microanalyses of rocks and other materials to reveal records of complex events recorded in zircon, monazite, and other samples.

Main Activities in FY2019

Reconstruction of paleoclimate in West Asia

Our goal is to reconstruct the paleoclimate of West Asia over a long period of time (from tens of thousands of years ago to several thousands of years ago), based on ^{14}C dating and isotopic analysis of stalagmites and travertines in Iran. This was in answer to a call for studies in the new academic field “The Essence of Urban Civilization: An Interdisciplinary Study of the Origin and Transformation of Ancient West Asian Cities,” conducted in collaboration with the University of Kurdistan (Iran), the University of Tsukuba, and the University of Toyama. In recent years, past fluctuations in precipitation were reconstructed from chemical analyses of stalactites, showing that precipitation characteristics change notably with climate change. However, there are few studies on the reconstruction of the paleoclimate in West Asia (especially in Iran and Iraq), and how precipitation characteristics changed in this region owing to past climate change is not well understood. In the current fiscal year, we performed ^{14}C dating, elemental analysis, and isotopic analysis on travertine samples and spring waters in the Zendan-e Soleyman and Baba Gurgur, and examined whether travertine recorded paleoclimatic information. Furthermore, to identify fluctuations in the dust component that impacts elemental and isotopic fluctuations in travertine, we visited Iran in October 2019 to set up a high-volume air sampler at the University of Kurdistan, and began sampling atmospheric aerosols (PM10).

Sampling of travertines from Zendan-e Soleyman, Iran.

^{14}C dating and multi-Sr isotopic analysis of bioapatite from cremated bones

In archeological research targeting excavated bones, the organic components of the bones, the collagens, are generally used. Using isotopes of its main components, carbon, nitrogen, and oxygen, samples can be dated, and diet studied. However, the method cannot be used on cremated bones that have been through high temperatures at which the organic matter has decomposed. The use of inorganic chemicals as an index is necessary. In the present study, we have focused on bioapatite, which is the inorganic component of bones. It was the first systematic high-precision multi-Sr analysis – radiogenic Sr isotope analysis ($^{87}Sr/^{86}Sr$) and stable Sr isotope ratio ($^{88}Sr/^{86}Sr$, $\delta^{88}Sr$) – of cremated bones used in addition to ^{14}C analysis. The samples were cremated bones of Jokei (AD1155–1213) stored in an urn excavated from under the Gorin Tower in Jisho-in, Nara Prefecture, and cremated bones (600–900 BP) excavated from the Binman-ji burial site in Shiga Prefecture. First, we showed that the amount of Ba in the cremated bones was a good index for the quantitative analysis of secondary alterations that the bones went through during burial in the soil. Then, we selected bone fragments with low amounts of Ba and limited secondary alterations for high- precision ^{14}C dating (Minami et al., 2019; Radiocarbon). We also performed multi-Sr isotopic analysis and showed that the Jokei was likely a vegetarian, while ancient people buried at Binman-ji had a meat-heavy diet: their trophic levels differed. This result is a breakthrough as it shows that the stable Sr isotope ratio of bioapatite in cremated bones is a new and useful indicator of dietary analysis.

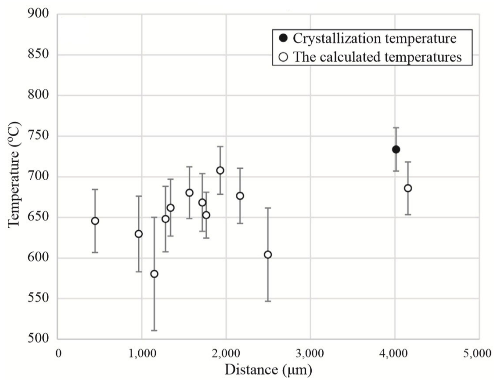
Binman-ji burial site in Shiga Prefecture.

Development of EPMA quantitative analysis of trace titanium (Ti) in quartz and application to estimate the cooling process of granitic magma

Quartz, silicon dioxide, may contain various trace elements. The Ti concentration of quartz reflects its crystallization temperature and the activity of Ti in the system; quartz crystallized at a higher temperature has a higher titanium concentration.

