Japan Geoscience Union Meeting 2015
(May 24th - 28th at Makuhari, Chiba, Japan)

HTT31-01 Room:304 Time:May 27 11:00-11:15

Geochemical and isotopic map of stream water as a basis of environmental traceability an example of northeastern Japan

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Traceability, which can trace the route of a material from its utilization to production area, is a key concept based on the precautionary principle of global environmental issues. All materials in the earth is composed of 92 elements, most of which have stable isotopes (SI). The ratio of SI of an element can be utilized as a traceability index regarding the source and process of the element in the environment. In order to establish a traceability method in earth environment using multiple elements and SI, Research Institute for Humanity and Nature (RIHN) has been installed analytical instruments of elements and SI in the laboratory, and started a project of making water quality map, which aims to elucidate the spatial distribution of the concentration and SI ratios of elements in terrestrial water. This is because elements in organisms and agricultural-fishery products are derived ultimately from ambient water, and the concentrations and SI ratios of elements in the water vary geographically rather than seasonally. This geographical variation of terrestrial water is attributed to the amount and quality of atmospheric precipitation and the geology and human activities in the watershed. Accumulating the data of water quality map provides basic information on traceability studies including water-material circulation, biodiversity, and climate change as well as agricultural and fishery products and food.

In order to elucidate the impact of atmospheric precipitation and the role of chemical weathering and human activities on fresh water, we determined the concentrations of 6 major ions and 46 trace elements and the isotopic ratios of hydrogen, oxygen, and strontium for about 1000 stream waters in the Iwate, Miyagi, Akita, and Yamagata prefectures of northern Tohoku, and compared them with geomorphology, geology and the geochemical map of river sediments.

The δD and δ^{18}O values of stream water tend to decrease with latitude and elevation, but their relation is expressed as δD=6.6δ^{18}O. The slope value of 6.6 is lower than 8.0 in the precipitation and it becomes low in the water of high δD and δ^{18}O values, indicating that river water experienced with evapotranspiration, particularly in southern and coastal areas where both values are high.

The ^{87}Sr/^{86}Sr ratio of stream water in igneous rock watershed ranges from 0.704 to 0.706, which is slightly higher than that of water in the Green Tuff region (0.706-0.707). The ^{87}Sr/^{86}Sr ratios of sedimentary rocks in the north Kitakami region (0.7085-0.7124) are higher than those in the south Kitakami one. The distribution of Sr/Ca ratio of stream water in the igneous rock watershed also resembles to that of watershed rocks. This correspondence of the Sr isotopic ratio and the Sr/Ca ratio of stream water with those of watershed rock reflects the major source of Sr as well as Ca in the water is largely derived from rocks.

However, although the distribution patterns of alkali-earth elements (Sr and Ca) in the water is similar each other, it is different from those in the river sediment. The distribution pattern of other major and trace elements is also different between river water and river sediment. This inconsistent pattern of elements indicates the important role of element fractionation during the chemical weathering of rocks into water, such as the adsorption and desorption of elements on the sediment particles.

Rare-earth elements of river water in the Miyagi prefecture tends to be higher than those in the Iwate prefecture. As REE are present as colloidal form, this tendency suggests that rocks in the Green-tuff region supply colloidal particles than granite and sedimentary rock which widely distribute in Iwate prefecture. Adsorption-desorption reaction play an important role on the water quality of water.

Keywords: geochemical map, isotopic map, stream water, traceability, northeastern Japan

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HTT31-02 Room:304 Time:May 27 11:15-11:30

Sources of atmospheric lead inferred from isotope ratios of HCl-soluble Pb and the residual Sr-Nd of aerosol particles

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Lead is one of the major pollutants of atmospheric environment. Around the East Asia, China is known as the major emission source of atmospheric lead. It is obvious that the influence reaches Japan across the sea. However, it is not clear which part of China is the major pollutant source of Japanese air and if there is seasonal change of sources. In order to reveal these, we conducted Pb isotope analysis of HCl-soluble component of aerosol particles sampled with high temporal resolution from August 2011 to August 2012 at the Omura City, north Kyushu. In association with Sr-Nd isotope ratios of HCl-insoluble component and air-mass back trajectory analysis, the four regions, the north, inland, south China, and Korea, were recognized as the discrete sources of atmospheric lead. Among the four areas, the north China is important in amount of lead. Atmospheric lead in the Omura City is mainly derived from the north and inland China during fall and winter. On the other hand, it is transported from Korea during a few days in fall, and from the south China during a few days in summer.

