

日本原子力研究開発機構東濃地科学センター JAEA-AMS-TONO における加速器質量分析装置を用いた地球科学研究  
Geoscience studies using by AMS at JAEA-AMS-TONO in the Tono Geoscience Center of the Japan Atomic Energy Agency

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JAEA-AMS-TONO は、1997 年に日本原子力研究開発機構 東濃地科学センターに設置された。当 AMS は、5MV タンデム型ペレトロン加速器を有し、<sup>14</sup>C、<sup>10</sup>Be 及び <sup>26</sup>Al 測定を行い、高レベル放射性廃棄物の地層処分に関わる地質環境の安定性研究などに利用している。また、<sup>14</sup>C 及び <sup>10</sup>Be 測定は、機構で行う施設供用制度のもと、地球科学や環境科学、考古学などの分野で大学や他の研究機関の方にも利用していただいている。

以下に、当 AMS を使った地球科学研究での放射性炭素 (<sup>14</sup>C) 年代測定について紹介する。安江らは、<sup>14</sup>C 年代を用いた黒色土の層序対比と断層変位基準について検討している (本大会で発表予定)。岐阜県阿寺断層のトレンチから採取した黒色土の <sup>14</sup>C 年代を調べ、土壌の年代は深い方から 4000 年から 2000 年であり、土壌はほぼ一定の速度で堆積していることを示した。また、今泉ら (2006) は、秋田県横手盆地東縁断層地帯の千屋断層において <sup>14</sup>C 年代を用いて断層の活動時期を推定した。土壌の年代は 1000~1300 年を示し、千屋断層は 1896 年に起こった陸羽地震によって生じたことを明らかにした。佐々木ら (2006) は、内陸の気候変動について調べ、岐阜県大湫盆地から採取した花粉と堆積物の <sup>14</sup>C 年代から、一万年 BP 前から温暖期が始まっていることを示した。

平成 25 年度より <sup>10</sup>Be のルーチン測定を開始し、宇宙線生成核種である <sup>10</sup>Be の深度分布を用いた花崗岩地帯の長期的な侵食速度の推定に関わる研究を産業技術総合研究所とともに開始した。また、近年 <sup>26</sup>Al のルーチン測定に向け、装置調整や試験測定を行っている。整備は順調に進んでおり、近年中のルーチン測定化を目指している。今後、岩体の露出年代や堆積速度、火山碎屑物の評価等に <sup>10</sup>Be 及び <sup>26</sup>Al 測定を利用していく予定である。

キーワード: 加速器質量分析, 年代測定, C-14, Be-10, Al-26  
Keywords: AMS, Dating, C-14, Be-10, Al-26

## ABOX-SC法を用いた炭化物の<sup>14</sup>C年代測定 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 <sup>14</sup>C-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 K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>-H<sub>2</sub>SO<sub>4</sub> treatment in a sealed tube at 60°C for 20 hr (Brock et al., 2010). The mixed solution of K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> and H<sub>2</sub>SO<sub>4</sub> 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 CO<sub>2</sub> contaminants adsorbed during sample treatment and contaminants remained after the ABOX treatment. Finally, the residue of OREC is oxidized completely into CO<sub>2</sub> at 850°C for 1 hr, and the CO<sub>2</sub> is graphitized to be <sup>14</sup>C-dated.

In this study, we apply the ABOX-SC method to some charcoal samples of known age to measure <sup>14</sup>C 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

## 日本産樹木年輪の AMS 炭素 14 年代測定による較正曲線の作成 AMS radiocarbon dating of Japanese tree rings for regional calibration curve

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炭素 14 年代を暦上の実際の年代に修正するには、年輪年代法などで実年代の判明した樹木年輪の炭素 14 年代をもとにした「較正曲線」が用いられる。先に公開された北半球用の較正曲線「IntCal13」は主に北半球高緯度地域の樹木年輪の測定結果に基づいているが、太陽活動の 11 年周期を相殺するため、また炭素 14 年代測定に必要な試料量を確保するためにも多くは 10 年単位の年輪が測定されている。AMS による炭素 14 年代測定 (AMS-<sup>14</sup>C 法) は 1mg 以下の炭素量でも測定が可能なことから、1 年単位の樹木年輪などより詳細な測定に応用できる。近年は測定精度が向上し、これまで較正曲線の誤差に隠されてきた炭素 14 濃度における北半球内の不均質性が次第に明らかになりつつある。日本版較正曲線の作成を目指して、これまで継続されてきた AMS による日本産樹木の炭素 14 年代の測定結果を紹介する。

