Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-01 Room:311 Time:May 1 09:00-09:15

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 (14C) and 10Be, 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, 14C 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 (14C) dating. The AMS system requires only 1mg of carbon in precise determination of 14C/12C and 13C/12C isotope ratios, and this advantage has broadened the applicability of 14C measurements. Nowadays, AMS contributes to almost all kinds of research that utilize 14C dating in archeology, cultural property science, geology, and those that employ 14C tracer in environmental sciences, medical sciences and even forensic studies.

For example, a Tandetron AMS system dedicated to 14C measurement was installed at Nagoya University, and its routine operation for 14C 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 14C 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 14C measurements by participating in international 14C inter-comparison tests, and confirmed that our14C 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 14C measurements.

Quite frequent applications of 14C dating with AMS to the Quaternary samples in the last bidecade 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 14C ages are from +-17 to +-30 yr, mainly owing to the 14C counting statistics; (3) calibration of 14C age to the calendar age scale become quite popular, for 14C ages up to 50,000 cal BP; (4) marine reservoir effect on 14C age has been recognized and investigated recently, and a realistic correction for the effect is becoming possible partly.

Along with 14C, other cosmogenic radioisotopes such as 10Be, 26Al, 36Cl, 129I 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

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(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-02 Room:311 Time:May 1 09:15-09:30

Present status of YU-AMS

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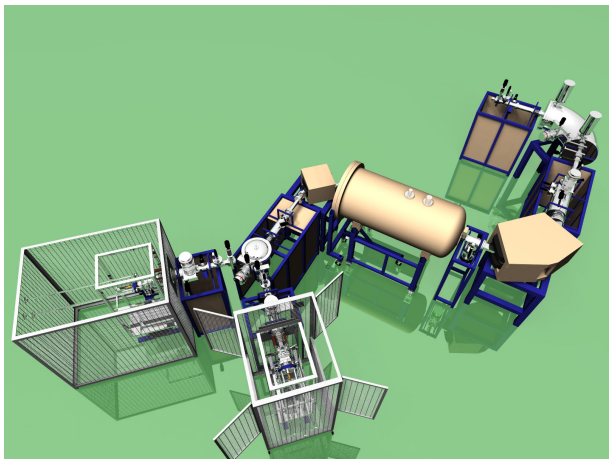
Yamagata University (YU) installed an AMS (YU-AMS) system in the Kaminoyama Research Institute to meet the requirement of ^{14}C AMS for microdosing and medical studies as well as that of radiocarbon dating in the same facility. An automated graphitization line was also installed in the same research 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



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HTT35-03 Room:311 Time:May 1 09:30-09:45

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 dicussed.

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Keywords: AMS, isobar, laser, photodetachment

Japan Geoscience Union Meeting 2014
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HTT35-04 Room:311 Time:May 1 09:45-10:00

Development of C14-free laboratory animals.

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In the early stage of pharmaceutical researches, ^{14}C labeled chemicals with very high radioactivity are administrated to laboratory animals to study pharmacokinetics. However, when $^{14}C/^{12}C$ of the laboratory animals could be reduced, the radioactivity level of the chemicals is expected to be reduced lower. Also when ^{14}C-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 ^{14}C 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 ^{14}C-free lives as feed for the ^{14}C-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 ^{14}C free water set in a glove box that was filled with artificial air (N_2, O_2 and dead carbon CO_2 with proper concentration) irradiating with some artificial lights. ^{14}C concentrations in the plants and the microbes were measured by AMS. Since the modern carbon (natural carbon) CO_2 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 CO_2 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. $^{14}C/^{12}C$ of the euglena was measured by AMS and the ^{14}C 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.

References:

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

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

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HTT35-05 Room:311 Time:May 1 10:00-10:15

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

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Radiocarbon (^{14}C) dating method has contributed to the age determination of samples of the past 50,000 years in geology and archaeology. However, since ^{14}C date is not in agreement with the calendar year, the calibration using a dataset which consists of ^{14}C data of calendar year known samples is required for it. Moreover, since there is regional difference of ^{14}C concentration in the atmosphere (regional ^{14}C 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 ^{14}C 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 tandetron 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 ^{14}C dates of tree-ring samples showed that it will be older than IntCal13 for an average of 28 $\pm$ 22 years, a maximum of 76 $\pm$ 21 years (in AD488). These ^{14}C 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 ^{14}C concentration is easy to be supplied (Nakamura et al. 2012). This measurement result might suggest that ^{14}C 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 ^{14}C offset, Southern Japan, tree-ring, Yaku cedar

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HTT35-06 Room:311 Time:May 1 10:15-10:30

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 Tandetron 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

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HTT35-07 Room:311 Time:May 1 10:30-10:45

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 (ˆ{14} C) 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

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HTT35-08 Room:311 Time:May 1 11:00-11:15

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 CO_2, which has atmospheric ^{14}C values in isotopic equilibrium with atmosphere, and carbonate-dissolved CO_2, which has ^{14}C-free (dead) carbon through interaction with cave host bedrock. As a result, drip water contains a percentage of dead carbon, which will make the ^{14}C ages of the stalagmite older. Therefore, a correction of the dead carbon fraction is needed for ^{14}C dating of stalagmites. In recent years, young stalagmites of 10-20 ka have been ^{14}C dated by comparing the ^{14}C on samples of known calendar age with the tree ring record of atmospheric ^{14}C 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 ^{14}C concentrations of drop water coupled with soil CO_2, atmospheric CO_2, and host limestone, in order to reveal possibility of accurate and precise ^{14}C dating on stalagmite in Japan.