Keywords: Aerosol, Atmospheric lead, cross-border pollution

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HTT31-03 Room:304 Time:May 27 11:30-11:45

Multi-tracers approaching to groundwater and surface water interaction in Ono basin surrounded by steep mountains, Japan

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Mountainous headwaters and lower stream alluvial plains are important as water recharge and discharge areas from the view point of groundwater flow system. Especially, groundwater and surface water interaction is one of the most important processes to understand the total groundwater flow system from the mountain to the alluvial plain.

We performed tracer approach and hydrometric investigations in a basin with an area 948 square km surrounded by steep mountains with an altitude from 250m to 2060m, collected 258 groundwater samples and 112 surface water samples along four streams flowing in the basin. Also, Stable isotopes ratios of oxygen-18 (^{18}O) and deuterium (D) and strontium (Sr) were determined on all water samples.

The ^{18}O and D show distinctive values for each sub-basin, due to different average recharge altitudes among four sub-basins. Also, Sr isotope ratio shows the same trend as ^{18}O and D, due to different geological covers in the recharge areas among four sub-basins.

The ^{18}O, D and Sr isotope values of groundwater along some rivers in the middle stream region of the basin show close values as the rivers, and suggesting that direct recharge from the river to the shallow groundwater is predominant in that region. Also, a decreasing trend of discharge rate of the stream along the flow supports this idea of the groundwater and surface water interaction in the basin.

Keywords: Groundwater flow system, Circulatory system of groundwater and surface water, Hydrogen and oxygen isotope ratio, Sr isotopic composition, Multi tracers approach

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HTT31-04 Room:304 Time:May 27 11:45-12:00

Plantation of Cryptomeria japonica might alter dynamics of metal element

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In this study, we found dynamics of many metallic elements in catchment areas may be altered by plantation of Cryptomeria japonica.

Organisms can alter nutrient dynamics in ecosystems via physiological results such as respiration, decomposition and excretory processes. Many studies have established importance of the alteration of nutrient dynamics by organisms in ecosystems. Especially, dynamics of carbon, nitrogen and phosphorus can be altered by physiological responses of organisms. However, there are few studies that focused on effects of organisms on dynamics of metallic elements in ecosystems.

Our previous studies showed that the vegetation in catchment area might alter calcium concentration in the soils and water of streams, and affect the community structure of invertebrates in soils and streams. In these studies, we observed that concentration of exchangeable calcium in the Japanese cedar (C. japonica) plantations is about three times higher than in the evergreen broad-leaved forests. This might indicate C. japonica has characteristics that alter dynamics of metallic elements in soil. We focused on organic acids extracted from roots of tree because some studies showed root exudation of organic acids could elute materials in soil particles and base-rocks.

We conducted field survey and a pot experiment in Wayakama Experimental Forest of Hokkaido University. And we determined the reason why the elevation of calcium concentration in plantation of C. japonica occurs. In consequences, our results showed plantation of C. japonica might increase exchangeable metallic ion in soils through increased supply of organic acids to soil systems. And the some eluted metallic ions might be supplied to streams.

Keywords: stream, soil, metallic ion, plantation of Cryptomeria japonica, organic acids

Japan Geoscience Union Meeting 2015
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HTT31-05 Room:304 Time:May 27 12:00-12:15

Usefulness of strontium isotope composition for determining the geographical origin of Japanese horseradish

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The geological conditions in Shizuoka prefecture vary widely, from young volcanic rock area in the eastern part to old sediment or metamorphic rock in the western area. Dissolved element and isotopic compositions in springwater, which is located at the top of a river, would reflect the geological characteristics with the exception of effects of atmospheric deposition and human activity. This study aims to evaluate the usefulness of strontium isotope ratio ($^{87}Sr/^{86}Sr$) for determining the geographical origin of Japanese horseradish (*Wasabia japonica*) cultivated in the springs. We collected 58 springwater samples and 59 horseradish samples from 21 sites in Shizuoka prefecture. These samples were subjected to trace elements and $^{87}Sr/^{86}Sr$ analyses. The $^{87}Sr/^{86}Sr$ values differed, based on the geological characteristics of their site locations, and the value of horseradish sample was well accorded with that of water sample in the same location. The combination of trace element content, e.g. vanadium, and $^{87}Sr/^{86}Sr$ value of the horseradish allowed us to distinguish the production area clearly.

