Research strategy of multi-isotope studies on environmental research

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Environmental traceability method is based on material cycling, and thus considered to be applicable to the studies on various environmental issues. Especially, the method can be applicable to evaluate human impacts on ecosystem properties. In terrestrial ecosystems, heterogeneity of local environment in multiple elements and isotope ratios exists in geology, providing information to watershed ecosystems through water cycling, material cycling and movement of living organisms. The information can be used as an indicator of environmental traceability method.

The Research Institute for Humanity and Nature (RIHN) is operating a co-research framework, named “Isotope Environmental Research Collaboration”. The research pursues research development through multi-elemental analysis and multi-isotope analysis, considering the environmental traceability. To achieve the goal, various approaches are required including development of manuals for observation, standardization of analysis and construction of protocols for sharing data, etc.

It is recently considered that research collaboration with local people, local government, and local education is important to solve environmental issue. We propose the importance of research collaboration between universities and our role to enhance research collaboration based on RIHN initiative.

Keywords: Stable isotopes, Environmental traceability, Watershed

Estimation of air pollutant sources from the seasonal variation of Sr and Pb isotope ratios of aerosols

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[Introduction] Transboundary air pollution remains as a major environmental concern in Japan since it is located downwind of the Asian continental outflow. Due to the prevailing high pressure system in the Pacific, it has been generally accepted that the air quality near the ground surface of central Japan is relatively unaffected by the polluted air-mass originating from the Asian continent. However, our recent observations and increasing number of studies have seen episodes of high aerosol concentrations even in summer periods, indicating the significance of the polluted air-mass not only spring and winter but also in summer. However, as there are a variety of aerosol sources (e.g. refuse incineration, volcanic eruption) within Japan, it is difficult to distinguish domestic aerosols from foreign ones. In order to identify the sources of air pollutants in Japan, we determined the Sr and Pb isotopic ratios and elemental compositions of aerosols collected at Noto peninsula, Japan, in different seasons.

[Samples and analytical procedure] The sampling site is NOTO Ground-based Research Observatory (NOTOGRO) at the tip of Noto peninsula (37°45’N, 137°36’E). All of the samples were collected weekly on the roof of a building (15 m a.g.l.) using a High-volume air sampler (AH-600F, SHIBATA) with a constant flow rate of 700 L/min. Only the coarse fraction exceeding 2.5 μm in diameter was collected on a 12.6 x16.6 cm^2 SiO_2 filter through an impactor. Filters were extracted with 5% HOAc (acetic acid) solution. Residual fraction after extraction with HOAc was digested in $HF\text{-}HCl\text{-}HNO_3$ solution. Isotopic ratios of Sr and Pb were measured using a Thermo Scientific Triton thermal ionization mass spectrometer (TIMS) and Thermo Scientific Neptune multicollector-inductively coupled plasma-mass spectrometer (MC-ICP-MS), respectively.

[Results and discussion] The HOAc-leachate materials are characterized by low $^{87}Sr/^{86}Sr$ ratios and approach to a marine value of 0.70918, indicating the dominant contribution of sea salt aerosol. In contrast, the HOAc-residue materials have high $^{87}Sr/^{86}Sr$ ratios, and sample collected from 9 to 16 July show the highest $^{87}Sr/^{86}Sr$ ratios similar to surface soil silicates in the north China (0.71432 in average; Nakano et al., 2004). This result supports a view that the continental air-mass may reach to Japan even in summer season. About Pb isotope ratios of the HOAc-leachate materials, samples with low V/Mn showed similar Pb isotopic ratios reported in China. Major sources of V and Mn are from oil combustion and coal combustion, respectively. This result suggests that the air-mass over NOTOGRO in spring to early summer of 2015 are affected by the coal-derived aerosols in China.

Keywords: aerosol, Sr-Pb isotopes, transboundary air pollution

An attempt to estimate the source and accumulation of atmospheric deposition Sr and Pb isotope ratios in the bryophyte

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Recently, excessive input of heavy metals and other pollutions into the surface of environment have been developed through consumption of fossil fuels and yellow sand. Although there are many monitoring sites for atmospheric deposition in Japan, the studies that estimated the amounts of fall in forest ecosystem are very few due to difficulty of installation of sampler. In this study, we focused on relationship between atmospheric deposition and bryophyte. Since some kind of bryophyte obtains almost their nutrients from atmospheric deposition, and metallic elements are penetrated into their tissue. Therefore, we considered isotope ratios and heavy metal concentration of bryophyte tissue might provide a useful index of source and accumulation of atmospheric deposition. We collected bryophyte, soil and leaf in many part of Akita, Nagano Miyazaki prefecture, and analyzed heavy metal concentration and isotope ratios (Sr and Pb). Sr and Pb isotope ratios of the some bryophyte samples differed to the soil and leaf sample. This means bryophyte contain metals supplied from atmospheric deposition. In the future, we plant to analyze bryophyte collected another sites such as the Sea of Japan side. And because the uptake rates of heavy metal vary among bryophyte species, we have to select indicator species for estimate of atmospheric deposition.

