# **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 urgent issues. Determining when an event occurred in the past, via "dating," is important for understanding the present state of the Earth and predicting its future. We promote chronological studies on a broad range of subjects, from events in Earth's history, spanning 4.6 billion years, to archeological materials, cultural properties, and modern cultural assets.

The Division for Chronological Research conducts interdisciplinary research involving radiocarbon (<sup>14</sup>C) dating using accelerator mass spectrometry to understand changes in the Earth's environment and the cultural history of humankind from approximately 50,000 years ago to the present day, as well as research on and development of new <sup>14</sup>C analyses and dating methods. In addition, the laboratory studies near-future forecasts of Earth and space environments, focusing on spatio-temporal variations in cosmogenic nuclides, such as <sup>14</sup>C and <sup>10</sup>Be, and conducts research that integrates art and science through collaborations between researchers in archeology, historical science, and other fields. Furthermore, using the chemical U-Th total Pb isochron method (CHIME), which was first developed at Nagoya University, and radiometric dating of long-half-life radioisotopes (Sr-Nd-Hf), we have shed light on events in Earth's history from its formation 4.6 billion years ago to approximately 1 million years ago. An electron probe microanalyzer (EPMA) has 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 FY2021

### Development of carbon-14 detection technique by PIMS

Carbon positive-ion mass spectrometry (C-PIMS) is an effective method for detecting radiocarbons at low concentrations. In this new radiocarbon detection technology, an electron cyclotron resonance ion source (ECRIS) is used

to generate highly charged positive ions (e.g.,  $C^{2+}$ ), which are converted into negative ions (C-) in a charge exchange cell. The development of high-performance ECRIS is important for the practical application of C-PIMS. At the ISEE, we are conducting theoretical and experimental studies on whether a small ECR ion source using a solid-state 2.45-GHz microwave generator is a viable option for the practical application of C-PIMS. Theoretical estimates predict that the generation of a 40-keV C<sup>2+</sup> ion beam of approximately 40 mA will enable the detection of natural radiocarbons.

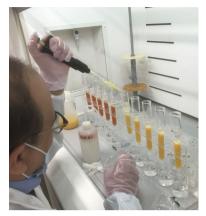


Beam injection system of PIMS.

## Model construction of long- and short-term structural evolution of Zagros orogenic belt

Understanding the igneous activity and formation process of the Zagros orogenic belt in Western Asia, which is part of the Alpine–Himalayan orogenic belt, is important for understanding the formation of the Earth's continental crust and for

analyzing the formation process of resource deposits. This year, we invited Prof. Azizi from the University of Kurdistan, Iran to Nagoya University for three months, from November 2021 to February 2022, to research the long-term evolution of long-lived radioisotopes (Sr-Nd-Hf) (Azizi *et al.*, 2021, *Journal of Petrology*; Daneshvar *et al.*, 2021, *Minerals*; Nouri *et al.*, 2021, *Lithos*; Gholipour *et al.*, 2021, *Lithos*). We also initiated an effort to develop a new structural development history model by adding a short time series of light elements such as Li and Be. The relatively short-lived cosmic ray-produced nuclide <sup>10</sup>Be can be useful for clarifying isotope recycling in subduction zones and post-impact systems. In 2021, we set up columns of cation and anion exchange resins for Be separation and conducted Be extraction from 40 rock samples, for which chemical and Sr-Nd-Hf isotopic ratio data were obtained.



Column separation of rock samples for extraction of Be fraction.

# Formation process of gigantic whale bone concretions in the Oga Peninsula, Akita Prefecture, Japan

More than 100 gigantic spherical carbonate concretions containing whale bone are exposed in the Unozaki coast of the Oga Peninsula in Akita Prefecture, Japan. This study examined the formation mechanism of these concretions, which has not been elucidated before, using field surveys and geochemical analyses. The results suggested that these gigantic whale bone concretions were formed by the rapid burial of the whale bone on the seafloor during wave storms and the decomposition of its carbon component by benthic organisms. The observation of the primary dolomite suggests that gigantic concretions may have formed in a reducing environment. Subsequently, gigantic spherical carbonate concretions were formed by growth of calcite. (This research



Giant whalebone concretion in the Oga Peninsula, Akita Prefecture, Japan.

was supported by the 2021 Akita Geopark Research Grant and was conducted with the cooperation of the Culture and Sports Division of Oga City and the Oga Peninsula–Oogata Geopark Association.)

# Relationship between Antarctic primary productivity and sea ice changes with sub-decadalscale climate variability during the last 11,400 years

The marine margin of Antarctica plays an important role in the global carbon cycle through the formation of Antarctic bottom water and primary productivity. An international joint research team that included researchers from the University of Tokyo, Nagoya University, Victoria University of Wellington, and Stanford University investigated a 170-m-long sediment core recovered from the Adélie Basin along the Wilkes Land margin of East Antarctica. The

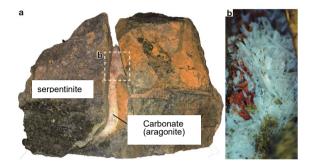


Off the coast of Wilkes Land, East Antarctica.

study revealed that sea ice variations off the coast of East Antarctica have been closely linked to sub-decadal-scale climate modes (for example, the El Niño–Southern Oscillation, Southern Annular Mode, and Indian Ocean Dipole) during the past ~11,400 years (Johnson *et al.*, 2021). The study findings can be used to improve models for predicting the global carbon and marine biogeochemical cycles.