キーワード: 炭素 14 年代法, 樹木年輪, 較正年代, 大気成分, 地域効果

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

## 長崎県鷹島沖海底泥土中から引き上げられたされた元寇関連資料の 14C 年代測定 Radiocarbon dating of archeological remains related with the 13th century Mongol Invasion to Japan

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2011 年 10 月 24 日の夕刊で、鎌倉時代に来襲した元寇の沈没船とみられる船体が長崎県松浦市の鷹島沖で見つかった事が大々的に報道された。元寇の際に使われた船とされる木材などが過去に見つかったが、今回は船の構造がわかる状態で発見されており、元寇船復元の研究が大いに進むことが期待されている。

著者らは、2006 年 5 月 10?11 日と長崎県松浦市鷹島町の松浦市立鷹島歴史民俗資料館および鷹島埋蔵文化財センターを訪問し、既に鷹島周辺の海底から採取され保存されていた元寇関連資料を分取させて頂いた。この調査は、科学研究費補助金基盤研究 (B)「海底遺跡出土遺物の調査・分析・保存に関する基礎的研究」を遂行するうえで企画したものである。海底から採取された考古学・文化財科学における遺物が正しく元寇に関連するものであることを年代測定から確かめるために、採取した試料について、名古屋大学年代測定総合研究センター所有のタンデトロン加速器質量分析計による放射性炭素年代測定を行った。

その後、2011 年に発見された沈没船の調査が進み、鷹島沖合の水深 20?25m の海底において、約 1m に砂泥に埋まって見つかった沈没船の船体の竜骨 (キール) の調査において採取されたさまざまな遺物の中から、貝殻類を分析研究に提供頂けることになった。採取された貝殻には、浅海底の何処かに以前生息していたものであり沈没船と何の関わりも持たないものが多いが、一方では、航行使用中の船の船底に付着して生育する船体汚損種の貝も含まれている。そこで、提供して頂いた貝殻の 14C 年代測定を行った。これらの結果をここに報告する。

キーワード: AMS 14C 年代測定, 歴史資料, 沈没船遺物, 元寇, 船体汚損貝

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

## 南九州，肝属低地の泥炭層の AMS 放射性炭素年代 AMS radiocarbon dating of peaty layers in Kimotsuki lowland, southern Kyushu

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南九州の肝属低地には、中期完新世以降の泥炭層が堆積しており、その放射性炭素 (<sup>14</sup>C) 年代を使ったウイグルマッピングが適用可能である。また、泥炭層には開聞岳、桜島および霧島火山から飛来した複数のテフラが挟在しており、これらを用いて、<sup>14</sup>C 年代の再現性を検証することができる。一方、それらによってのテフラの年代を正確に決定することができる。このポスター発表では、ここで得られたコア試料についての <sup>14</sup>C 年代を示し、これらから決定した各テフラの年代を報告する。

キーワード: 肝属低地, 泥炭層, テフラ, 放射性炭素年代

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

## 韓国沿岸の完新統中の貝と植物の放射性炭素年代値の差異 Offset in radiocarbon ages between shell and plant pairs in the Holocene sediments around the Korea

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2009 年以降、韓国地質資源研究院において韓国沿岸の放射性炭素海洋貯留効果を評価するための研究プロジェクトが実施されてきた。採取年代が特定されている西暦 1950 年以前の海洋試料が少ないので、この地域の放射性炭素海洋貯留効果の評価は難しかった。そこで、61 本の堆積物コア試料を打撃式ボーリングコア採取器によって 52 地点から 1m 毎に採取した。採取地点は Daum や Google といったウェブサイトの空中写真の判読によって選定され、1918-1926 年に作成された 1/50000 の地形図や古い空中写真も参考にされた。それぞれのコア試料の長さは最大で 5m であり、すべての試料の全長は 132m である。岩相や貝の群集組成の解析結果によって、同じ層準に含まれている貝と植物片が選定された。これらの試料は物理的と化学的に繰り返し洗浄され、KIGAM の自動グラフィット化装置によって還元され、KIGAM の AMS システムによって放射性炭素年代値が測定された。この発表では韓国沿岸の放射性炭素海洋リザーバー効果の時空間的な多様性について報告します。