Keywords: Fossil fuel, strontium isotope ratio , atmospheric deposition

Stream water chemistry and dynamics of sulfur derived from atmospheric deposition in a forested catchment in central Japan

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[Introduction]

Ijira catchment is located in the downwind side of the Chukyo Industrial Area in central Japan and has been historically experiencing large-scale depositions of sulfur and nitrogen from the atmosphere. The catchment was acidified and nitrogen-saturated in the mid-1990s, according to previous studies (Yamada et al. 2007; Nakahara et al. 2010). However, recovery from acidification and nitrogen saturation has been observed recently.

[Methods]

We used the following monitoring data from the Ministry of the Environment of Japan: long-term data of stream water chemistry since 1988, wet deposition (rainwater) monitoring data since 2000, the input-output budget since 2007, and isotopic data of sulfur and strontium in rainwater, soil solution and stream water since 2014. Moreover, sulfur isotopic data of tree rings in Cryptomeria Japonica from the Chukyo Area was also used for analysis of long-term trends on stream water chemistry and dynamics of sulfur derived from atmospheric deposition in the forested catchment.

[Results and discussion]

The stream water pH declined from 7.3 in 1994/1995 to 6.6 in 2003, and then promptly recovered to a value of approximately 7 thereafter. Simultaneously, the NO_3^- concentration increased until 2002/2003 and thereafter started declining in 2005. During the period of acidification with NO_3^- leaching, the SO_4^{2-} concentration reached the highest value in 1994 with a mean concentration of 210 $\mu mol_c\ L^{-1}$, and then gradually declined to 127 $\mu mol_c\ L^{-1}$ in 2013. In addition, the concentrations of dissolved organic carbon were high from the mid-1990s to the early-2000s. The mean annual SO_4^{2-} input from 2007 to 2012 was 0.9 ±0.1 $kmol_c\ ha^{-1}\ year^{-1}$, while the mean annual output from the stream for the corresponding period was 2.3 ±0.5 $kmol_c\ ha^{-1}\ year^{-1}$. Even after taking into account various uncertainties, the output of SO_4^{2-} exceeded the input. The mean sulfur isotopic ratios ($\delta^{34}S$) of SO_4^{2-} in rainwater and soil solution at 20 cm depth were 4.6‰ and 3.8‰, respectively, while that in the stream water was −13‰. Recent sulfur inputs appear to be retained in relatively shallow soil layers. The sulfur in shallow layers may have contributed to the high concentrations in the mid-1990s. Reports in the literature suggest the existence of geological sources with significantly low $\delta^{34}S$ values (from −14‰ to −8‰) near the study catchment. Therefore, it is possible that the SO_4^{2-} derived from geological sources contributes to the large discrepancy, although dendrochronology suggests certain effects of the atmospheric inputs with lower $\delta^{34}S$ (from −7‰ to +1‰) in the 1960s/1970s in the Chukyo Industrial Area.

[Acknowledgements]

This study was conducted based on the monitoring data from the Ministry of the Environment of Japan and the related research outputs. Strontium isotopic analysis was conducted by the support of Joint Research Grant for the Environmental Isotope Study of Research Institute for Humanity and Nature.

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Keywords: acidification, nitrogen saturation, sulfur, isotope

Application of Multiple-indicator to groundwater flow and chemistry study in the alluvial fan

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The Tedori River Alluvial fan between the Tedori and Sai Rivers in central Japan contains abundant groundwater resource and it supplies the region drinking and industrial water. The paddy and crop-rotated paddy fields occupy 45% of its total area (140km^2). To maintain sustainable groundwater use and prevent groundwater contamination, it is necessary to investigate the flow paths and sources of the groundwater. In this study, 83 water samples mainly from shallow groundwater, river water, and paddy water, were collected in the alluvial fan during the paddy irrigation period. We analyzed stable isotope ratios of H, O, and Sr and concentrations of major dissolved ions and trace elements.