Keywords: geographical origin, Japanese horseradish (Wasabia japonica), strontium isotopic composition, trace element, Shizuoka prefecture

Japan Geoscience Union Meeting 2015
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HTT31-06 Room:304 Time:May 27 12:15-12:30

A possibility of the elucidation of smelting process in pre-modern Japan by stable lead isotope ratio analysis

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Keywords: stable lead isotope ratio, pre-modern Japan, smelting process

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HTT31-07 Room:304 Time:May 27 12:30-12:45

Recent progresses in the ICP-mass spectrometry as rapid, accurate and flexible analytical tool for isotopes geosciences

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Rapid and unremitting developments in inorganic mass spectrometry, including a multiple collector-ICP-mass spectrometry (MC-ICPMS), have revolutionized the precision of the isotopic ratio measurements, and the applications of the inorganic mass spectrometry in geochemistry, metrology and biochemistry were beginning to appear over the horizon. Analytical community is actively solving problems, such as spectral interference, mass discrimination drift, high-yield chemical separation and purification processes, or reduction of the contamination of analytes. The variations in isotopic ratios of the heavy elements can provide new insights into past and present geochemical and biochemical processes.

Stable isotope tracers are now increasingly being used in studies of elemental metabolism, bioavailability or toxicity of nutrients, as well as evaluating the elemental turnover time. Besides an absence of harmful radiation, this approach has the further advantage of enabling multi-element studies, in which different isotopes can be added to the same meal. The metabolism of higher organisms can be transcribed as stable supply of the most essential elements through transfer, absorption, and storing processes, which form the basis of homeostasis function. Because of the homeostasis control, fluctuations or changes in the concentration of the essential nutrients would be highly restricted to maintain the biochemical functions. This suggests that the impairments in the metabolism or nutritional status of both the essential and toxic metal elements could not be evaluated only by the concentration of the elements in fluids. Variations in the isotopic composition of the elements induced through dietary or metabolism processes have potential to become novel biochemical markers for assessing impairments in metal metabolism or nutritional status of the elements.

Iron is one of the most important inorganic nutrients for all terrestrial plants and animals, and the natural variations in isotope ratio of Fe have been used to trace the food chain. For land organisms, it is widely recognized that the Fe isotope ratios (^{56}Fe/^{54}Fe and ^{57}Fe/^{54}Fe) changes by 0.1% with increase the trophic level (Walczyk and Blankenburg, 2002, 2005). In contrast, the Fe isotope data for marine organism of lower trophic levels (plankton, shrimp and tuna) did not vary significantly from the Fe isotope ratio for the seawater. The small variations in the Fe isotope ratios for marine organisms could be explained either by higher intake efficiency of Fe from the dietary foods, or by the smaller isotope fractionation due to intake of hemo-Fe (Fe(II)). However, it should be noted that the reported Fe isotope ratios for marine organisms were very limited, and therefore, possible link between the ^{56}Fe/^{54}Fe and ^{57}Fe/^{54}Fe ratios and the trophic level was not clearly demonstrated. To investigate this, we have measured the ^{56}Fe/^{54}Fe and ^{57}Fe/^{54}Fe for series of marine creatures of various trophic levels using multiple collector-ICP-mass spectrometer (MC-ICPMS). The measured ^{56}Fe/^{54}Fe isotopes for marine organisms of higher trophic levels became significantly lower than those for lower trophic levels animals. Several important features of the Fe isotopes for marine creatures could be derived from the present results. The obvious changes in the Fe isotope ratios could be due to different Fe biocycling for higher trophic level animals. Another important feature obtained from the Fe isotopes was that the definition of the trophic level, based on the ^{13}C/^{12}C and ^{15}N/^{14}N isotope, would not reflect the food chain for inorganic nutrients including Fe. The details of the mechanism in the variation of the ^{56}Fe/^{54}Fe ratios for both the marine and land organisms will be discussed in this presentation.

Keywords: stable isotope, isotope signature, MC-ICP mass spectrometry, Fe biocycle, trophic level

Japan Geoscience Union Meeting 2015
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HTT31-08 Room:304 Time:May 27 14:15-14:30

Use of multiple isotope tracers to study coastal ecosystem

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Environmental traceability method is based on material cycling, thereby applicable to the studies on various environmental issues. Especially, the method is useful to evaluate human impacts on ecosystem properties. However, comprehensive use of the method is under development. In this study, we show potential applicability to use multiple isotope tracers in the study of coastal marine ecosystem, using isotope elements of terrestrial origin as well as the elements circulating in the system.