キーワード: 放射性炭素年代測定, 海洋貯留効果, 沿岸堆積物, 韓国

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

## 南極湖沼群における湖底生物群集の生長速度推定 Estimation for the growth rate of benthic biotic communities in Antarctic lakes

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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  $\mu\text{g-C/L/day}$  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 Sôya 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  $-20\text{ }^{\circ}\text{C}$ . 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.

キーワード: 湖底堆積物, 加速器質量分析, 湖沼, 南極, 成長速度  
Keywords: sediment, AMS, lakes, Antarctic, growth rate

## 林地化に伴う黒色土壌有機物動態 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 ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ , and  $\Delta^{14}\text{C}$  values), and  $^{13}\text{C}$  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  $\delta^{13}\text{C}$  values correlate positively with  $\delta^{15}\text{N}$  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  $\Delta^{14}\text{C}$  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  $\delta^{13}\text{C}$  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.

キーワード: 土地利用, 林地化, 土壌有機物,  $^{14}\text{C}$

Keywords: land use, reforestation, soil organic matter,  $^{14}\text{C}$

## 放射性炭素同位体を用いた冷温帯落葉広葉樹林における土壌有機炭素蓄積能の評価 Potential sink of soil organic carbon in a Japanese cool-temperate forest based on bomb radiocarbon based residence time

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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 (<sup>14</sup>C) inventories, was 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. <sup>14</sup>C contents of LOM in surface layer were similar to those of atmospheric CO<sub>2</sub> and roots, whereas those in deep layer are significantly low (<sup>14</sup>C < -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 <sup>14</sup>CO<sub>2</sub> profile to determine the rate of CO<sub>2</sub> production from heterotrophic respiration of two SOM fractions. The <sup>14</sup>C values of soil CO<sub>2</sub> profile was constant down to 75 cm depth, which were close to those of atmospheric CO<sub>2</sub> and fine roots, suggesting that most of soil CO<sub>2</sub> is derived from recent photosynthetic fixed C. These results indicate that this forest might be higher sequestering soil carbon as low density fractions semi-permanently, which is also concerned about instability of near future climate change.

## 化合物群レベル放射性炭素同位体分析 (CCSRA) と分子組成解析を用いたインドコルカタ運河堆積物中 PAHs の起源識別 Source diagnosis of PAHs using compound class specific $^{14}\text{C}$ analysis and Monte Carlo source apportionment at Kolkata canal

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多環芳香族炭化水素 (PAHs) はその有害性から主要な大気汚染物質として注目されている。その排出を制御するために起源の定量的識別が求められている。近年、分取キャピラリー GC (PCGC) と加速器質量分析計 (AMS) を用いた極微量スケール放射性炭素同位体測定を組み合わせたにより、環境試料中の個別の有機化合物または化合物群について放射性炭素年代を測定できるようになった。例えば Kanke ら<sup>1</sup> や Kumata ら<sup>2</sup> は、この化合物レベル放射性炭素同位体測定 (CSRA) を堆積物やエアロゾル試料中の PAHs に適用し、化石炭素と現代炭素の寄与を識別することに成功している。しかし、堆積物試料など石油燃焼と石炭燃焼など異なる種類の化石燃料の寄与が想定されるケースでは、CSRA のみでの起源推定には不十分であった。

本研究では、インド第 3 の都市コルカタ (人口約 1500 万人) 市内の運河から高濃度の PAHs で汚染された堆積物を採取し、PAHs の化合物群レベル放射性炭素同位体測定 (CCSRA) によってバイオマス/化石炭素の起源を識別したほか、メチル化 PAHs/PAHs 比、石油起源マーカー物質であるホパン、PAHs の異性体比のモンテカルロシミュレーションによる 3 エンドメンバーモデルでの解析を併用した複合的なアプローチによって、PAHs の汚染源を解析した。

