

Review of development of AMS in the past 30 years and future perspective

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One of the main aims of Quaternary research is to understand global environmental changes of the past and to predict the expected changes in the near future. To pursue this aim, high-resolution age estimation is particularly important. Dating methods so far used for Quaternary research can be classified into five categories: (1) age estimation based on the decay procedure of cosmogenic radioisotopes such as radiocarbon (^{14}C) and ^{10}Be , U-series nuclides and a K-Ar pair; (2) age estimation by cumulative dose from natural nuclear radiation and cosmic rays such as TL, OSL, ESR, FT dating methods; (3) age estimation with geological or geochemical evidences of prehistoric events such as paleomagnetic reversals or excursions, oxygen stable-isotope-ratio variations, tephra stratigraphy; (4) age estimation with paleontological records of prehistoric events such as diatom, pollen, nanno-plankton and shellfish assemblages, as well as semi-global fluctuations of tree ring width; (5) age estimation based on archeological evidences. These dating methods are selectively applied dependent on the characters of geological and archeological events to be analyzed. Among the radio-isotopic dating methods, ^{14}C dating is most frequently used because of its applicability to many different types of Quaternary samples, as well as age range covered by this method (a few hundred to 50,000 yr BP).

Developments of accelerator mass spectrometry (AMS) have triggered a wide area of application in radiocarbon (^{14}C) dating. The AMS system requires only 1mg of carbon in precise determination of $^{14}\text{C}/^{12}\text{C}$ and $^{13}\text{C}/^{12}\text{C}$ isotope ratios, and this advantage has broadened the applicability of ^{14}C measurements. Nowadays, AMS contributes to almost all kinds of research that utilize ^{14}C dating in archeology, cultural property science, geology, and those that employ ^{14}C tracer in environmental sciences, medical sciences and even forensic studies.

For example, a Tandem AMS system dedicated to ^{14}C measurement was installed at Nagoya University, and its routine operation for ^{14}C measurement was started in 1983 for the first time in Japan. In 1996, another AMS system (HVE-Model-4130-AMS) was purchased and has been used for high precision ^{14}C measurements. By 30 minutes measurement of carbon isotopes repeated for consecutive three days for a sample, one-sigma uncertainty of ± 17 to ± 30 years is achieved. A reproducibility test for 2000-year-old archeological samples yielded a fluctuation error as small as ± 11 years. We also have evaluated accuracy in our ^{14}C measurements by participating in international ^{14}C inter-comparison tests, and confirmed that our ^{14}C results were quite consistent with the consensus values by all the participants. After the critical tests, we are sure that our AMS system can be applicable to historical samples that require high precision as well as high accuracy ^{14}C measurements.

Quite frequent applications of ^{14}C dating with AMS to the Quaternary samples in the last decade are promoted by the following reasons: (1) a very small amount of carbon (about 1mg of carbon for the final target preparation) is required; (2) uncertainties of ^{14}C ages are from ± 17 to ± 30 yr, mainly owing to the ^{14}C counting statistics; (3) calibration of ^{14}C age to the calendar age scale become quite popular, for ^{14}C ages up to 50,000 cal BP; (4) marine reservoir effect on ^{14}C age has been recognized and investigated recently, and a realistic correction for the effect is becoming possible partly.

Along with ^{14}C , other cosmogenic radioisotopes such as ^{10}Be , ^{26}Al , ^{36}Cl , ^{129}I are also measured with AMS systems. We briefly describe history of development of domestic AMS groups as well as worldwide AMS groups, along with the research fields of AMS applications and future perspective.

