

# 9-1. Research Divisions

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## Division for Chronological Research

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### Research topics and keywords

- Radiocarbon ( $^{14}\text{C}$ ) dating by Accelerator mass spectrometry (AMS)
- Developing radiocarbon ( $^{14}\text{C}$ ) pre-treatment and measurement techniques
- Analysis of cosmogenic nuclides
- CHIME (chemical U-Th total Pb isochron method)
- Microanalysis and spectroscopy
- Geochronology
- Isotope analysis

### Introduction to Division for Chronological Research

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, “dating,” is of great importance to predict future states of the Earth. Therefore, we conduct chronological studies on a broad range of subjects from events in Earth’s history spanning approximately 4.6 billion years, to archeological materials, cultural properties, and modern cultural assets. The Tandetron dating group conducts interdisciplinary research that involves radiocarbon ( $^{14}\text{C}$ ) dating using accelerator mass spectrometry (AMS) to understand changes in the Earth’s environment and the cultural history of humankind from approximately 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  $^{14}\text{C}$  and  $^{10}\text{Be}$ , and conducts research that integrates art and science through collaborations between researchers in archeology, historical science, and other fields. The micro-scale spatial dating group uses CHIME (the chemical U-Th total Pb isochron method), which was first developed at Nagoya University, to shed light on events in Earth’s history from the formation of Earth 4.6 billion years ago up to approximately 1 million years ago. With an electron probe microanalyzer (EPMA), non-destructive microanalyses of rocks and other samples are performed to reveal records of complex events recorded in zircon, monazite, and other materials.

### Main Achievements in FY2017

#### 1. Climate and tectonic history in the Levant

The area surrounding the Dead Sea, the Levant, was the locus of humankind’s migration out of Africa and, thus, has been a home for peoples since the Stone Age. Understanding the climate and tectonic history of this region provides valuable insights into archaeology and studies of human history, and helps in gaining a better picture of future climate and tectonic scenarios. An International Continental Scientific Drilling Program (ICDP) deep drilling project was performed in the Dead Sea between November 2010 and March 2011. The project was conducted by the ICDP and scientists in Israel, Germany, Japan, Norway, Switzerland, and the United States. We have worked on deciphering evidence of past climates and crustal deformation from collected sediment cores. AMS  $^{14}\text{C}$  dating of plant fossils from the upper 150 m of the sediment core revealed the occurrence time of climate and tectonic movement events over the past 50,000 years. We explored the relationship between these findings and the migration of early modern humans (*Homo sapiens*).



Drilling, observation and sampling in the Dead Sea; drilling at a water depth of 300 meters.

## 2. Small-scale $^{14}\text{C}$ dating of sediment core drilled from off Zambezi River, Africa

International Ocean Discovery Program (IODP) Expedition 361 drilled six sites at the southeast African margin and in the southwest Indian Ocean using a D/V JOIDES Resolution, from January to March 2016. Site U1477 was located at the continental slope of the western Mozambique Channel near the Zambezi River delta, and the sediment at Site U1477 is mainly composed of silty clay, with an abundance of planktonic foraminifer of less than 1%. The small-scale  $^{14}\text{C}$  dating (~80 micrograms of carbon) results for planktonic foraminifer from the U1477 core indicate that the sedimentation rate (~1.2 m/kyr) of this core marks it as a very early sediment, and the age of the lowermost part of the core (~181 m) is approximately 150 ka. In future research, high-resolution reconstructions of African climate and environmental changes over the past 150,000 years will be obtained.



D/V JOIDES Resolution.

## 3. Reconstruction of hydrological variability during the late last glacial period from Lake Baikal sediment in southern Siberia

We reconstructed the hydrological variability (river input and chemical weathering intensity) at Lake Baikal in southern Siberia, using two independent sedimentological and geochemical proxies. This is a key region for bridging the spatial gap in climate reconstructions from the late glacial period. We found that the millennial-scale hydrological variability during the late last glacial period (31–11.5 cal ka BP) is associated with solar activity changes, with wet (dry) climate conditions in the Siberian region corresponding to solar maxima (minima). Millennial-scale biological responses to hydrological changes resulting from solar activity during the last glacial period were also observed. Our results indicate that solar-induced millennial-scale hydroclimate variability appears to control Eurasian biological change during the late last glacial period.



Lake Baikal and the terminal facet.