The drip water samples showed ^{14}C of 1130 BP to 980 BP and $\delta^{13}C$ 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 ^{14}C of 1025±140 BP and $\delta^{13}C$ 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 μm/yr. The calibrated age of RYGS12 was estimated by comparing the ^{14}C with the IntCal13 calibration curve, resulting that the stalagmite had a constant dead carbon fraction through its growth time and gives ^{14}C 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 ^{14}C measurement can be performed on stalagmites in the Ryugashi Cave.

The RYGS12 sample showed rapid decrease of $\delta^{13}C$ from -8.3 ‰ to -11.8 ‰ at around AD1450 The decrease suggests an increase of soil input to the stalagmite, since soil CO_2 has low $\delta^{13}C$ 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 $\delta^{13}C$ decrease might be caused by the Meio earthquake. In the presentation, we will present ^{14}C result on another stalagmite sample RYG08 of 30 cm in length.

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

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HTT35-09 Room:311 Time:May 1 11:15-11:30

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

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HTT35-10 Room:311 Time:May 1 11:30-11:45

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

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HTT35-11 Room:311 Time:May 1 11:45-12:00

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

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Current research indicates that future atmospheric CO_2 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 planed vertical ^{14}C 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 ^{14}C analysis by an accelerator mass spectrometer (AMS), soil samples adjusted to a weight of approximately 3mg-C were first converted to CO_2 gases by combustion with CuO and Ag foil at 900 ℃, and then purified cryogenically in a vacuum line. The CO_2 gas samples were reduced to graphite with H_2 gas over Fe powder. The $^{14}C/^{12}C$ ratios of the sample graphite were measured at the Tandetron AMS Facility in the Mutsu Office of the Japan Atomic Energy Agency. The ^{14}C results are expressed as $\Delta^{14}C$. The typical analytical error of the $\Delta^{14}C$ values was about $\pm$ 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 $\Delta^{14}C$ profile of the warming plot was unique with a maximum (220 ‰) at 5cm depth, although the $\Delta^{14}C$ of the control plot was approximately constant from surface to 10cm depth. In terms of $\Delta^{14}C$ vertical profile above 10cm depth, although the $\Delta^{14}C$ of the warming plot above the 3cm depth having POC $>$15 wt% were lower than those of the control plot, the $\Delta^{14}C$ 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

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HTT35-12 Room:311 Time:May 1 12:00-12:15

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

SASA, Kimikazu[1*] ; KUROSUMI, Kazuna[1] ; SUEKI, Keisuke[1] ; TAKAHASHI, Tsutomu[1] ; MATSUSHI, Yuki[2] ; TOSAKI, Yuki[3] ; HORIUCHI, Kazuho[4] ; UCHIDA, Tomoko[5] ; MATSUZAKI, Hiroyuki[6] ; MOTOYAMA, Hideaki[7]

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^{36}Cl is cosmogenic nuclide (half-life:301 kyr) produced mainly by a reaction of $^{40}Ar(p, n\alpha)^{36}Cl$ 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 cm^{-2} yr^{-1}. The variation of ^{36}Cl flux in early Holocene shows similar fluctuations of ^{10}Be flux in the same ice core. $^{36}Cl/^{10}Be$ is constant at 0.10 $\pm$ 0.01 in early Holocene. This means that this value can be used for radioactive age dating of the old ice core. $^{36}Cl/^{10}Be$ varies in the last deglaciation. It suggests that the decrease in $^{36}Cl/^{10}Be$ ratio is linked to climate change.

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

Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-13 Room:311 Time:May 1 12:15-12:30

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

YAMAGATA, Takeyasu[1*] ; NARAZAKI, Yukinori[2] ; NAGAI, Hisao[1] ; MATSUZAKI, Hiroyuki[3]

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The concentrations of ^{7}Be and ^{10}Be 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 ^{10}Be/^{7}Be that was caused by strong stratosphere-troposphere exchange (STE) were appeared in February to June, and various concentrations but constant ^{10}Be/^{7}Be 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

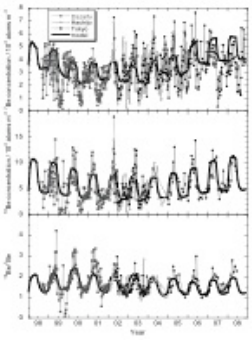

Fig. 1 The decadal variations of ^{7}Be, ^{10}Be concentration and ^{10}Be/^{7}Be in the atmosphere in Dazaifu, Hachijo-Island and Tokyo during 1998 to 2008.

Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-14 Room:311 Time:May 1 12:30-12:45

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

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[1]University of Tsukuba, [2]High energy acclerator research organization, [3]Shimizu corporation, [4]The National Institute of Advanced Industrial Science and Technology, [5]Kyoto University, [6]The University of Tokyo

The long-lived radionuclides ^{36}Cl and ^{129}I 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 ^{36}Cl and ^{129}I 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 ^{o} 20' N - 37 ^{o} 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 H_2O_2 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 ^{36}Cl/Cl were determined by AMS at Tandem Accelerator Complex, University of Tsukuba. Preparation of Iodine-129 target was following ordinary method. Isotopic ratios of ^{129}I/I were determined by accelerator mass spectrometry (AMS) at MALT, the University of Tokyo. Moreover, we determined ^{137}Cs 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 ^{36}Cl and ^{129}I in surface soils. The measured ^{36}Cl/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 x 10^{-13} and 4.1 x 10^{-13}. It was shown that the ^{36}Cl/Cl ratios are lower at both sea sides. The concentrations of ^{129}I and ^{129}I/I ratios in surface soil (0-10 cm) at 28 points were determined to be 0.18 - 1.13 mBq/kg and 4.3 x 10^{-9} - 11.7 x 10^{-9}, respectively.