これまでの調査から、コルカタ市内運河の  $\Sigma$  14-parental-PAHs 濃度は  $15.9 \pm 11.6 \mu\text{g/g}$  ( $n=12$ ) と、他の熱帯アジア地域 8 カ国都市域の沿岸堆積物中 PAHs の濃度範囲 ( $0.21 \pm 0.17 \mu\text{g/g}$  (マレーシア  $n=17$ )  $\sim 1.76 \pm 1.53 \mu\text{g/g}$  (カンボジア  $n=4$ )) と比べ、極端に高いことが分かっている<sup>3</sup>。

PAHs 生成温度指標であるメチル化 PAHs/PAHs 比が  $0.47 \pm 0.24$  と低いことから、燃焼由来の PAHs の影響が強いと考えられた。

コルカタでの燃焼起源は、自動車排ガス、レンガ製造での石炭燃焼、家庭用調理ストーブでの薪、石炭燃焼があげられる。C30-hopane/ $\Sigma$  PAHs 比 ( $0.09 \pm 0.05$ ) も含めた分子組成解析から、このうちガソリン車排ガスと家庭用調理ストーブでの石炭燃焼については、影響は限定的と判断できた。

コルカタ運河堆積物から精製した  $\Sigma$  178 (フェナントレン+アントラセン)、 $\Sigma$  202 (フルオランテン+ピレン)、 $\square$  HMW (分子量 228 以上のペアレント PAHs の合計) の放射性炭素同位体比 ( $p\text{MC} \pm \sigma$ ) は、それぞれ  $10.6 \pm 0.1$ ,  $5.9 \pm 0.4$ ,  $7.6 \pm 0.5$  (KKNC)、 $8.4 \pm 0.5$ ,  $8.3 \pm 0.4$ ,  $8.5 \pm 0.3$  (KKSC) であり、化石炭素由来の PAHs が卓越した。2000 年 (試料採取から 6 年前) の大気中  $\text{CO}_2$  と化石炭素をエンドメンバーとした同位体マスバランス計算から、 $\Sigma$  178、 $\Sigma$  202、 $\Sigma$  HMW-PAH へのバイオマス燃焼の寄与率はそれぞれ 9.8, 5.5, 7.0 % (KKNC)、7.8, 7.7, 7.9 % (KKSC) と試算された (Fig)。化石燃料燃焼に由来する残りの 90-94% の PAHs を、石炭燃焼 (レンガ製造) とディーゼル排ガスの 2 種類の混合によるものと仮定し、MPy/Py 比を用いて計算すると、石炭燃焼 (レンガ製造) とディーゼル排ガスの寄与率はそれぞれ 60~65%、29~33% と試算された。文献に報告されている石油、石炭、バイオマスの各種燃料の燃焼由来 PAHs の分子量 202 (フルオランテンとピレン)、276 (インデノ [123-cd] ピレンとベンゾ [ghi] ペリレン) の異性体比をエンドメンバーとして用いたモンテカルロシミュレーション<sup>4</sup> でも、石油、石炭燃焼由来 PAHs の寄与率はそれぞれ 11%, 50% (KKNC)、13%, 56% (KKSC) と、化石炭素起源 PAHs の大部分が石炭燃焼によることを示した。

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キーワード: 化合物群レベル  $^{14}\text{C}$  測定, PAHs, モンテカルロシミュレーション, 分子組成解析, 起源識別

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

## ヒト歯牙エナメル質中<sup>14</sup>Cを用いた人体年齢の精密推定 Accurate age estimation using <sup>14</sup>C in human teeth enamel.

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Radiocarbon (<sup>14</sup>C) concentration in the atmosphere showed a stable value until 1955. However, as a result of the nuclear bomb testing, modern <sup>14</sup>C concentration in the atmosphere dramatically increased during late 1950s and early 1960s. These bomb-produced <sup>14</sup>C is then oxidized to form CO<sub>2</sub>, and incorporated into plants by photosynthesis. Then, by eating plants or animals fed by these plants, the <sup>14</sup>C concentration in human body reflects the <sup>14</sup>C value of atmospheric CO<sub>2</sub> at a certain time. Recent studies insisted that these <sup>14</sup>C can play important role for forensic analysis, especially age estimation using <sup>14</sup>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 <sup>14</sup>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 <sup>14</sup>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 <sup>14</sup>C concentration in the atmosphere indicates 5 different zones according to different peak <sup>14</sup>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 °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 <sup>14</sup>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 <sup>14</sup>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 <sup>14</sup>C curve. We now are preparing for analysis of <sup>14</sup>C of root dentine collagen and root inorganic matter. Their results will give more compelling data, now discover what is waiting for you!!