Keywords: accelerator mass spectrometry, cosmogenic nuclides, radionuclide, age measurement, ion nuclide separation, ion particle counting

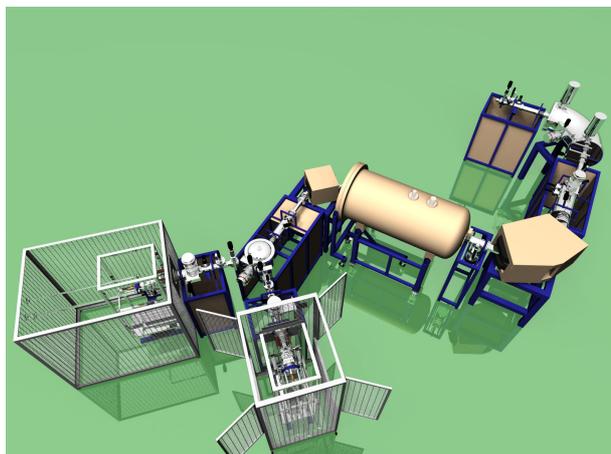
Present status of YU-AMS

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Yamagata University (YU) installed an AMS (YU-AMS) system in the Kaminoyama Research Institute to meet the requirement of ¹⁴C AMS for microdosing and medical studies as well as that of radiocarbon dating in the same facility. An automated graphitization line was also installed in the same research institute for sample preparation. This AMS system is the first AMS system installed in a university in north Japan (Tohoku-Hokkaido region). The facility also provides radiocarbon dating for samples from other universities, institutes and public organizations. Currently, we are planning to install a second new ion source and an automated graphitization line until March in 2014. In this paper, we describe the status of the YU-AMS system.

Keywords: AMS, microdose



Development of isobar suppression system using Laser in Accelerator mass spectrometry

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For Accelerator mass spectrometry (AMS), isobar separation is quite important to improve measurement accuracy and the background. In order to suppress isobar interference, gas-filled magnet or gas counter have been conventionally used [1,2]. Nuclides of interest are separated from isobars by interaction between ions and materials in these devices.

In the 1980s, Berkovits et al. tried to remove stable isobars before acceleration with laser light[3]. In this method, the difference of the electron affinity (EA) was utilized for isobar suppression. If the EA of the nuclide of interest is higher than the EA of the isobar nuclide, only negative isobar can be selectively neutralized by photodetachment with photons of energy, which is higher than the EA of isobar nuclide but lower than the EA of the nuclide of interest. Consequently, only nuclide of interest can be injected into the accelerator and isobar suppression can be effectively achieved. However, due to the limited laser performance, the laser-ion interaction time was too short to suppress isobar sufficiently at that time. Therefore, this technique has not been in practical use yet.

Recently, as laser improved in quality and the way to increase the laser-ion interaction time effectively was proposed, development of isobar suppression system is going on. For example, Liu et al. developed the RFQ ion cooler to slow ions [4]. This apparatus is filled with a buff gas and ions collide with gas molecules, which results in the deceleration of ions and the long interaction time. This photodetachment system can remove isobar interference in AMS measurements for nuclides, such as Cl-36 (EA=3.62eV) with S-36 (EA=2.08eV), Ni-59 (EA=1.156eV) with Co-59 (EA=0.661eV) [4]. Furthermore, even if the EA of the nuclide of interest is lower than the EA of the isobar nuclide, photodetachment could be useful by converting the nuclides into the molecular ions and reversing these electron affinities.

In order to make laser interact with ion beams effectively, the ion beam optics including the devices like the ion cooler should be optimized. This device will be installed after the electrostatic deflector or after the injection magnet in the beam line. In this study, as a preliminary step, optimization of the ion beam optics including some components to decelerate ion will be discussed.

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Keywords: AMS, isobar, laser, photodetachment

Development of C14-free laboratory animals.

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In the early stage of pharmaceutical researches, ¹⁴C labeled chemicals with very high radioactivity are administrated to laboratory animals to study pharmacokinetics. However, when ¹⁴C/¹²C of the laboratory animals could be reduced, the radioactivity level of the chemicals is expected to be reduced lower. Also when ¹⁴C-free (DC ; denoted as dead carbon) laboratory animals were developed, the radioactivity level for the study is expected to be reduced down by five orders of magnitude or lower by using AMS technique. That means that we need not the ¹⁴C labeled chemicals but natural level chemicals for the study of pharmacokinetics. In this case, it is also expected that we can be free from the hazardous high radioactivity and from the strictly regulated troublesome laboratory to handle the high level radioactive materials.