The depth profiles of ^{36}Cl/Cl, ^{129}I and ^{137}Cs were examined that the difference of distribution. The concentrations are higher at close surface in each nuclide. The results of ^{36}Cl/Cl profiles in soil cores up to 1 m long suggested that bomb-produced ^{36}Cl remains in uppermost sections, typically for ˜20 cm deep, in undisturbed soil layers. The observed close correlation between organic matter content and ^{36}Cl/Cl ratio implies that presence of biological activity contributes the retention of fallout ^{36}Cl in the surface zone. The concentration of ^{129}I 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 ^{137}Cs are not detection in deeper than 40 cm. It is showed that ^{137}Cs was lower mobility in soils. Therefore, the sampling soils were not a disturbance.

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

Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-P01 Room:Poster Time:May 1 18:15-19:30

Geoscience studies using by AMS at JAEA-AMS-TONO in the Tono Geoscience Center of the Japan Atomic Energy Agency

KOKUBU, Yoko[1*] ; MATSUBARA, Akihiro[1] ; HANAKI, Tatsumi[1] ; YASUE, Ken-ichi[2] ; UMEDA, Koji[2]

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The JAEA-AMS-TONO facility was established in 1997 at the Tono Geoscience Center, Japan Atomic Energy Agency (JAEA). Our AMS system is a versatile system based on a 5MV tandem Pelletron type accelerator (National Electrostatic Corporation, US) and has been made available for ^{14}C-, ^{10}Be- and ^{26}Al-AMSs. These multi-nuclide AMSs have been mainly applied to neotectonics and hydrogeology, in support of our research on geosphere stability applicable to the long-term isolation of high-level radioactive waste. Furthermore, the ^{14}C- and ^{10}Be-AMSs are used for geoscience, environmental science and archaeology by researchers of universities and other institutes under the JAEA's common-use facility program.

Major contribution of radiocarbon (^{14}C) dating through our ^{14}C-AMS to geoscience studies are as follows. Yasue *et al.* identified fault displacement and stratigraphic correlation of black soils based on ^{14}C ages (presented in this conference). They conducted ^{14}C dating of the black soil collected from a trench wall of the Atera Fault, Gifu. The results of ^{14}C date show that the soil age varies from 4,000 to 2,000 y with depth of the sampling points and the soil was deposited at approximately constant rate. Imaizumi *et al.* (2006) estimated the faulting age based on ^{14}C dating of soils at the Senya Fault in the Toen Fault Zone, Yokote Basin, Akita. It was found that the ages range between 1000 - 1300 y, indicating that the Senya Fault was caused by the Rikuu Earthquake in the year of 1896. Sasaki *et al.* (2006) studied local climate change in an inland basin. Pollen records and ^{14}C ages of sediments in Ohkute Basin, Gifu were used to reconstruct past climate change. The results suggested that the local climate has been warmer for the last 10000 yBP.

Since the fiscal year of 2013, the ^{10}Be-AMS has been routinely measured and used to study long-term erosion rates of weathered granitic soil surfaces using cosmogenic ^{10}Be depth profile under the joint research program with the National Institute of Advanced Industrial Science and Technology (AIST). Recently, we have started development of ^{26}Al-AMS. The system tuning and test measurement have been carried out for routine measurement. The development has so far done well and the routine measurements of the ^{26}Al-AMS will be started in near future. The ^{10}Be- and ^{26}Al-AMSs will be used to estimate the exposure age of basement rocks as well as the sedimentation rate and the assessment of volcaniclastic material ejected during volcanic eruptions.

Keywords: AMS, Dating, C-14, Be-10, Al-26

Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-P02 Room:Poster Time:May 1 18:15-19:30

Radiocarbon dating of charcoal by the ABOX-SC method

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Charcoal is one of the most important samples for radiocarbon dating. It is necessary to remove contaminants from charcoal sample to obtain the reliable date. ABA (acid-base-acid) method is usually used for chemical pretreatment of charcoal: First, a sample is treated with HCl to remove carbonate contaminant. Next, the sample is treated with NaOH to remove organic contaminants derived from soil during burial. After then, the sample is treated with HCl again to remove absorbed atmospheric carbon dioxide during NaOH treatment. The residue is combusted with CuO at 850?C and graphitized to be 14C-dated.

However, the ABA treatment often cannot completely remove contaminants from poorly-preserved and/or old charcoals (>about 30 ka). Bird et al. (1999) showed that the ABOX-SC (acid-base-oxidation stepped combustion) method removes organic contaminants more efficiently than the ABA treatment. The age of the charcoal sample treated with the ABOX-SC was reported to be older than that of the charcoal sample treated with the ABA (Brock et al., 2010).

The ABOX-SC method consists of 3 step chemical pretreatments: HCl and NaOH treatments followed by K2Cr2O7-H2SO4 treatment in a sealed tube at 60?C for 20 hr (Brock et al., 2010). The mixed solution of K2Cr2O7 and H2SO4 removes organic contaminants effectively from charcoal samples, and can extract carbon fraction of oxidation resistant elemental carbon, OREC, which is resistant to oxidation and is less affected by contamination during burial (Bird et al., 1999). After the ABOX chemical treatment, the OREC is heated at 630?C with CuO for 2 hr to remove atmospheric CO2 contaminants adsorbed during sample treatment and contaminants remained after the ABOX treatment. Finally, the residue of OREC is oxidized completely into CO2 at 850?C for 1 hr, and the CO2 is graphitized to be 14C-dated.

In this study, we apply the ABOX-SC method to some charcoal samples of known age to measure 14C ages. The charcoal samples, which were excavated from Tang-e Sikan cave in Arsanjan city, Iran, have been dated at about 40 ka and 26 ka by the ABA method, and are considered to be attributed to Upper Paleolithic period. In this study, we confirm the age difference of Paleolithic charcoal samples by the two pretreatment methods of ABA and ABOX-SC.