キーワード: ヒト歯牙, エナメル質, <sup>14</sup>C, 法医学, 核実験, 年齢推定

Keywords: human tooth, enamel, <sup>14</sup>C, forensic science, nuclear bomb testing, age estimation

## 波照間島と落石岬における大気中二酸化炭素の放射性炭素同位体比観測 Observations of atmospheric radiocarbon in carbon dioxide at Hateruma Island and Cape Ochi-ishi, Japan

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Atmospheric radiocarbon in carbon dioxide ( $^{14}\text{CO}_2$ ) is a powerful tracer for understanding of carbon cycles, e.g. oceanic and biospheric  $\text{CO}_2$  exchanges and  $\text{CO}_2$  emissions from fossil fuel combustion. Observation sites for radiocarbon concentrations,  $\Delta^{14}\text{C}$ , 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  $^{14}\text{CO}_2$  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.  $\text{CO}_2$  was extracted from the whole air at NIES and  $\text{CO}_2$  samples were converted to graphite and analyzed ratios of  $^{14}\text{C}/^{12}\text{C}$  by accelerator mass spectrometry (AMS, National Electrostatics Corp., 1.5SDH) at Paleo Labo Co., Ltd., Japan. Analytical precision in  $\Delta^{14}\text{C}$  determined from statistical uncertainty (number of  $^{14}\text{C}$  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  $\Delta^{14}\text{C}$  was  $-5$  ‰  $\text{yr}^{-1}$  in average but large IAV was observed at both stations: large decreases in 2007-2008 and in 2010-2011 ( $-8$  to  $-9$  ‰  $\text{yr}^{-1}$ ) and almost zero decrease in 2009. We also observed clear seasonal cycle of  $\Delta^{14}\text{C}$ . The peak-to-peak amplitudes in the seasonal cycle determined from the smooth curve fits were 7 ‰ at both stations and the maximum of  $\Delta^{14}\text{C}$  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  $\Delta^{14}\text{C}$  seasonal cycle between HAT and COI suggested that the atmospheric  $\Delta^{14}\text{C}$  at COI was influenced by  $\text{CO}_2$  emitted from terrestrial biosphere.

## 放射性炭素同位体を用いた沖縄辺戸岬、福江島におけるPM2.5エアロゾルの起源推定 Radiocarbon based source apportioning of PM2.5 carbonaceous aerosols at Cape Hedo, Okinawa and Fukue island, Japan

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Radiocarbon (<sup>14</sup>C) 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 <sup>14</sup>C 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 <sup>14</sup>C 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 <sup>14</sup>C content represents in the unit of pMC. Results of EC-<sup>14</sup>C in both sites were 25-30pMC in Cape Hedo and 18-44pMC in Fukue, respectively. These results mean that relative apportionments 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-<sup>14</sup>C 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

## 蔵王山の火山湖と温泉における $^{129}\text{I} / ^{127}\text{I}$ を用いた火山活動モニタリングの試み Study on monitoring of volcanic activity using $^{129}\text{I} / ^{127}\text{I}$ 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<sup>[1]</sup>. Since the water quality of crater lake are correlating with volcanism changes<sup>[2][3]</sup>, 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  $^{129}\text{I} / ^{127}\text{I}$  ratios (atomic ratio of radioiodine and stable iodine) in crater lake and hot spring of Zao volcano.