The δ^2H and δ^{18}O values in the shallow groundwater are relatively low along the Tedori River and increased with the distance from the river; this trend would be caused by dilution effect by the river water. However, there is little contribution of the paddy water with high values reflecting the influences of the evaporation effect. Concentrations of Mg^{2+}, Ca^{2+}, Sr^{2+}, HCO_3^-, and SO_4^{2-} in the groundwater have a similar distribution trend: lower along the Tedori River and higher in the central area of the fan where paddies are relatively widespread. The $^{87}Sr/^{86}Sr$ ratios of the groundwater are related to near-surface geology. The groundwater in sediment from the Tedori River has relatively high $^{87}Sr/^{86}Sr$ ratios, whereas that from the Sai River in the north of the fan has low $^{87}Sr/^{86}Sr$ ratios. The river water of the Tedori River is also high ratio reflecting sediments in the upper river basin. The three stable isotopes indicate that the groundwater in the central and southern fans is recharged from the Tedori River, whereas recharge in the north is from the Sai River.

There are linear relationships between $^{87}Sr/^{86}Sr$ ratio and the reciprocal concentrations of Sr^{2+}, Mg^{2+}, and Ca^{2+}. These geochemical characteristics suggest that groundwater recharged from the Tedori River flows towards the central fan and then runs off the sea. It mixes with waters from precipitation and paddy water that have become enriched in these components during downward infiltration. The results obtained by application of multiple-indictor are consistent with our hydrological observation results: groundwater contour maps and river water balances, and groundwater flows reproduced by the three dimensional numerical simulation.

Keywords: Strontium isotope, Paddy field, Groundwater-river interaction

Characteristics of the distribution of $\delta^{18}O$, δD and d-excess values in groundwater and spring water at Fukushima and Niigata prefecture

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Stable isotopes of oxygen and hydrogen are useful to understanding the recharge are of groundwater and spring water. The object of this study is to grasp the detailed distribution of stable isotopes of oxygen ($\delta^{18}O$) and hydrogen (δD) in groundwater and spring water at Fukushima and Niigata prefecture.

Stable isotopes of oxygen and hydrogen are relatively high near the coastal area and relatively low at inland area, thus the inland effect is confirmed. And isotopes are relatively low in high altitude area (e.g. mountainous area), the altitude effect and temperature effect are also recognized.

From the distribution map of d-excess values, it is obvious that the d-excess is relatively low at Pacific coastal regions (Fukushima prefecture) and relatively high at coasts of Japan Sea regions (Niigata prefecture). It is assumed that the difference of d-excess values is affected by origin of the water vapor. The d-excess values change near the Ou mountains, and not change near the Abukuma mountains. Elevation of the Ou mountains is higher than Abukuma mountains, hence the water vapor is prevented by the Ou mountains is considered.

Keywords: Fukushima, Niigata, groundwater, spring water, stable isotope, d-excess

Natural abundance of ^{15}N in Japanese forest soils

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Natural abundance of ^{15}N of bulk soil N has been investigated in many studies with emphasis on soil N decomposition, N loss, and chronosequence of N dynamics in forests. $\delta^{15}N$ of bulk soil N generally increases with soil depths, which is considered as a consequence of the loss of ^{15}N-delpleted N via leaching and gaseous loss both coupled with N mineralization and nitrification as well as the incorporation of ^{15}N-enriched N into bulk soil N via microbial biomass. However, the details on how ^{15}N of bulk soil N can be discriminated during these processes are not clear.

We analyzed the $\delta^{15}N$ of bulk soil N collected in the GRENE (Green Network of Excellence) environmental information project and the ReSIN (Regional and comparative Soil Incubation study on Nitrogen dynamics in forest ecosystems) project (Urakawa et al. 2014, 2016). Mineral forest soils were collected in each watershed from five soil profiles with different soil depths down to 40 or 50cm depth. We used a modified EA-IRMS in TUAT with higher sensitivity to measure the $\delta^{15}N$ of bulk soil N with low N concentrations. We analyzed the soil samples (488 samples from 32 watersheds) with higher N concentration than ca. 0.1% (with less than 8mg sample weight to run) to reduce the risk of incomplete combustion.

$\delta^{15}N$ of bulk soil N ranged from -3.2 to +10.2 permill with N concentrations ranging from 0.1 to 1.3%. Isotopic fractionation factors for bulk soil N, estimated from the relationship between N concentrations and $\delta^{15}N$ values, ranged from 0.6 to 7.2 permill. The differences in mycorrhizal association of plants would influence the isotopic fractionation factor (Hobbie and Ouimette 2009), although we found no significant relationships between mycorrhizal associations (ECM, AM) and the isotopic fractionation factors. Climate factors such as MAT and MAP suggested as the factors affecting $\delta^{15}N$ of bulk soil N (Amundson et al. 2003) were not significantly correlated with $\delta^{15}N$ of bulk soil N. In the presentation we will discuss the possible factors influencing $\delta^{15}N$ of bulk soil N and isotopic fractionation factors in the forest watersheds.