In Dec. 2010, we started the project to produce ¹⁴C-free lives as feed for the ¹⁴C-free laboratory animals. This is a preliminary report on the project, and parts of the investigation were already reported at 13th Japanese Symposium on AMS (Kobayashi et al., 2011). Here will be also introduced some recent studies.

When lives were cultivated in DC surroundings they grow up inevitably to be DC lives. For example, photosynthetic lives like vegetables or some kinds of microbes with chloroplast like euglena (midorimushi ; in Japanese) can easily be DC lives when they were cultivated in DC surroundings.

For the first step, we cultivated some plants and euglena using ¹⁴C free water set in a glove box that was filled with artificial air (N₂, O₂ and dead carbon CO₂ with proper concentration) irradiating with some artificial lights. ¹⁴C concentrations in the plants and the microbes were measured by AMS. Since the modern carbon (natural carbon) CO₂ could not be perfectly removed from the air and the water and air tightness of the glove box system was also not perfect, the percentages of dead carbon to modern carbon (DC ratio) in the samples could not be higher than about 80 %.(Kobayashi et al., 2011a ; Kobayashi et al., 2011b)

While investigating the cause of the imperfect DC ratio, we got 96% DC Euglena by using a little different way. That is to use a small glass bowl with an airtight lid, in which were set CO₂ tablets, a small amount of chemical fertilizer and Euglena. From outside of the bowl, fluorescent lamps irradiated adequate amount of light to them. One or two months later, the euglena was carefully collected on glass filters in a glove box filled with DC air. ¹⁴C/¹²C of the euglena was measured by AMS and the ¹⁴C concentration was 3.71±0.02 pMC (percent Modern Carbon) which leads to DC ratio as 96% (Kobayashi et.al., 2012).

In order to make sure that mice really eat the euglena tablets, we fed the mice with euglena tablets which were mixed with chlorella powder by 20%. The tablet is on the market for people as a health food. After three months feeding on three mice, there were no large differences compared to the other three mice which were fed by ordinary food for comparison. The experiment concluded that mice will grow up healthy by eating the euglena tablet.

Those two above data show the possibility to realize the DC animals.

Recently, we have started to extend the project to realize for business, supported by a grant.

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Kobayashi, K. et al. (2011a), Production of ¹⁴C-free plants and animals (in Japanese) ; Kobayashi, K. et al. (2011b), Production of ¹⁴C-free plants and animals ; Kobayashi, K. et al. (2012), Production of dead carbon lives (in Japanese)

Keywords: Pharmacokinetics by AMS, C14-free lives, Euglena

¹⁴C measurement of the Southern Japanese tree by the AMS method for high-precision radiocarbon calibration

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Radiocarbon (¹⁴C) dating method has contributed to the age determination of samples of the past 50,000 years in geology and archaeology. However, since ¹⁴C date is not in agreement with the calendar year, the calibration using a dataset which consists of ¹⁴C data of calendar year known samples is required for it. Moreover, since there is regional difference of ¹⁴C concentration in the atmosphere (regional ¹⁴C offset; Hogg et al. 2002), in order to be high-precision calibration, the dataset for each area needs to be established.

The Center for Chronological Research (CCR), Nagoya University has measured ¹⁴C data of tree-rings of a Yaku cedar from the Southern Japan, in order to establish a calibration dataset for Japan. Previous measurement has shown that the cedar shows ¹⁴C date older than global standard calibration dataset IntCal13 (Reimer et al. 2013) in various times for the past 2000 years (Nakamura et al. 2013). This report shows the measurement result of the tree-rings formed in the 5th century.

Sample is a Japanese cedar from Yaku Island, Kagoshima prefecture (sample code: Yaku_A). Dendrochronological analysis with a master-chronology (Kimura unpublished) has carried out, and the calendar year of this sample is known. After exfoliating the annual rings of AD434-502 of a sample in one year respectively, only even-numbered years was measured (n = 35). The tandemron AMS II of CCR was used for this measurement. Measuring only even-numbered years in order to understand a whole tendency previously, it also measures the samples of odd-numbered years in the future.