## 4. Chemical characteristics of $^{10}\text{Be}$ in bed-sediments of rivers flowing into Lake Biwa

We investigated the chemical characteristics of meteoric  $^{10}\text{Be}$  extracted from bed-sediments in the Yasu and Ado rivers flowing into Lake Biwa, central Japan. The sediments were sieved into 5 grain-size fractions (180–150  $\mu\text{m}$ , 150–125  $\mu\text{m}$ , 125–63  $\mu\text{m}$ , 63–32  $\mu\text{m}$ , and <32  $\mu\text{m}$ ), and each of the fractions was sequentially leached into three phases: exchangeable, amorphous oxy-hydroxide, and crystalline oxy-hydroxide phases. The results were as follows: 1) most  $^{10}\text{Be}$  (~95% of the total  $^{10}\text{Be}$ ) was located in the amorphous and crystalline oxy-hydroxide phases of bed-sediments for all grain size fractions; 2) the  $^{10}\text{Be}$  concentrations of all the leachates decreased with increasing grain size; 3) the bed-sediments from upper streams showed higher  $^{10}\text{Be}$  concentrations than those from lower streams; and 4) meteoric  $^{10}\text{Be}$  concentrations in river bed-sediments is strongly dependent on river water pH as well as grain size.



Upstream (left) and downstream (right) parts of the Ado river.

## 5. Cultural history of PaleoAsia

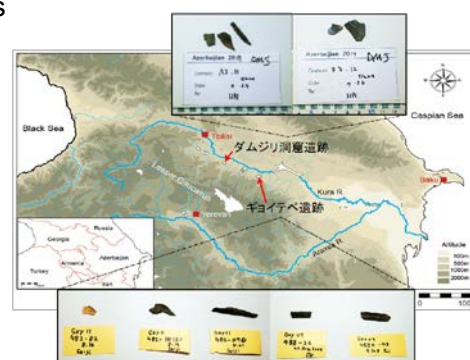
Modern humans (*Homo sapiens*) originated in East Africa approximately 200,000 years ago and migrated to the Eurasian continent while adapting to a diversified environment in Asia some 50,000 years ago, subsequently replacing more primitive species including Neanderthals. We conducted an interdisciplinary research project to understand the Cultural History of PaleoAsia (supported by Scientific Research on Innovative Areas, MEXT Grant-in-Aid Project FY2016–2020). We are advancing field studies of ruins and wetlands (Vietnam, Mongolia, Oman, Iran, Jordan, Pakistan) to explore the residence environments and lifestyles of early modern humans and past climate conditions.



Excavating cave ruins in Iran.

## 6. Radiocarbon dating of carbonate hydroxyapatite in fossil bones

We are aiming for accurate  $^{14}\text{C}$  dating of carbonate hydroxyapatite (CHA), a bone inorganic component, in cremated bone with little remaining organic carbon from collagen. This year, we investigated seven burned bones collected from the Göytepe and Damjili archeological sites in Azerbaijan. The bone samples had low CHA crystallinity, and  $^{14}\text{C}$  dates ~400 (Göytepe) and 1000–1700 (Damjili) years younger than expected. The results indicate that the bones had been burned at a low temperature (<600°C), that they were heavily contaminated through diagenetic alteration during burial because of low CHA crystallinity, and that the degree of contamination from foreign carbon into the bones varied with burial conditions.



Burned bone samples from Azerbaijan.

## 7. Radiocarbon dating of the famous ancient sutras written in Asuka and Nara periods

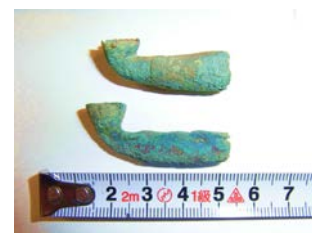
We measured the radiocarbon ages of six famous ancient sutras written in the Asuka and Nara periods: Myohorenge kyo attributed to Nakatomi no Kamatari (AD614–669), Gogatsu Tsuitati kyo (AD770), Uokai kyo (AD770), Hyakumanntou Darani (AD764–770), Jishinin Darani (AD764–770), and Todaiji Nigatsudo Yakegyo (AD729–767). The calibrated radiocarbon ages of six samples excluding Myohorenge kyo and Jishinin Darani were consistent with their paleographical ages. In particular, Hyakumanntou Darani was shown to be the oldest print in the world. If Myohorenge kyo was truly written by Nakatomi no Kamatari, it would be the oldest existing sutra; however, its calibrated radiocarbon age was 1299–1404 cal AD and, hence, this theory was refuted by the radiocarbon dating. Jishinin Darani had been known as one of the oldest existing prints; however, radiocarbon dating indicated that Jishinin Darani was printed in 1527–1656 cal AD and, thus, is not one of the oldest prints.



Hyakumanntou Darani.