Elements constituting living organisms are obtained from the environments. From ecological point of view, the living organisms are obtaining environmental information where they live and what they eat. In other words, the living organisms record indicators of surrounding environmental conditions. Elements in hard tissue, e.g. otolith, sequentially record environmental variables as they grow. The calcium carbonate and trace metals in otolith are primarily derived from the water, therefore, they contain information of the water bodies that the fish have previously occupied. In contrast, elements in soft tissue are continuously metabolized and have each turnover time. Some elements in the organism are derived from the surrounding water, others are derived from the diet. These things considered, multiple tracers in organisms have various information.

Our project, ”Development of multi-isotope tracer techniques for evaluating functions of coastal ecosystem” funded by JST-CREST, is aiming at developing a method to evaluate coastal ecosystem using multiple isotope tracers. We show potential advantage of the use of multiple isotope tracers to study coastal ecosystem.

Keywords: Stable isotope, Traceability, Coastal ecosystem

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HTT31-09 Room:304 Time:May 27 14:30-14:45

Stable nitrogen isotope ratios of amino acids reveal the differences in trophic position of benthic fishes in Sendai Bay

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Coastal area provides many ecosystem services such as fishery resources, while it is one of vulnerable ecosystems affected by overfishing, pollution, and development. For the suitable conservation and management, it is necessary to clarify the food web structure and its dynamics. In coastal ecosystem, since some fish species show a widespread migration, their spatio-temporal scales are critical for unraveling the food web dynamics.

Estimation of trophic positions using carbon and nitrogen isotope ratios of bulk tissues are powerful approach for clarifying the food web structure. In the management of fishery resources, trophic positions estimated by stable isotope ratios are regarded as important index of fish resources.

Recent studies reveal that stable nitrogen isotope ratio of individual amino acids ($\delta^{15}N_{AA}$) is useful for estimating trophic levels (TLs) of organisms. In the amino acid metabolism of organisms, glutamic acid experiences deamination and transamination, which consequences great isotopic enrichment per TL. On the other hand, phenylalanine conserves its amine during metabolism, resulting in little isotopic enrichment per TL. Therefore, the TLs of organisms can be determined by following equation:

$$TL = (\delta^{15}N_{Glu} - \delta^{15}N_{Phe} + \beta)/7.6 + 1$$

where $\delta^{15}N_{Glu}$ and $\delta^{15}N_{Phe}$ are stable nitrogen isotope ratios of glutamic acid and phenylalanine of an organism, respectively, and β is the nitrogen isotopic difference between glutamic acid and phenylalanine of a primary producer. For accurate estimation of TL, β values for aquatic and terrestrial primary producers are defined as -3.4 ‰ and +8.4 ‰, respectively.

In this study, we estimated TLs of bastard halibut (*Paralichthys olivaceus*) and stone flounder (*Kareius bicoloratus*) in Sendai Bay using $\delta^{15}N_{AA}$ values. After hatch and settling, juvenile fishes inhabit shallow sea areas (water depth $<$10m), and most of them move to deeper offshore areas with growth. However, some large adult fishes are caught at shallow sea areas.

To reveal the changes of TL with growth and differences of TL among different habitats in these fishes, we collected fish samples from various habitats in Sendai Bay from 2012 to 2014. Amino acids of fish muscle tissues were purified by HCl hydrolysis, followed by N-pivaloyl/isopropyl derivation. The values of $\delta^{15}N_{AA}$ were determined by isotope ratio mass spectrometry coupled to a gas chromatograph via combustion and reduction furnaces.

Analysis of $\delta^{15}N_{AA}$ clarified increase of TL from juveniles to larger adults of bastard halibut and stone flounder. Furthermore, difference of TL between adult fishes with even body size collected in shallow sea areas and deeper offshore areas suggested that some adult fishes stay at shallow sea areas for a long period. In offshore areas, difference of TL among habitat areas and seasons suggested the variation of food resources. Furthermore, we considered the possibility for tracing the migrations of individual fish using information from $\delta^{15}N_{AA}$.