#### Carbon cycle processes in shallow subduction zones

Carbon in the Earth's surface layer is brought to the Earth's depths as plates are subducted and then returned to the surface in a cyclic process. The importance of the shallow carbon cycle process, in which subducted carbon returns to the Earth's surface from relatively shallow areas, has long been highlighted; however, the time scale of this cycle is not well understood. In this study, carbonate (aragonite) mineral veins in forearc mantle rocks collected from Kaigamekaiyama (6,400-m depth) in the Izu–Ogasawara Trench were analyzed and found to have stable carbon isotope ratios similar to those of deep-sea water and low <sup>14</sup>C concentrations. This indicates that carbonates that originated from carbon in old seawater and were transported into the Earth's



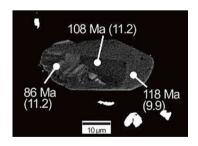
(a) Carbonate (aragonite) produced by precipitation in a fracture of altered mantle rock (serpentinite). (b) Enlarged view of the area enclosed by the white dotted line in (a). Aragonite is needle-shaped.

interior remained in the fore-arc mantle of the upper plate for tens of thousands of years or more. This finding is important and advances our understanding of the global carbon cycle by adding a new indicator, <sup>14</sup>C, to rocks that have been studied geologically and mineralogically. This research was conducted in collaboration with the Japan Agency for Marine-Earth Science and Technology, Tohoku University, Niigata University, and the National Institute of Advanced Industrial Science and Technology and was supported by the ISEE Joint Research Program (Accelerator Mass Spectrometry Analysis) (Oyanagi *et al.*, 2021, *Communications Earth & Environment*).

## Geochronological study of the Pingze–Dongshan metamorphic belt, Fujian, China

Geochronological studies of the Pingze-Dongshan metamorphic belt in Fujian Province, China are important for understanding the late Mesozoic tectonic development history of the continental margin in South China. Therefore, we

measured the ages of monazite and zircon in mica schist and intrusive granite in this region. U-Pb isotopic ages were determined by LA-ICP-MS at National Taiwan University and NanoSIMS at the University of Tokyo and U-Th-Pb CHIME ages were determined by EPMA at ISEE. The results indicated that the age of peak metamorphism of the mica schist reached its metamorphic peak at approximately 100 Ma. One group of zircons had a simple age cluster (145–30 Ma) while the other showed a wide age spectrum of two populations (c. 1.8 Ga and c. 190 Ma) that are uncommon in this region (Lin *et al.*, 2021, *Journal of Asian Earth Sciences*).



Backscattered electron image and CHIME ages of monazite (from Lin *et al.*, 2021)

# Structural developmental history of Hida and Yoengnam belts based on zircon ages and Hf isotopic ratios

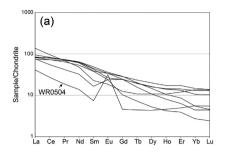
Integrated zircon U-Pb chronology and Hf isotope geochemistry of igneous and metamorphic rocks from the Hida Belt, southwest Japan, have led to a new understanding of the evolution of the arc system. The igneous parts of zircon in basic-medium orthogneisses from the Tateyama and Tsunokawa areas yielded Permian <sup>206</sup>Pb/<sup>238</sup>U ages. The

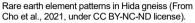
overgrown parts are 250–240 Ma in age and have a relatively high Th/U ratio (> 0.2). These parts may have been formed by a thermal event caused by the intrusion of the Hida granitoids. Based on the whole rock chemistry, Sr-Nd isotopic ratios, Hf isotopic ratios, and zircon ages, the Hida Belt was an island–arc system that formed along the North China Craton with the Yeongnam Belt in the Paleozoic–Mesozoic (Cho *et al.*, 2021, *Geoscience Frontiers*).

#### Isotope Ratio Meeting 2021

The Isotope Ratio Meeting of the Mass Spectrometry Society of Japan is held annually to allow researchers and students in Japan to use isotope analysis as their main research tool to exchange information on current problems in both hardware and software and to develop isotope ratio measurements and applied research fields. This is the only meeting in Japan that provides a forum for the exchange of isotope research using mass spectrometry as a research method.

This year, the ISEE took the lead in holding the online meeting on November 10–12, 2021, in collaboration with the Graduate School of Environmental Studies of Nagoya University. The meeting was attended by 94 participants, including 29 students. Two special scientific lectures, three invited lectures (one of which was given by Associate Professor Fusa Miyake of ISEE on "Investigation of past extreme solar events using cosmic ray-produced nuclides"), and seven live video introductions from the laboratories were given. In the laboratory introductions, the analytical equipment was demonstrated and the audience was able to catch a glimpse of the laboratories, making it a meaningful opportunity for exchange among isotope researchers.







Poster for Isotope Ratio Meeting 2021.

# Regional Contribution Program 2021 of Nagoya University "Let's Learn about Radiation from the Natural World"

The Regional Contribution Program 2021 of Nagoya University, "Let's Learn about Radiation from the Natural World," was conducted as a hands-on learning experience for upper elementary school students. The purpose of this program was to understand the nature of radiation and how humans are constantly exposed to radiation from the natural world. We have been considering ways to implement the "hands-on" learning program for COVID-19. We decided to send students educational materials for simple experiments that could be done safely at home, and to conduct experiments and discussions that would be difficult to perform at home in an online format. We distributed simple radiation measurement instruments and radiation sources to the students and asked them to measure the radiation levels in the samples and their surroundings. The results were discussed in an online format. Additionally, a fog-box experiment was conducted online. This allowed the participants to observe radiation emitted from minerals and the atmosphere, as well as radiation from outer space. Although the online format limited the number of possible experiments, it had the advantage of allowing participation from outside the Nagoya area. Participants from the Kansai area were able to examine differences in natural radiation doses that were thought to be due to geological differences.



Poster for 2021 Regional Contribution Program of Nagoya University.