Natural  $^{129}\text{I}$  (half-life: 15.7 million year) are produced by nuclear spallation reaction of  $^{129}\text{Xe}$  with cosmic ray in the atmosphere and spontaneous fission of  $^{238}\text{U}$  in the geological layer. In the ocean, steady-state  $^{129}\text{I} / ^{127}\text{I}$  ratio of the seawater is estimated to be  $1.5 \times 10^{-12}$ <sup>[4]</sup>. Sunken iodine by the ocean plate having lower  $^{129}\text{I} / ^{127}\text{I}$  ratio (older  $^{129}\text{I}$  age) compared to the steady-state ratio of seawater, are supplied to the atmosphere mainly via magmatic activity. In general,  $^{129}\text{I} / ^{127}\text{I}$  ratio in hot spring water and brine water are used as indicator of origin and behavior of iodine in the water<sup>[5][6]</sup>.  $^{129}\text{I} / ^{127}\text{I}$  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  $\mu\text{m}$  filter,  $^{129}\text{I} / ^{127}\text{I}$  ratio were measured for the isotopic diluted water samples by adding carrier ( $^{127}\text{I}$  standard) at MALT, The University of Tokyo.  $^{127}\text{I}$  concentrations were measured by ICP-MS, and original  $^{129}\text{I} / ^{127}\text{I}$  ratio of water samples were estimated.

Water temperature and pH were 10.2 °C and 3.3 at Okama; 40.0 °C and 3.3 - 4.0 at Kamoshika Hot Spring.  $^{129}\text{I} / ^{127}\text{I}$  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 ( $1.5 \times 10^{-12}$ )<sup>[4]</sup>. Since  $^{129}\text{I} / ^{127}\text{I}$  ratio of anthropogenic metric water were over  $9.0 \times 10^{-12}$ <sup>[7]</sup>, surface water of Okama and Kamoshika Hot Spring water were very likely to be strong affected by the meteoric water including anthropogenic  $^{129}\text{I}$ . For the monitoring of volcanic activity using  $^{129}\text{I} / ^{127}\text{I}$  ratio, it is necessary to decide the site as few anthropogenic  $^{129}\text{I}$  as possible through the measuring of  $^{129}\text{I} / ^{127}\text{I}$  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, crater lake, hot spring,  $^{129}\text{I} / ^{127}\text{I}$ , AMS

## 福島第一原子力発電所起源 I-129 の逐次抽出法による土壌中の分布調査 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 <sup>129</sup>I ( $T_{1/2} = 1.57 \times 10^7$  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. <sup>129</sup>I in soil was measured by AMS and stable iodine (<sup>127</sup>I) was measured by ICP-MS at MALT (Micro Analysis Laboratory, Tandem accelerator), The University of Tokyo. It was found that <sup>129</sup>I was concentrated near surface but distributed deeper compared with <sup>137</sup>Cs ( $T_{1/2} = 30$  y). From the estimation of relaxation length using depth profiles, the FDNPP derived <sup>129</sup>I move 0.6 cm/y downward and <sup>137</sup>Cs 0.3 cm/y for it. It was also found that <sup>129</sup>I seems to move downward more quickly than <sup>137</sup>Cs.

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

Recent studies on the X-ray absorption fine structure (XAFS), especially near edge structure (XANES), reported that the stable iodine (<sup>127</sup>I) in soil existed as an organic component<sup>[1]</sup>. However, it had not yet been proved that it was also the case with the accident derived <sup>129</sup>I because it had been incorporated in the soil system only recently and the abundance of <sup>129</sup>I in soil was more than 8 orders of magnitude smaller than sub-ppm level stable iodine (<sup>127</sup>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 <sup>129</sup>I can be quantified by AMS method of the fraction added with carrier. The fraction of the organic component for <sup>127</sup>I and <sup>129</sup>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 <sup>129</sup>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 ( $\text{IO}_3^-$ ) absorbed in amorphous oxides, especially goethite or delta- $\text{MnO}_2$ . They are formation of monodentate mononuclear outer-sphere species and bidentate, binuclear inner-sphere species<sup>[2]</sup>. The latter iodine are linked to organic carbon directly by a covalent bond.

[1] Y. S. Shimamoto et al., 2011, *Environ. Sci. Technol.*, **45**, pp2086-2092

[2] T. Nagata et al., 2010, *Geochim. Cosmochim. Acta.*, **74**, pp6000-6013