Keywords: Radiocarbon, Charcoal, ABOX-SC method

Japan Geoscience Union Meeting 2014
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HTT35-P03 Room:Poster Time:May 1 18:15-19:30

AMS radiocarbon dating of Japanese tree rings for regional calibration curve

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Calibration of radiocarbon data can be achieved by comparison of measured radiocarbon age of samples with known calendar age. Tree rings that determined by dendrochronology are one of the important data set for calibration. IntCal13 calibration curve was launched mainly based on trees grown in the northern high latitude. These rings were sampled in ten years each at once to cancel the variation of solar activity, and to obtain sufficient sample size for conventional radiocarbon measurement as well. AMS radiocarbon dating can measure less than 1mg of carbon efficiently and is capable of date annual tree rings. Recent advance in accuracy of AMS radiocarbon measurement reveals that the resolution of IntCal may be insufficient for precise calibration. In particular, regional effect on the calibration curve had turned out to be a major problem. AMS radiocarbon dating of Japanese tree rings with actual age has been carried out to accomplish Japanese regional calibration curve. Tree ring preserves atmospheric ^{14}C concentration at that time, therefore the offset between radiocarbon age of Japanese tree ring and IntCal should indicate the inhomogeneity in the northen hemisphere.

Keywords: radiocarbon dating, tree ring, calibrated age, atmospheric inventory, regional effect

Japan Geoscience Union Meeting 2014
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HTT35-P04 Room:Poster Time:May 1 18:15-19:30

Radiocarbon dating of archeological remains related with the 13th century Mongol Inversion to Japan

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[1]Center for Chronological Research, Nagoya University

The shallow sea floor off Takashima, Matsuura, Nagasaki Prefecture, has been investigated archeologically as a potential site where many Mongolian warships exist under the sea sediments. It is historically recorded that more than 4000 Mongolian warships were destroyed by a typhoon during the Mongol invasion to Japan in 1281. The underwater investigations have been performed since 1980, and a lot of archeological remains related with the invasion have been collected there. In 2006, we were allowed to get some archeological remains for 14C dating with AMS. The samples were fragments of palm-bark ropes, lacquer products, bamboo ropes and charred rice. 14C ages for the samples were all consistent with the age of Mongol invasion in 1281.

Recently a body of submerged wrecks most probably originated from Mongolian warship has been discovered in the 1m-deep horizon of the sea sediment off Takashima. During the survey of the new warship, shell samples were collected near the ship. Some shells were recognized to be hull-fouling species, which may have grown up on the bottom of Mongolian warship and preserved along with the broken ship in the sea sediment. We have conducted 14C dating for some shell samples and found out that shells belong to hull-fouling species showed 14C ages consistent with the time of Mongol Inversion. Some other shells not belonging to hull-fouling species showed younger or older dates as compared with the time of Mongol invasion.

Keywords: AMS 14C dating, historical remains, remains from submerged wrecks, Mongol invasion, shell of hull-fouling species

Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-P05 Room:Poster Time:May 1 18:15-19:30

AMS radiocarbon dating of peaty layers in Kimotsuki lowland, southern Kyushu

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Since the middle Holocene, peaty layers have accumulated on the Kimotsuki lowland in south Kyushu, Japan. They can be applied radiocarbon (ˆ{14} C) wiggle-matching for establishing high-resolution chronology. Moreover, several tephra intercalated with the peat bed which originated from Kaimondake, Sakurajima, and the Kirishima volcanoes, and they can certify the reproducibility of ^{14} C dates. On the other hand, the age of the tephra by them can be determined correctly. We present here the results of AMS ^{14} C dating on the core sample, and report the age of each tephra determined from these dates.

Keywords: Kimotsuki lowland, peaty layer, tephra, radiocarbon date

Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-P06 Room:Poster Time:May 1 18:15-19:30

Offset in radiocarbon ages between shell and plant pairs in the Holocene sediments around the Korea

NAKANISHI, Toshimichi[1*] ; HONG, Wan[1] ; SUNG, Kisuk[1] ; SUNG, Kilho[1] ; NAKASHIMA, Rei[2]

[1]KIGAM, [2]AIST

Since 2009, a research project to evaluate the marine reservoir effects of the coastal sites of Korea has been progressed by KIGAM. Estimating the reservoir effect of this area is difficult because age-known marine samples obtained before AD 1950 are rare. In order to solve this problem, 61 sediment cores were collected with 1 m intervals by a percussion drilling tool from 52 coastal sites in the southern area of the Korean Peninsula. These drilling sites were roughly preselected by the interpretation of modern air photos of internet map services provided by the websites such as Daum and Google. Topographic maps in 1918-1926 with 1/50000 scale and old air photos were also used for the site selection. The length of each core was less than 5 m and the total length was 132 m. Based on analysis of lithology and mollusk assemblages, we selected marine shell and terrestrial plant pairs from same horizons. These samples were cleaned by physical and chemical pretreatments, and reduced by automatic graphitization system in KIGAM. The radiocarbon ages of the samples were measured by the AMS facility of KIGAM. This presentation will report about spatial and historical variation of radiocarbon marine reservoir effect around Korea.