Thirty five ¹⁴C dates of tree-ring samples showed that it will be older than IntCal13 for an average of 28 ± 22 years, a maximum of 76 ± 21 years (in AD488). These ¹⁴C dates were mostly located in the middle of IntCal13 and SHCal13 (the calibration dataset for the Southern Hemisphere; Hogg et al. 2013).

Since Yaku Island touch the northernmost end of the Intertropical Convergence Zone in a summer, it is thought that the Southern Hemisphere atmosphere with regularly low ¹⁴C concentration is easy to be supplied (Nakamura et al. 2012). This measurement result might suggest that ¹⁴C concentration in the atmosphere of the Japanese neighborhood fell in the 5th century, and the atmospheric supply from the Southern Hemisphere may have become strong. Sakamoto et al. (2013) measured the tree-ring samples in the 5th to 6th century of the Japanese cedar from Nagano Prefecture Central Japan, and they has reported that the data is older than a IntCal13. Our result harmonizes with the measurement result of the trees from Nagano, and this time can consider a possibility that the influence of the Southern Hemisphere atmosphere had reached to central Japan. From now on, the tree-rings of Yaku cedar in formed the 6th century will measure, and it will compare with the result of Sakamoto et al. (2013).

Keywords: radiocarbon calibration, regional ¹⁴C offset, Southern Japan, tree-ring, Yaku cedar

C-14 dating and geochemical analyses of the tsunami sediments in continuous soil deposits from Tohoku area, Japan

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Past tsunami sediments preserved in continuous soil and lake sediments are crucial and unique clues to reconstruct the past tsunami invasion area. Generally, the tsunami sediments originated from sea floor, sandy beach and/or coastal land soils containing gravels, sands, muds, shells and microfossils. In particular, muddy tsunami sediments should be found to detect the limit of tsunami invasion areas, because relative small particles move to more inland area with tsunami in comparison with sand deposits. Additionally, dating of tsunami sediments is indispensable to refer for historical disaster events. Therefore, we have to make age models of continuous soil deposits with tsunami sediments and new techniques for detection of invisible muddy tsunami sediment in strata. In this study, we performed the nine radiocarbon measurements of plant residues in continuous soil deposits as well as geochemical characteristics of tsunami sediments. 2m-continuous soil deposits were taken by the handy geoslicer (Fukkenn co. ltd.) from the Pacific coast of Tohoku area in northeast Japan. The samples were composed of cultivated surface soils, peaty clay, silt and sub-rounded medium sands. The sandy deposits were found between the peaty clay layers. To show the sedimentary ages of sandy deposits, plant residues were taken from the sandy and peaty clay layers in the continuous soil sediments. The plant residues were washed with ultra pure water using ultra sonic cleaner to remove soil particles containing relative old carbon. Then, the samples were treated sequentially with 1.2M-HCl, 1.2M-NaOH and 1.2M-HCl at 60 degrees of Celsius for 3 hours. After neutralization and freeze-drying, the samples were combusted in evacuated quartz tubes. Then, the purified carbon dioxide was reduced to graphite using Hydrogen gas with iron catalysts. Radiocarbon measurements were performed by the Tandem AMS system (Model-4130, HVEE) in Center for Chronological Research, Nagoya University. Total organic carbon contents of the plant residues were from 45.9 to 54.5 wt.% (50.4 wt.% in average) and stable carbon isotope ratios of the plant residues ranged between -26.7 and -30.1 permil (vs. PDB), which consist with those of modern terrestrial C3 plants. As a result of this study, part of the calibrated ages of plant residues taken from just above the sandy tsunami sediments was about 1000-1300 cal BP, and these ages were agreed well with those of the Jogan earthquake and tsunami in the Sendai plain.

Keywords: Radiocarbon dating, Tsunami deposits, Jogan tsunami, Geochemistry, EDXRF

AMS radiocarbon dating of tephra layers on Adak Island, central Aleutian

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The Holocene tephra layers distribute on Adak Island located in the central part of the Aleutian Islands. Radiocarbon (¹⁴C) age determination of charcoal fragments in the soil layer has been performed. Moreover, tephra is intercalated with sand layer of dune. Since sand dune has high depositional rate and low contamination of organic matter, a possibility of polluted charcoal sample from below and above tephra is low. Thus reliable age was able to be obtained by the AMS method to these small samples. We report those results.