Keywords: coastal ecosystem, food web, fishery resources, migration tracing

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HTT31-10 Room:304 Time:May 27 14:45-15:00

The effects and solution of ”mixing problem” in the dietary analysis using stable isotopes

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Dietary analysis using stable isotopes can reveal dietary information in the long timeframe, and it can be applied to ancient animals. These advantages of stable isotope analysis allowed them to be used in the various field of science including anthropology, ecology and archaeology. Especially, stable isotope analysis using dual isotopes; carbon ($d^{13}C$) and nitrogen ($d^{15}N$) is the most widely used in every field. Carbon stable isotopes possess distinctly different isotope ratios between C_3 and C_4 plants due to fractionation during photosynthetic carbon fixation. The stable nitrogen isotope ratios increase along with food chain. In this reason, stable isotope analysis using these stable isotopes provides highly reliable dietary information. In addition, recent studies sometimes use stable isotope mixing model, which can evaluated the proportional contribution of each food resource to the diets of individual or groups of target animal.

However, there are several points to be kept in mind when we interpret the result of stable isotope analysis, and the ”mixing problem” is one of them. This is attribute to the geometry of sources and mixtures in a mixing diagram, and the problem sometimes diffuses or constrains the possible source contributions. The mixing problem frequently occurs as the number of sources increases (e.g. over 4 sources in dual isotopes). If the geometry of sources and mixtures were likely to involve the mixing problem, the interpretation of stable isotope analysis should be undetermined regardless of the use of mixing model. However, this problem is not sufficiently recognized even in modern times.

In this presentation, we show the effect and solution of the mixing problem using field data of Hokkaido brown bears (*Ursus arctos*). Brown bears are opportunistic omnivore and they consume various diet items including C_3 plants, crops (including C_4 plants), terrestrial animals and salmon. Previous studies showed that the mean $d^{13}C$ and $d^{15}N$ values of C_3 plants, terrestrial animals and salmon of Hokkaido were almost linearly distributed, and the points of Hokkaido brown bears were also mostly plotted on the line. This is exactly the case of mixing problem, and we tried to evaluate the effects of the mixing problem using mixing model analysis and additional use sulfur stable isotopes which can isotopically separate the marine and terrestrial diet.

We collected bone collagen of brown bears in Shiretoko peninsula and their diet items (C_3 herbs, C_3 fruits, corn, terrestrial animals and salmon), and measured carbon, nitrogen and sulfur stable isotope ratios. Then we estimated proportional contribution of each diet items to individual bear’s diet using mixing model (SIAR) both in dual (carbon and nitrogen) and triple isotopes (carbon, nitrogen and sulfur), and compared the results of these estimates.

In the results of SIAR using dual isotopes, proportions of each diet items (C_3 herbs, C_3 fruits, corn, terrestrial animals and salmon) were 33.2%, 28.1%, 8.4%, 19.0%, 6.4%, respectively. On the other hand, dietary proportions estimated by triple stable isotopes were 36.5%, 28.3%, 6.8%, 11.4% 10.5%, respectively. Mean absolute differences of proportions of each diet items were highest in terrestrial animals (12.0%). In the dual isotope analysis, the proportion of terrestrial animals tended to be overestimated (max: 53.3%) and salmon and C_3 herbs were likely to underestimated.

Our results showed that the mixing problem practically results in the bias of dietary estimation by stable isotope mixing model. When the geometry of sources and mixtures were likely to involve the mixing problem, the proportions of diet items in the intermediate position of mixing diagram should be overestimated. In this case, interpretation of stable isotope data is difficult, and therefore, addition of other stable isotope elements or discussion about the expected bias should be needed.

Keywords: stable isotope, carbon, nitrogen, sulfur, mixing model, brown bear

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HTT31-11 Room:304 Time:May 27 15:00-15:15

Marine ecological study using long-lived radionuclides

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Recent advance in technology enables us to monitor fish migrations and marine mammals behaviors to understand their ecological information. These fundamental data is critical to establish management programs of marine resources. However the logger size limits their applicability to smaller size tests and no information is available for the period before the capture and release of loggers.

We therefore have been trying to develop methods to monitor ecology and environmental data using geochemical fingerprints remaining in the body of marine organisms. That includes radiocarbon, iodine and other trace amount of nuclides. In the presentation, we will introduce our recent studies on this topic in particular using radiocarbon that is measured by Single Stage Accelerator Mass Spectrometry installed at the Atmosphere and Ocean Research Institute, the only and the first machine in Japan. The success of the study is depending on the reduction of sample sizes for each measurements but this new AMS provides stable and reliable measurements with trace amount of samples.

Keywords: isotope, radionuclides, accelerator mass spectrometry, ecology

AMS building (left) and Single Stage Accelerator Mass Spectrometer (right)