Keywords: Radiocarbon dating, Marine reservoir effect, Coastal sediments, Korea

Japan Geoscience Union Meeting 2014
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HTT35-P07 Room:Poster Time:May 1 18:15-19:30

Estimation for the growth rate of benthic biotic communities in Antarctic lakes

TANABE, Yukiko[1*] ; UCHIDA, Masaki[2] ; KONDO, Miyuki[3] ; UCHIDA, Masao[3]

[1]Waseda Institute for Advanced Study, Waseda University, [2]National Institute of Polar Research, [3]National Institute for Environmental Studies

Antarctica is an ice continent. It has one of the most extreme environments for life in the world. There are very little ice-free regions with life in it, so these regions are sometimes called polar oasis within the polar desert. The ice-free regions are scattered along the coastal regions and around mountainous peaks (Nunataks) in continental Antarctica or concentrated in Antarctic Peninsula in maritime Antarctica. The biota is simple due to lack of remarkable organisms at the top of food webs, and primary producers such as cyanobacteria, algae, lichens, mosses, heterotrophic microorganisms and metazoans dominate the sparse communities. A remarkable diversity of lakes exists in Antarctica, ranging from hypersaline with nearly 10 times the conductivity of seawater, to brackish and freshwater, sub-glacial, permanently ice-covered and seasonally ice-covered lakes. These lakes are unproductive with typical photosynthetic levels of 0.5 — 30 μg-C/L/day from phytoplankton. Phytoplankton cannot bloom and hardly survives in the water column during the best light-available summer around the Syowa region. This results from low annual levels of photosynthetically active radiation and ice cover that attenuate light into the water column or photo-inhibit photosynthetic systems due to continuous low temperatures and the lack of any significant input of inorganic nutrients. Despite such severe situations, one of the most productive ecosystems in continental Antarctica is found in freshwater lakes, where benthic microbes form thick mats, and aquatic mosses can flourish on the lakebeds of the Syowa region. These lakes were exposed by glacial retreat after the Last Glacial Maximum. The benthic mats consist of almost organisms, dominantly cyanobacteria, algae, and mosses in the lakes, because there are a little inorganic particles and organic matters inflow to the lakes from the water catchment, and as pointed out above, almost no phytoplankton in the water column. This negligible level of any sedimentation and turbulence situation is specific to Antarctica, which is suitable to estimate the growth rate of benthic biotic communities in fine-scale. We collected sediment cores from 17 freshwater lakes in S$\hat{o}$ya Coast on the south area of Syowa station, continental Antarctica during January-February in 2009 — 2010. The core samples were vertically sliced in each 1 cm as soon as possible after sampling in the field hut, and transported to Japan at -20 ℃. Then, we analyzed the samples by using an AMS (accelerator mass spectrometry), and estimated the growth rate of the benthic biotic mats in each Antarctic lake.

Keywords: sediment, AMS, lakes, Antarctic, growth rate

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HTT35-P08 Room:Poster Time:May 1 18:15-19:30

Black SOM dynamics during reforestation of Japanese grassland

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The dynamics of the polyaromatic structures of black humic acids (HAs), which are presumably derived from charred materials, are of significant interest for the global carbon cycle. However, the details of those dynamics are not yet well understood. We investigated differences in the degree of darkness (A600/C values), isotopic ratios (δ13C, δ15N, and Δ14C values), and 13C NMR spectra of size-separated black HAs extracted from Japanese volcanic ash soils in order to estimate the variations in the polyaromatic structures of black HAs during ca. 100 years of natural reforestation of Japanese pampas grassland. For several hundred years, all the study sites were managed similarly as grassland by burning. Subsequently, their management differed: at site G (Miscanthus sinensis: C4 plant), maintenance as of the time of this study was still performed by mowing, while at sites P (Pinus densiflora: C3 plant) and Q (Quercus crispula: C3 plant), maintenance was discontinued ca. 30 and 100 years ago, respectively. Thus, the sites range from grassland (site G) to coniferous forest (site P) to broad-leaved forest (site Q). For all HA size fractions at all sites, we found that δ13C values correlate positively with δ15N values, although the gradients are much lower for fractions of small to medium molecular size than for fractions of medium to large molecular size (denoted as lower-size and higher-size fractions, respectively). Overall, for the lower-size fractions, the contribution ratio of C4-plant-derived carbon shows a significant positive correlation with A600/C values and a negative correlation with Δ14C values, and their aromatic characteristics are greater than those of higher-size fractions within the same black HA. Furthermore, the relative proportion of lower-size fractions decreases with reforestation, especially from site P to Q. The δ13C values for all size fractions are similar for sites G and P, but are relatively low for site Q. The aryl C contents of the lower-size fractions are lower and the O-alkyl C contents and the aliphaticity (alkyl C:O-alkyl ratio) are clearly higher for sites P and Q than for site G. These results strongly suggest that stimulation of HA biodegradation might be achievable by continuous input of new plant litter during reforestation, even for lower-size HA polyaromatic structures, despite the fact that lower-size HAs biodegrade more slowly than higher-size HAs.

Keywords: land use, reforestation, soil organic matter, 14C

Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-P09 Room:Poster Time:May 1 18:15-19:30

Potential sink of soil organic carbon in a Japanese cool-temperate forest based on bomb radiocarbon based residence time

KONDO, Miyuki[1*] ; UCHIDA, Masao[1] ; OHTSUKA, Toshiyuki[2] ; MURAYAMA, Shohei[4] ; SHIRATO, Yasuhito[3] ; SHIBATA, Yasuyuki[1]

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Functional roles of SOC pool for carbon dynamic remains almost unknown. In this study, residence time (RT) based on carbon and radiocarbon (14C) inventories, was f investigated in a Japanese temperate forest (Takayama) under Asian Monsoon climate, and the potential of soil carbon sequestration were also investigated. Soil organic matter was divided to two fractions as low density humified material (LOM) and high density mineral-associated material (HOM). Our results were thoroughly compared with those in a temperate forest (Harvard forest) conducted using a similar approach [Gaudinski et al., 2002]. The LOM was the major part of the SOC (76%) and its contribution was higher even in the deep layer. 14C contents of LOM in surface layer were similar to those of atmospheric CO2 and roots, whereas those in deep layer are significantly low (?14C <-200 per mil) as well as HOM fractions, although LOM fraction seems to consist of labile carbon. RTs for low density fractions as derived from their radiocarbon content are 53 ?330 yrs BP in surface layer and 1760 ?2780 years BP. Storage of SOC in our site was larger, irrespective of depths and differed considerably from that in Harvard forest. We also measured soil 14CO2 profile to determine the rate of CO2 production from heterotrophic respiration of two SOM fractions. The ?14C values of soil CO2 profile was constant down to 75 cm depth, which were close to those of atmospheric CO2 and fine roots, suggesting that most of soil CO2 is derived from recent photosynthetic fixed C. These results indicate that this forest might be higher sequestrating soil carbon as low density fractions semi-permanently, which is also concerned about instability of near future climate change.