Keywords: Adak Island, Holocene, tephra, radiocarbon date

Radiocarbon dating of stalagmites from the Ryugashi Cave, Shizuoka

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Stalagmites are cave deposits precipitated from drip water. Drip water consists of carbon derived from soil CO₂, which has atmospheric ¹⁴C values in isotopic equilibrium with atmosphere, and carbonate-dissolved CO₂, which has ¹⁴C-free (dead) carbon through interaction with cave host bedrock. As a result, drip water contains a percentage of dead carbon, which will make the ¹⁴C ages of the stalagmite older. Therefore, a correction of the dead carbon fraction is needed for ¹⁴C dating of stalagmites. In recent years, young stalagmites of 10-20 ka have been ¹⁴C dated by comparing the ¹⁴C on samples of known calendar age with the tree ring record of atmospheric ¹⁴C during a period of overlap (Hoffmann *et al.*, 2010; Southon *et al.*, 2012). This procedure involves the implicit assumption that dead carbon fraction in stalagmite remained constant through its growth time. In this study, therefore, we examined dead carbon fraction in two stalagmites from the Ryugashi Cave in Hamamatsu, Shizuoka by investigating seasonal variation in ¹⁴C concentrations of drip water coupled with soil CO₂, atmospheric CO₂, and host limestone, in order to reveal possibility of accurate and precise ¹⁴C dating on stalagmite in Japan.

The drip water samples showed ¹⁴C of 1130 BP to 980 BP and $\delta^{13}\text{C}$ of -10.1 ‰ to -9.1 ‰, which are lower in fall and winter, and higher in spring and summer, and have the annual means of ¹⁴C of 1025±140 BP and $\delta^{13}\text{C}$ of -9.4±0.4 ‰. The RYGS12 stalagmite of 7 cm in length showed 945±30 BP at its top and 2150±40 BP at its bottom, and had a growth rate of about 60 $\mu\text{m}/\text{yr}$. The calibrated age of RYGS12 was estimated by comparing the ¹⁴C with the IntCal13 calibration curve, resulting that the stalagmite had a constant dead carbon fraction through its growth time and gives ¹⁴C ages of 1050 years older than the true age. The carbon isotopic fractionation between drip water and stalagmite was negligible. The results indicate that high-resolution ¹⁴C measurement can be performed on stalagmites in the Ryugashi Cave.

The RYGS12 sample showed rapid decrease of $\delta^{13}\text{C}$ from -8.3 ‰ to -11.8 ‰ at around AD1450. The decrease suggests an increase of soil input to the stalagmite, since soil CO₂ has low $\delta^{13}\text{C}$ of -22.0 ‰. It is reported that there was a great earthquake of magnitude 8.6 (Meio earthquake) accompanied by a catastrophic tsunami in this study area in AD1498. Therefore, the $\delta^{13}\text{C}$ decrease might be caused by the Meio earthquake. In the presentation, we will present ¹⁴C result on another stalagmite sample RYG08 of 30 cm in length.

Keywords: stalagmite, radiocarbon age, carbon isotope ratio, oxygen isotope ratio

14C-based source apportionment of carbonaceous component in PM2.5 in Nagoya city

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The measurement of radiocarbon (¹⁴C) has been used to estimate the relative contributions of fossil and contemporary carbon sources in particulate matter throughout the world. In this study, we use ¹⁴C to provide quantitative estimates of carbon origin sources in Nagoya in April 2003 to March 2004. pMC and OC/EC showed similar seasonal variations and high values (range 39.8 to 68.4, 1.0 to 2.0; average 53.4, 1.5, respectively) in May and early June, whereas the values of pMC stayed relatively low values (range 28.3 to 41.9, 0.7 to 1.2; average 34.2, 1.0, respectively) after middle June. To estimate the source region of high pMC values, backward air mass trajectories were calculated during the sampling period in April to June. The air mass appeared to have passed through eastern Siberia when the pMC values showed high values in May and early June which also showed high values of OC/EC ratio and OC concentrations. In 2003, many researchers reported the influence of large forest burning in Siberia. This large forest burning occurred in spring to summer. The smoke from this forest burning had reached to Korea, Japan, and North America. The influences from the Siberian forest fires had an important implication for air quality over East Asian region. We conclude that high pMC values measured in spring are originated from large forest fires in Siberia and transported long distance.