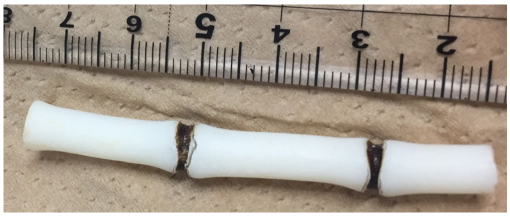
If the titanium concentration in a microvolume of a quartz grain is measured, the details of crystal growth and temperature change can be estimated, leading to elucidation of the formation process of the rock that contained the grain. A problem with this method is that, as Ti concentration becomes more sensitive at lower temperatures, for quartz not crystallized under super high temperatures, accuracy in addition to precision is required. Thus, we developed a method to measure the titanium concentration in an area to the order of microns, using accurate and non-destructive EPMA.

Since a method has already been developed to measure titanium at 10 µg/g or less in zircon, with a relative error of 10% or less (2σ) (Yuguchi et al., 2016), we applied this method to the quartz. However, as quartz is easily damaged by electron bombardment, we determined a new beam diameter and X-ray counting method suitable for quartz. We then applied this method to Toki Granite in Gifu Prefecture and estimated the crystallization temperature of the quartz. We revealed that the quartz crystals were formed in the range from 600°C to 750°C. The zonal structure observed by cathodoluminescence imaging reflected the temperature and diffusion of titanium in granitic magma. As such, it was shown that quartz that displays oscillatory zoning in cathodoluminescence imaging formed under a conditions of slow cooling.



Distance from the quartz grain boundary and estimated crystallization temperature (Yuguchi et al., 2020).

Elucidating the growth process of axial internode skeletons in deep-sea isidid octocorals

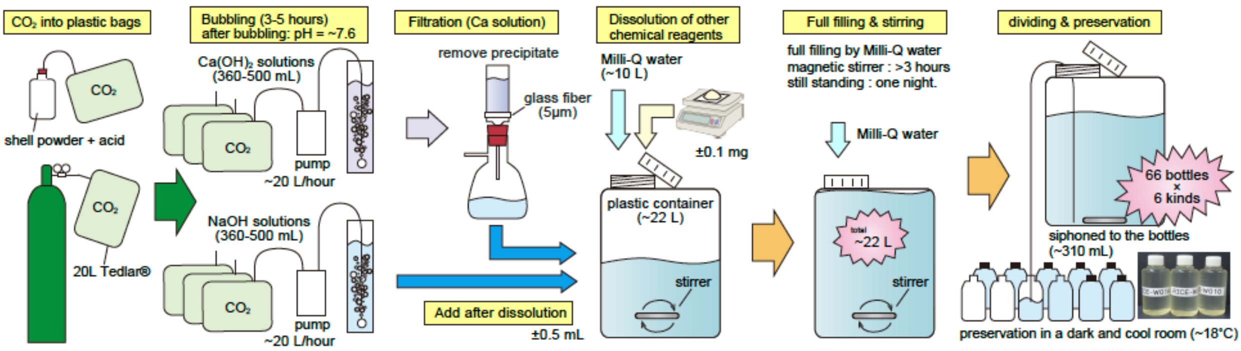


Isidid octocorals used for analysis.

The calcareous skeletons of deep-sea corals are being used as an archive of paleooceanic information; however, as growth processes differ depending on classification, it is necessary to understand the growth processes and growth rates for palaeoceanographic research. The isidid octocorals consist of elongated calcitic skeletons with hollow structures in the center. The skeletons of corals have annual rings, and their central axis is a thin opening. It was assumed that the center was old, and the outer part was young, but a previous study of ^{14}C dating reported that the age was younger in the center in multiple samples. It is unknown if the cause of this rejuvenation is growth from the center or a change in the age of the body of water in contact with the specimen. If there are two formation processes for the skeleton, the crystal orientation is likely different. Thus, in the present study, we performed a crystal orientation analysis of a thin section of isidid axis using electron backscatter diffraction. The crystal orientation analysis of the isidid bone axis showed that the whole specimen was structured with uniform regularity. This result supports the idea that skeleton formation only occurs at the surface of the bone axis. If there was no reversal of growth direction, ^{14}C dating of isidid skeletons can be used as a proxy for the age of water masses.