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HTT35-P10 Room:Poster Time:May 1 18:15-19:30

Source diagnosis of PAHs using compound class specific 14C analysis and Monte Carlo source appotionment at Kolkata canal

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Atmospheric polycyclic aromatic hydrocarbons (PAHs) originate mostly from incomplete combustion of carbon-based fuels. PAHs account for most (35-82%) of the total mutagenic activity of ambient aerosols. Reduction of air pollution by PAHs is essential for an effective air quality control, which requires reliable source apportionment. It has been reported that atmospheric pollution by PAHs in Indian megacities, such as Kolkata, Mumbai and Chennai, is comparable to the highest levels across the globe and Kolkata air exhibit the highest level among them. Also, our previous survey revealed that sediments from Kolkata city canals have the highest PAHs concentrations (i.e., 15.9±11.6 μg of $\sum$14-parent PAHs/g dw, n=12) among the 174 surface sediments from 8 tropical Asian countries.[1] Examining methylated-to parental PAHs ratios of three homologous series and C30-hopane/$\sum$PAHs ratios both in sediment samples and in probable source materials, the high level sedimentary PAHs were ascribed to those emitted from combustion sources. However, relative importances of combustion sources were not solved. The present study aimed to apportion sources of combustion to PAHs in highly contaminated sediments from Kolkata, India by using combined approaches of CCSRA technique, molecular fingerprinting and Monte Carlo source apportionment.

Furthermore, three- and four ring PAHs (MW178, 192, 202) in leftover extracts were harvested on PCGC and analyzed for 14C on AMS at NIES-TERRA, NIES (Tsukuba, Japan). PAHs isolated from Kolkata canal sediments showed mostly fossil carbon isotopic signatures, i.e., ^{14}C signal of PAHs with MW178, 202 and those with MW?228 were 10.6±0.1, 5.9±0.4, 7.6±0.5 pMC (KKNC), 8.4±0.5, 8.3±0.4, 8.5±0.3 pMC (KKSC). By using source end-members of MW202 and MW276 isomer pair ratios, Monte Carlo source apportionment[2] revealed that most of fossil-PAHs were derived from coal combustion, i.e., relative contributions (median) from coal and petroleum combutions were 50% and 11% in KKNC and 13% and 56% in KKSC.

1. Saha M. *et al., Mar. Pollut. Bull.*, **2009**, 58 (2), 189-200
2. Sheesley R.J. *et al., Atmos. Environ.*, **2011**, 45(23), 3874-3881

Keywords: compound class specific radio carbon analysis, PAHs, monte carlo simulation, molecular fingerpringing, source apportionment

Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-P11 Room:Poster Time:May 1 18:15-19:30

Accurate age estimation using ^{14}C in human teeth enamel.

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Radiocarbon (^{14}C) concentration in the atmosphere showed a stable value until 1955. However, as a result of the nuclear bomb testing, modern ^{14}C concentration in the atmosphere dramatically increased during late 1950s and early 1960s. These bomb-produced ^{14}C is then oxidized to form CO_2, and incorporated into plants by photosynthesis. Then, by eating plants or animals fed by these plants, the ^{14}C concentration in human body reflects the ^{14}C value of atmospheric CO_2 at a certain time. Recent studies insisted that these ^{14}C can play important role for forensic analysis, especially age estimation using ^{14}C in human teeth enamel.

Teeth enamel is such a harder part of the human body that they are hardly destroyed by a natural process. And, the most important is, there is no turnover of enamel after its formation has completed. Although there are previous works which estimate the birth year of individuals by using ^{14}C concentration in enamel, their samples are teeth from Swedish, Scottish and American people, and study areas are mainly at high latitudes of the northern hemisphere. The precision of age estimation using teeth enamel is determined by enamel formation time of teeth and atmospheric ^{14}C concentration in a certain area at a certain year. It is known that teeth formation time of Japanese is different from that of Caucasian. It has been found that ^{14}C concentration in the atmosphere indicates 5 different zones according to different peak ^{14}C concentration of the nuclear bomb testing. These zones are named NH zone1, NH zone2, NH zone3, SH zone3 and SH zone1-2 from north to south. The boundary between NH zone1 and NH zone2 is Ferrel cell - Hadley cell boundary. It is nearly located at 35 ^{o}N. So it means that previous works mainly focused on NH zone1 samples, not NH zone2 samples. One of NH zone2 samples, teeth enamels of Japanese have not studied sufficiently. The aim of this study is to clarify whether age estimation using teeth enamels of Japanese can determine the precise year of birth of individuals and to discuss the mechanism of carbon fixation of enamels or other parts of the teeth.

7 of 44 collected tooth samples have been analyzed. They are 5 third molars and 2 second molars. The year of birth of each individual is 1943, 1946, 1951, 1951, 1951 for third molars, and 1933 and 1959 for second molars. In order to get the estimated year of birth, a model age for enamel completion of Japanese was subtracted from the year given by the ^{14}C analysis of samples. The result shows that age estimation using teeth of Japanese gives precise age determination. Needless to say, taking account of the degree of individual variation and possibility of differences in local environment or in diet is important, however, this result seems to be uninfluenced by those effects. Larger number of, more and more various parts of teeth (for example, first molars, anteriors, such as early-completed teeth.) have to be analyzed.