Keywords: atmospheric aerosol, PM2.5, radiocarbon

Decadal change in bomb-produced radiocarbon in the Pacific Ocean revealed by WHP repeat hydrography

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Radiocarbon produced by nuclear weapon tests is one of ideal tracers for the air-sea gas exchange and ocean circulation. In the 2000s, radiocarbon in dissolved inorganic carbon was measured during revisit cruises along the WOCE (World Ocean Circulation Experiment) lines of P01 (47N approx., 2007), P03 (24N approx. 2005), P06 (32S approx., 2003), P10 (149E approx., 2005), P13N (165E approx., 2011), P14N/C (179E approx., 2007), P17N (135W approx., 2001), and P21 (17S approx., 2009) conducted in the 1990s in the Pacific Ocean. Comparison of radiocarbon data from the 1990s and 2000s revealed decadal changes of radiocarbon concentration in the thermocline, most of which were due to temporal changes in the bomb-produced radiocarbon. Vertical profiles and vertical-integrated inventories of the bomb radiocarbon in the subarctic and equatorial regions have not changed significantly. In the subtropical regions, radiocarbon decreased in upper thermocline from surface to about 500-m depth. In contrast, radiocarbon increased in lower thermocline from about 500-m to 1500-m depths. In the southern and northeastern subtropical regions, the two opposing directions in radiocarbon change resulted in small temporal changes of the total inventory of the bomb radiocarbon. On the other hand, the water column inventory significantly decreased in the northwestern subtropical region because the radiocarbon decrease in the upper thermocline was larger than the radiocarbon increase in the lower one. These decadal changes are primarily due to the meridional transport of the bomb radiocarbon from high latitude into temperate zone. The decrease in the vertical-integrated radiocarbon in the northwestern subtropical region implies that the turnover time of the thermocline circulation in the region is faster than those in the other subtropical regions in the Pacific Ocean. In addition the loss of the bomb-radiocarbon in the North Pacific Ocean could be explained by its transformation to the Indian Ocean via Indonesian Through Flow. This work was partially supported by Japan Society for the Promotion of Science (JSPS) KAKENHI Grant Number 18310017 and the Common-Use Facility Program of JAEA (2007A-F03, 2007B-F05, 2008A-F02, 2009A-F05, 2010A-F06, and 2011A-F04).

Keywords: bomb-produced radiocarbon, Pacific Ocean, ocean circulation

Study on property of soil organic matter decomposition by global warming using radiocarbon

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Current research indicates that future atmospheric CO₂ concentration may be increased more than predicted value by furthering of soil organic matter decomposition due to global warming. The information on soil organic matter decomposition property in long-term warmer environment has not yet been obtained. We have carried out artificial soil warming experiment in six forest sites having different vegetation in Japan for long time. We planned vertical ¹⁴C measurement of soil core at an even-green Japanese oak forest in Setonaikai region (Higashi-Hiroshima). A soil core was collected from both the soil warming and the control plot in December 2011, and was cut into 1cm layers in laboratory. Each sample was hydrolyzed with 1N HCl overnight to remove inorganic carbon in the sample, and then was analyzed particulate organic carbon (POC) and organic nitrogen (PON) by an elemental analyzer. For ¹⁴C analysis by an accelerator mass spectrometer (AMS), soil samples adjusted to a weight of approximately 3mg-C were first converted to CO₂ gases by combustion with CuO and Ag foil at 900 °C, and then purified cryogenically in a vacuum line. The CO₂ gas samples were reduced to graphite with H₂ gas over Fe powder. The ¹⁴C/¹²C ratios of the sample graphite were measured at the Tandem AMS Facility in the Mutsu Office of the Japan Atomic Energy Agency. The ¹⁴C results are expressed as Δ¹⁴C. The typical analytical error of the Δ¹⁴C values was about ± 4 ‰ based on the 1σ value of the counting statistics.