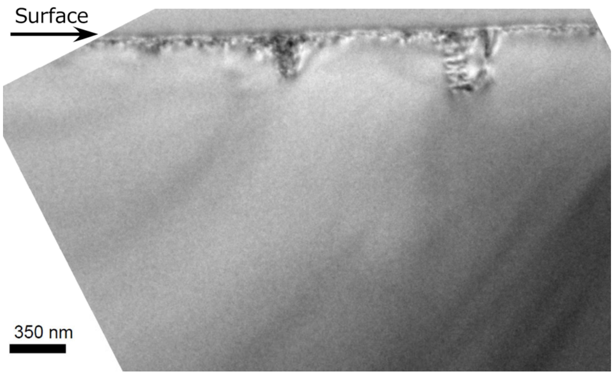
Preparation of water sample for the ^{14}C inter-comparison program

Since 2013, our research group, jointly with National Institute of Advanced Industrial Science and Technology, is conducting a basic verification to prepare for the full-scale operation of the ^{14}C analysis inter-comparison program for dissolved inorganic carbon in water samples (RICE-W: Radiocarbon Inter-comparison on Chemical Experiments, Water series). The important part of the inter-comparison verification is that water samples with the same ^{14}C concentration are distributed to each facility and that the ^{14}C concentration does not change from the time of distribution to the time of analysis. In this study, we verified what the containers and water samples meet these conditions well. We examined three types of containers made of glass, PAN resin, and PP resin. It was concluded that the PAN resin container was ideal because it showed superior gas barrier performance, and the handling such containers is convenient. In case of a natural sample, the effect of biological activity cannot be ignored. Thus, the addition of toxins such as $HgCl_2$ is necessary (Takahashi et al., 2018, NIMB). We abandoned the preparation of inter-comparison samples based on natural samples, and instead prepared inter-comparison samples that were completely and artificially controlled. We mixed some reagents to prepare a set of inter-comparison samples (RICE-W09-W14) with a chemical composition that mimics natural samples. The ^{14}C concentrations of RICE-W09-W14 covers the range from background to modern. We posted RICE-W09-W14 to some ^{14}C research facilities for ^{14}C comparison.



Preparation procedure for ^{14}C inter-comparison water sample (RICE-W09−W14).

Evaluation of surface damage by diamond polishing treatment



Transmission electron microscope image of olivine (bright field image).

In the correction calculation generally used in EPMA quantitative analysis, it is assumed that electrons enter the surface of a homogeneous sample that has been mirror polished orthogonally. For mirror polishing, 0.25–0.5 µm diamond or alumina is generally used. Usually, this method does not cause any issues in quantitative analysis. However, this polishing method is assumed to damage a fraction or half of a grain diameter on the sample surface. Thus, when the analytical area is shallow, it is possible that the analysis is performed on the damaged area instead of the actual sample. To assess the surface damage, we observed the surface of an olivine sample that had been polished with 0.25 µm diamond with a transmission electron microscope. We observed dislocation that was likely caused by polishing in the area dozens of mm from the surface. In some areas, there were dislocations of several hundred nm to 1 µm. Therefore, when using particle polishing, surface analysis must be performed, considering the possibility that the damage can extend beyond the grain diameter in some areas.

The 8th East Asia Accelerator Mass Spectrometry Symposium

The 8th East Asia Accelerator Mass Spectrometry Symposium was held in Sakata and Hirata Hall, Nagoya University from December 3 to December 6, 2019. EA-AMS is held every other year in Japan, South Korea, China, or Taiwan to present and discuss the development of measurement methods in the accelerator mass spectrometry of radiocarbon (^{14}C), iodine-129 (^{129}I), chlorine-36 (^{36}Cl), and so on, and their applications to environmental and Earth sciences. The 8th symposium was held jointly by the ISEE Division for Chronological Research and the JAEA Tono Geoscience Center. This symposium was attended by 115 accelerator mass spectrometry researchers from across the world, including East Asia (51 from Japan, 33 from China, eight from South Korea, and seven from Taiwan), Australia (four), the USA (three), Switzerland (three), and Poland (two). It started with an ice-breaker event on the evening of December 2nd, followed on the 3rd and 4th by presentations on technological developments, application research, and the current situations of facilities for accelerator mass spectrometry (40 oral presentations and 42 posters) along with discussions. On the 5th, there was a field trip to view an accelerator mass spectrometer, and on the final day (6th), there were oral presentations and a discussion on accelerator mass spectrometer research and future outlooks. There was meaningful information exchange and networking among accelerator mass spectrometry researchers.