To determine whether an individual is born before or after the peak of atmospheric ^{14}C concentration (in 1964, in NH zone2), root of teeth have to be analyzed. Since root completion age is some years after enamel completion age, it can be easily found that the sample age is whether rising or falling part of the atmospheric ^{14}C curve. We now are preparing for analysis of ^{14}C of root dentine collagen and root inorganic matter. Their results will give more compelling data, now discover what is waiting for you!!

Keywords: human tooth, enamel, ^{14}C, forensic science, nuclear bomb testing, age estimation

Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-P12 Room:Poster Time:May 1 18:15-19:30

Observations of atmospheric radiocarbon in carbon dioxide at Hateruma Island and Cape Ochi-ishi, Japan

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Atmospheric radiocarbon in carbon dioxide ($^{14}CO_2$) is a powerful tracer for understanding of carbon cycles, e.g. oceanic and biospheric CO_2 exchanges and CO_2 emissions from fossil fuel combustion. Observation sites for radiocarbon concentrations, $\triangle^{14}C$, are not many enough to evaluate the global and regional carbon flux. We present an analysis of trends, interannual variability (IAV) and seasonal cycle of $^{14}CO_2$ in background air from July 2004 to December 2012 at two NIES/CGER monitoring stations; Hateruma Island (HAT; latitude 24.06N, longitude 123.81E) and Cape Ochi-ishi (COI; latitude 43.16N, longitude 145.50E). The air samples were collected in 2 L Pyrex glass flasks. The sampling frequency was monthly. CO_2 was extracted from the whole air at NIES and CO2 samples were converted to graphite and analyzed ratios of $^{14}C/^{12}C$ by accelerator mass spectrometry (AMS, National Electrostatics Corp., 1.5SDH) at Paleo Labo Co., Ltd., Japan. Analytical precision in $\triangle^{14}C$ determined from statistical uncertainty (number of ^{14}C counts) was $\pm$1.7-2.0 ‰ for most samples. The repeatability of measurements using modern reference air was $\pm$1.9 ‰ . A decreasing trend in $\triangle^{14}C$ was -5 ‰ yr^{-1} in average but large IAV was observed at both stations: large decreases in 2007-2008 and in 2010-2011 (-8 to -9 ‰ yr^{-1}) and almost zero decrease in 2009. We also observed clear seasonal cycle of $\triangle^{14}C$. The peak-to-peak amplitudes in the seasonal cycle determined from the smooth curve fits were 7 ‰ at both stations and the maximum of $\triangle^{14}C$ appeared in July and the minimum in January at HAT, and the maximum in September and the minimum in May at COI. The differences in phase of $\triangle^{14}C$ seasonal cycle between HAT and COI suggested that the atmospheric $\triangle^{14}C$ at COI was influenced by CO_2 emitted from terrestrial biosphere.

Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-P13 Room:Poster Time:May 1 18:15-19:30

Radiocarbon based source apportioning of PM2.5 carbonaceous aerosols at Cape Hedo, Okinawa and Fukue island, Japan

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Radiocarbon (14C) analysis of the carbonaceous aerosol allows an apportionment of fossil and non-fossil sources of air-borne particulate matter (PM). A chemical separation of total carbon (TC) into its sub-fractions organic carbon (OC) and elemental carbon (EC) refines this powerful technique, as OC and EC originate from different sources and undergo different processes in the atmosphere. Although 14C analysis of TC, EC and OC has recently gained increasing attention, Nowadays gigantic brownish haze from various burning and combustion processes is also blanketing India and surrounding land and oceans during the winter season. In China and surrounding countries, same kind of atmospheric pollution are widely observed and occurred as well. Additionally this soot-laden Brown Cloud is affecting South and East Asian climate as much or more than carbon dioxide and cause hundreds of thousands of premature deaths annually, yet its sources have been poorly understood. In this study, we investigated the contribution of continent derived aerosol to Japan. Aerosol samples with diameter of 2.5mm were collected at Fukue island, one of Goto islands and at the Cape Hedo is located at the northern end of Okinawa Island. The 14C contents of EC of PM2.5 aerosols collected from October, 2009 and May, 2010 including the Kosa event in Cape Hedo and Fukue were measured. The 14C content represents in the unit of pMC . Results of EC-14C in both sites were 25-30pMC in Cape Hedo and 18-44pMC in Fukue, respectively. These results mean that relative apportions of biomass burning and fossil fuel were 25-30% and 18-44% in Cape Hedo and 25-35% and 65-75% in Fukue, respectively. The observed variations of pMC in Cape Hedo during February and March were relatively smaller than those of Fukue, which was more than 20%. According to back trajectory analysis in this duration, because ca. 70% of air mass in both sites was derived from the continent. The aerosols particulate matter to be transferred to Cape Hedo from continent would be relatively smaller than those to Fukue. Our data of EC-14C obtained during the Kosa event showed the relatively higher contribution of biomass burning sources in Fukue although these interpretation need to consider variation of the magnitude and concentration of EC in both sites. In further study we need to investigate details of the source of EC during this period.

Keywords: Radiocarbon, PM2.5, aerosol, source apportioning

Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-P14 Room:Poster Time:May 1 18:15-19:30

Study on monitoring of volcanic activity using ^{129}I / ^{127}I ratios in crater lake and hot spring at Zao volcano

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Volcanic tremors and mountain gradient changes have been detected at Zao volcano in Miyagi and Yamagata since January 2013, volcanic activity began to intensify although Zao volcano will not erupt immediately[1]. Since the water quality of crater lake are correlating with volcanism changes[2][3], basic water quality of crater lake and hot spring at Zao volcano have been studied by the group of Tohoku University from September 2013. As a part of this project, we are trying to monitor the volcanic activity using ^{129}I / ^{127}I ratios (atomic ratio of radioiodine and stable iodine) in crater lake and hot spring of Zao volcano.