Both of POC and PON weight percent in the soil were high above 3cm depth and decreased sharply with depth from 5cm to 15cm. Both of POC and PON of the soil warming plot were 20-30% lower than those of the control plot irrespective of depth above 15cm depth. The result indicates that the soil warming experiment was encouraged the microbial decomposition of soil organic matter up to comparatively deep layer. The Δ¹⁴C profile of the warming plot was unique with a maximum (220 ‰) at 5cm depth, although the Δ¹⁴C of the control plot was approximately constant from surface to 10cm depth. In terms of Δ¹⁴C vertical profile above 10cm depth, although the Δ¹⁴C of the warming plot above the 3cm depth having POC >15 wt% were lower than those of the control plot, the Δ¹⁴C of the warming plot below the 3cm depth were obviously higher than those of the control plot. The results indicate that microbes selectively decomposed young POC at surface layer and old POC at intermediate layer by the soil warming experiment.

Keywords: soil carbon, radiocarbon, global warming, organic matter decomposition, forest soil

Cosmogenic $^{36}\text{Cl}/^{10}\text{Be}$ ratio in the Antarctic ice core during the last deglaciation and early Holocene

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^{36}Cl is cosmogenic nuclide (half-life:301 kyr) produced mainly by a reaction of $^{40}\text{Ar}(p, n\alpha)^{36}\text{Cl}$ in the upper atmosphere. Cosmogenic nuclides fall on the Earth's surface at a rate depending on the nuclide production rates and hence reflecting the cosmic ray intensity. Therefore we can reconstruct fluctuations of cosmic ray intensity, by determining the past ^{36}Cl depositional flux. Such fluctuations of cosmic ray intensity may indicate paleo solar activity and/or variations in the Earth's geomagnetic field.

In this presentation, we report the results of cosmogenic ^{36}Cl measurements during 10.55 - 18.42 kyr b2k in the ice core drilled at the Dome Fuji station, Antarctica (Motoyama et al., 2007). ^{36}Cl in the ice was measured with the accelerator mass spectrometry (AMS) system at the University of Tsukuba (Sasa et al., 2010). The results show that ^{36}Cl conc. is $0.21 - 1.80 \times 10^4$ atoms g^{-1} and ^{36}Cl flux is $0.54 - 3.25 \times 10^4$ atoms $\text{cm}^{-2} \text{yr}^{-1}$. The variation of ^{36}Cl flux in early Holocene shows similar fluctuations of ^{10}Be flux in the same ice core. $^{36}\text{Cl}/^{10}\text{Be}$ is constant at 0.10 ± 0.01 in early Holocene. This means that this value can be used for radioactive age dating of the old ice core. $^{36}\text{Cl}/^{10}\text{Be}$ varies in the last deglaciation. It suggests that the decrease in $^{36}\text{Cl}/^{10}\text{Be}$ ratio is linked to climate change.

Keywords: $^{36}\text{Cl}/^{10}\text{Be}$, Cosmogenic nuclide, Accelerator Mass Spectrometry, Radiometric age determination, Ice core

Correlation between the concentrations of cosmogenic Be-7, Be-10 in atmosphere and solar activities.

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The concentrations of ⁷Be and ¹⁰Be were investigated at Dazaifu, Fukuoka (1998-2002), Hachijo-Island (2002-2005) and Tokyo (2002-2008) during 1998 to 2008. The seasonal variations were same each year; high concentrations and high isotopic ratios of ¹⁰Be/⁷Be that was caused by strong stratosphere-troposphere exchange (STE) were appeared in February to June, and various concentrations but constant ¹⁰Be/⁷Be by vertical convection in troposphere were appeared in July to December. The concentrations were reconstructed by the box model formula. The parameters of the mean residence time and STE intensities, and period, were constant. The amplitudes of production rate were higher than the amplitude of cosmic ray neutron flux observed at Moscow near earth's surface by a factor of 4. Since the neutron flux amplitude at polar region that was little influenced by geomagnetic field was only 10% higher than Moscow, the high amplitudes of production rate were assumed that caused by changing of energy spectrum of galactic cosmic ray.