## 8. Radiocarbon dating to bronze implement

Radiocarbon dating is a useful method for samples that contain carbon derived from atmospheric  $\text{CO}_2$ . Verdigris,  $\text{CuCO}_3 \cdot \text{Cu}(\text{OH})_2$ , is rust produced on bronze implements, formed from Cu in bronze and  $\text{CO}_2$  from the atmosphere. When verdigris is initially formed, it produces a close film and limits generation of new rust and, therefore, verdigris should preserve atmospheric carbon from its time of formation. First, we improved the carbon extraction method for  $\text{CuCO}_3 \cdot \text{Cu}(\text{OH})_2$ , then applied the method to archaeological samples of known age, measured the radiocarbon ages and combined the verdigris radiocarbon dating samples to demonstrate that verdigris is a suitable medium for radiocarbon dating.



Bronze pipe sample excavated from Hekirichi Jin'ya site, Hokuto.

### 9. CHIME monazite age of Sanbagawa metamorphic rock

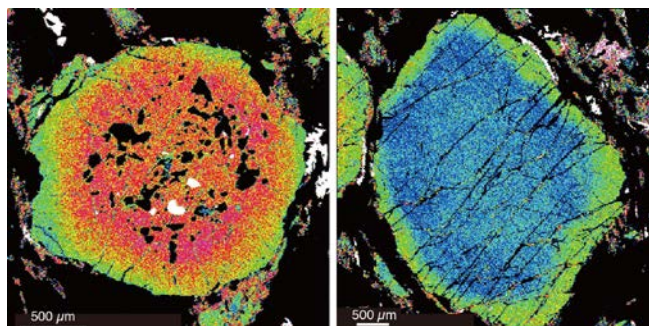
The Sanbagawa metamorphic rocks of epidote–amphibolite facies are extensively exposed in Nushima at the southern end of the Hyogo Prefecture, Japan. CHIME ages were determined for monazite grains from the Sanbagawa metamorphic rocks. CHIME monazite ages are  $92.3 \pm 3.2$  and  $90.3 \pm 4.7$  Ma for two chlorite–rich rocks collected from the reaction zone between pelitic schists and serpentinites. The monazite grains occur as isolated phases or aggregates with rutile/ilmenite in chlorite-rich matrix, and thus the CHIME monazite ages are interpreted to represent recrystallization close to the peak stage of prograde epidote-amphibolite facies metamorphism. This study is a first report of U-Pb age of the Sanbagawa epidote-amphibolite facies stage.



Monazite-bearing reaction zone between pelitic schists and serpentinites.

### 10. Zonal structures of garnet implying material mixing during prograde metamorphism

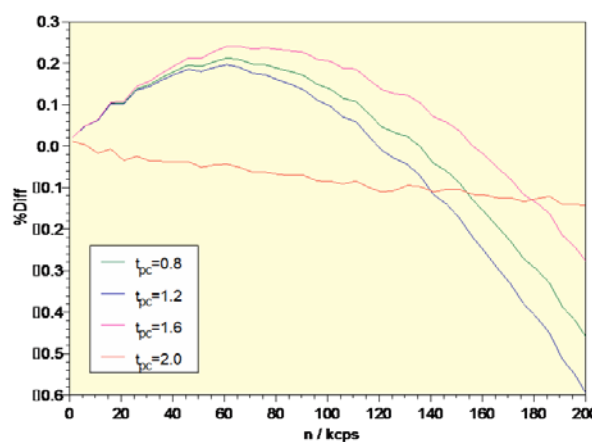
Kyanite-quartz eclogites, which were recrystallized under metamorphic conditions of 2.3–2.4 GPa/675–740°C, represent the deeper part of the Sanbagawa subduction zone. Garnet grains in the quartz eclogites are grouped into four types according to the compositional trends of their cores and these different type-zoned garnet grains sometimes coexist in a thin section. Central parts of these garnet grains usually show different compositional trends to each other, and their marginal parts share a similar compositional trend, implying that the equilibrium whole-rock compositional system changed during subduction metamorphism. This suggests that, in some cases, material interaction promoted by mechanical mixing probably controls mineral reactions and modification of mineral paragenesis of high-pressure metamorphic rocks.



Coexisting garnet grains showing different types of zonal structure.

### 11. Accurate dead time correction in quantitative electron probe microanalysis (EPMA)

A pseudo-fixed dead time circuit with a non-extendable approximation was developed to prevent systematic errors in quantitative electron probe microanalysis (EPMA) caused by inappropriate dead time corrections. EPMA of trace elements using wavelength dispersive spectrometers (WDS) requires high probe current and/or accelerating voltage. Therefore, the X-ray count rate of a standard material becomes extremely high when measured under the same conditions. Consequently, the accuracy of the dead time correction is a significant issue for quantification. The designed circuit suppresses the systematic error of the dead time correction to 0.6% or less when the true X-ray count rate is less than 200 kcps.



Monte Carlo simulation results of percentile errors.