Natural ^{129}I (half-life: 15.7 million year) are produced by nuclear spallation reaction of ^{129}Xe with cosmic ray in the atmosphere and spontaneous fission of ^{238}U in the geological layer. In the ocean, steady-state ^{129}I / ^{127}I ratio of the seawater is estimated to be 1.5×10^{-12}[4]. Sunken iodine by the ocean plate having lower ^{129}I / ^{127}I ratio (older ^{129}I age) compared to the steady-state ratio of seawater, are supplied to the atmosphere mainly via magmatic activity. In general, ^{129}I / ^{127}I ratio in hot spring water and brine water are used as indicator of origin and behavior of iodine in the water[5][6]. ^{129}I / ^{127}I ratio of hydrothermal at Zao volcano are considered to become lower by the supply of chronologically-old iodine in terms of global iodine cycle.

In September 2013, water samples of 2 L were collected from the surface of crater lake (Okama, diameter: 350 m, maximum depth: 35 m) located at 1,560 m in elevation and hot spring (Kamoshika Hot Spring) located at 1,230 m in elevation in the eastern side of Zao volcano. Water temperature and pH were measured on site. After water samples were filtered by 0.2 μm filter, ^{129}I / ^{127}I ratio were measured for the isotopic diluted water samples by adding carrier (^{127}I standard) at MALT, The University of Tokyo. ^{127}I concentrations were measured by ICP-MS, and original ^{129}I / ^{127}I ratio of water samples were estimated.

Water temperature and pH were 10.2 ℃ and 3.3 at Okama; 40.0 ℃ and 3.3 - 4.0 at Kamoshika Hot Spring. ^{129}I / ^{127}I ratios of Okama and Kamoshika Hot Spring were respectively, estimated to be $(1.5 \pm 0.4) \times 10^{-9}$ and $(0.78 \pm 0.2) \times 10^{-9}$, 500 - 1000 times higher than the steady-state ratio of sea water (1.5×10^{-12})[4]. Since ^{129}I / ^{127}I ratio of anthropogenic metric water were over 9.0×10^{-12}[7], surface water of Okama and Kamoshika Hot Spring water were very likely to be strong affected by the meteoric water including anthropogenic ^{129}I. For the monitoring of volcanic activity using ^{129}I / ^{127}I ratio, it is necessary to decide the site as few anthropogenic ^{129}I as possible through the measuring of ^{129}I / ^{127}I ratio of the Okama bottom water and some hot spring around Zao volcano. Continuous water quality survey of 1 - 2 times for Okama and 1 time per 1 - 2 months for hot springs are planned from June to November of this year.

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Keywords: Zao volcano, volcanic activity, crator lake, hot spring, ^{129}I / ^{127}I, AMS

Japan Geoscience Union Meeting 2014
(28 April - 02 May 2014 at Pacifico YOKOHAMA, Kanagawa, Japan)

HTT35-P15 Room:Poster Time:May 1 18:15-19:30

Speciation analysis of the Fukushima accident derived I-129 in the soil using sequential extraction method

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In previous study, we investigated the depth profile of the accident derived ^{129}I ($T_{1/2}$ = 1.57x10^7 y) and downward migration speed in soils of near field of Fukushima Dai-ichi Nuclear Power Plant (FDNPP), including crop fields and man-made fields. ^{129}I in soil was measured by AMS and stable iodine (^{127}I) was measured by ICP-MS at MALT (Micro Analysis Laboratory, Tandem accelerator), The University of Tokyo. It was found that ^{129}I was concentrated near surface but distributed deeper compared with ^{137}Cs ($T_{1/2}$ = 30 y). From the estimation of relaxation length using depth profiles, the FDNPP derived ^{129}I move 0.6 cm/y downward and ^{137}Cs 0.3 cm/y for it. It was also found that ^{129}I seems to move downward more quickly than ^{137}Cs.

To investigate the adsorption mechanism and the elemental process of migration of the accident derived ^{129}I in soil, it is important to know what kind of component the ^{129}I combines with.

Recent studies on the X-ray absorption fine structure (XAFS), especially near edge structure (XANES), reported that the stable iodine (^{127}I) in soil existed as an organic component[1]. However, it had not yet been proved that it was also the case with the accident derived ^{129}I because it had been incorporated in the soil system only recently and the abundance of ^{129}I in soil was more than 8 orders of magnitude smaller than sub-ppm level stable iodine (^{127}I).

In this study a progressive sequential extraction method including the dialysis was newly developed to obtain only the iodine sticking to the soil organic component. The advantage of sequential extraction over other method is that stable iodine can be quantified by direct analysis of the fraction and ^{129}I can be quantified by AMS method of the fraction added with carrier. The fraction of the organic component for ^{127}I and ^{129}I can be evaluated respectively by comparing with the other fraction and/or with the total concentration obtained by the bulk analysis (e.g. by the pyrohydrolysis).

Repeatability is 20% for the water soluble, oxides and organic fraction, 10% for Exchangeable fraction and 50% Residue (mainly minerals).

The results show that 60% of the total ^{129}I are associated with oxides and 30% associated with organic matter in crop field soil. The former, the oxides bond iodine, it takes a form of iodate (IO_3^-) absorbed in amorphous oxides, especially goethite or delta-MnO_2. They are formation of monodentaten mononuclear outer-sphere species and bidentate, binuclear inner-sphere species [2]. The latter iodine are linked to organic carbon directly by a covalent bond.

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