Keywords: Accelerator mass spectrometry, Cosmogenic nuclide, atmosphere, aerosol

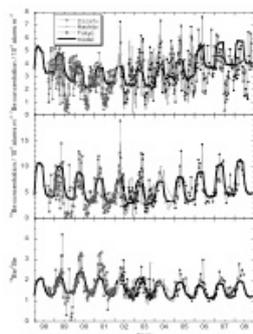


Fig. 1 The decadal variations of ⁷Be, ¹⁰Be concentration and ¹⁰Be/⁷Be in the atmosphere in Dazaifu, Hachijo-Island and Tokyo during 1998 to 2008.

Distributions of radionuclides Cl-36 and I-129 in surface soils before Fukushima accident

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The long-lived radionuclides ³⁶Cl and ¹²⁹I are generated by the nuclear tests or interaction with cosmic rays. They have descended to ground or sea level surface, and they have remained ground surface afterward. We have measured amount of ³⁶Cl and ¹²⁹I by accelerator mass spectrometry (AMS) before nuclear accident at the Fukushima No. 1 nuclear power plant.

We have collected surface soil samples from the Sea of Japan to the Pacific Ocean at the equal-latitude cross-sectional areas (37° 20' N - 37° 30' N) in the south Tohoku, Japan. Inorganic chlorine in soil developed an improved leaching process that uses diluted HNO₃ as an extractant, activated carbon to remove organic matters without decomposition, and H₂O₂ to remove residual organic matters. After leaching from soils, the AgCl samples for AMS-target made from the obtained solutions at ordinary treatment. Isotopic ratios of ³⁶Cl/Cl were determined by AMS at Tandem Accelerator Complex, University of Tsukuba. Preparation of Iodine-129 target was following ordinary method. Isotopic ratios of ¹²⁹I/I were determined by accelerator mass spectrometry (AMS) at MALT, the University of Tokyo. Moreover, we determined ¹³⁷Cs concentrations by gamma spectroscopy and LOI (loss on ignition used by an electric furnace) which related to the amount of the organic matter in soil.

We obtained the distributions of radionuclides ³⁶Cl and ¹²⁹I in surface soils. The measured ³⁶Cl/Cl ratios of 34 surface soil samples which were about 0-10 cm in depth from 6 sites at the equal-latitude cross-sectional areas were between 0.1×10^{-13} and 4.1×10^{-13} . It was shown that the ³⁶Cl/Cl ratios are lower at both sea sides. The concentrations of ¹²⁹I and ¹²⁹I/I ratios in surface soil (0-10 cm) at 28 points were determined to be 0.18 - 1.13 mBq/kg and 4.3×10^{-9} - 11.7×10^{-9} , respectively.

The depth profiles of ³⁶Cl/Cl, ¹²⁹I and ¹³⁷Cs were examined that the difference of distribution. The concentrations are higher at close surface in each nuclide. The results of ³⁶Cl/Cl profiles in soil cores up to 1 m long suggested that bomb-produced ³⁶Cl remains in uppermost sections, typically for ~20 cm deep, in undisturbed soil layers. The observed close correlation between organic matter content and ³⁶Cl/Cl ratio implies that presence of biological activity contributes the retention of fallout ³⁶Cl in the surface zone. The concentration of ¹²⁹I is shown the highest in uppermost surface. It is thought that the influence of global fallout has been received until now. In both nuclides, a constant amount exists in deeper than 30 cm. The concentrations of ¹³⁷Cs are not detection in deeper than 40 cm. It is showed that ¹³⁷Cs was lower mobility in soils. Therefore, the sampling soils were not a disturbance.

Keywords: AMS, Cl-36, I-129, soil