Keywords: Stable isotopes, Forest soils, Nitrogen dynamics

Tracing Atmospherically Deposited Nitrate in Forest Ecosystem Using Triple Nitrate Isotopes

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Nitrogen saturation is one of the forest environmental issues as a result of increasing anthropogenic emission of reactive nitrogen. Forest which reached nitrogen saturation exports residual nitrogen as dissolved inorganic nitrogen (mainly nitrate: NO_3^-) and the amount of NO_3^- leaching is one of the signal of nitrogen saturation. However, the mechanism of nitrate export from forest ecosystem is not well understood because of complexity of forest internal nitrogen cycle. In fact, NO_3^- has two sources, atmospherically deposited nitrate ($NO_3^-{}_{atm}$) and microbial nitrate ($NO_3^-{}_{microb}$) in forest ecosystem. These two sources could not be separated so far, but triple nitrate isotope analysis techniques enabled distinguishing $NO_3^-{}_{atm}$ from $NO_3^-{}_{microb}$, and revealed that the fraction of $NO_3^-{}_{atm}$ (f_{atm}) in stream water is about 10% worldwide. To clarify the mechanism of export of atmospherically deposited nitrate ($NO_3^-{}_{atm}$) to stream from forest catchment, we explored which factors of forest ecosystem influence f_{atm} in stream water.

We measured $\delta^{15}N$-NO_3^-, $\delta^{18}O$-NO_3^-, $\Delta^{17}O$-NO_3^-, and NO_3^- concentration along with forest hydrological pathways, through fall, soil water, ground water and stream water at Kiryu Experimental Watersheds (KEW) in central Japan.

We calculated the value of f_{atm} of four stream water, and mean value was about 10% except for one of the stream water which flows steep slope catchment. These results are consistent with the reports for stream. In soil, the concentration of NO_3^- and the values of f_{atm} were decreased along with the soil depth suggesting that $NO_3^-{}_{atm}$ was consumed immediately at surface soil. In contrast, the values of f_{atm} were not fluctuated in ground water which suggests that the value of f_{atm} in stream water is almost determined at soil.

Keywords: forest ecosystem, nitrate isotopes, oxygen isotope anomaly

Effects of rapid environmental changes on groundwater dissolved organic carbon dynamics in Tropical peat swamp

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Tropical peatland forests in Southeast Asia are considered to be one of the most important parts of larger ecosystems due to the huge amount of carbon stock and biodiversity they contain. Yet, recent rapid and intensive deforestation to procure timber and land for commercial plants or crops (Oil palm etc.) must have induced fundamental changes in the material cycling. We focused on the effects of human impacts such as deforestation, plantation, and manmade fires on groundwater dissolved organic carbon (DOC) dynamics in the natural forest, secondary forest, and degraded peatland (shown in Photo; originally there was peat swamp forest) area in East part of Sumatra Island and Central Kalimantan, Indonesia. We measured dissolved organic and inorganic matters in peatland groundwater both in wet and dry seasons. We compared the results of both bare land site after the deforestation and the fires and oil palm site planted after the fire. Both DOC quantity and quality differences are investigated between different land use types.

Keywords: Tropical peat swamp, Southeast Asia, Land use change

Absorption of radiocesium by fungi -estimation of soil hyphal distribution using stable isotopes-

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Many studies after the Chernobyl nuclear accident in 1986 have reported that fungal fruit bodies accumulated higher ^{137}Cs concentration than other organic materials in forest ecosystem. Some of the studies pointed out soil hyphal distribution as one of the main factors determining ^{137}Cs concentration in fungi, but the viewpoint has not yet been examined well. We therefore have attempted multi stable isotopes (^{13}C, ^{15}N and ^{34}S) to examine the relationship between hyphal vertical distribution and ^{137}Cs concentration in fruit bodies.

Study site was a broad-leaved forest dominated by konara oak, mixed with fir, located at 20 km southwest from the Fukushima Daiichi Nuclear Power Plant, in Kawauchi Village, Fukushima Prefecture. Fruit bodies and soil core samples (down to 30 cm below the soil surface) were collected. After oven-dried, the fruit bodies were ground into powder, and isotope ratio (d^{13}C, d^{15}N and d^{34}S) and ^{137}Cs concentrations of samples were measured. Each soil core was separated into 2-cm long, sieved after air-dried, and isotope ratio and ^{137}Cs concentrations were measured. For ^{34}S measurement, sulfur was extracted from samples with Parr bomb and collected as precipitation of $BaSO_4$. Litter and humus layers were also collected, treated and analyzed as other samples.

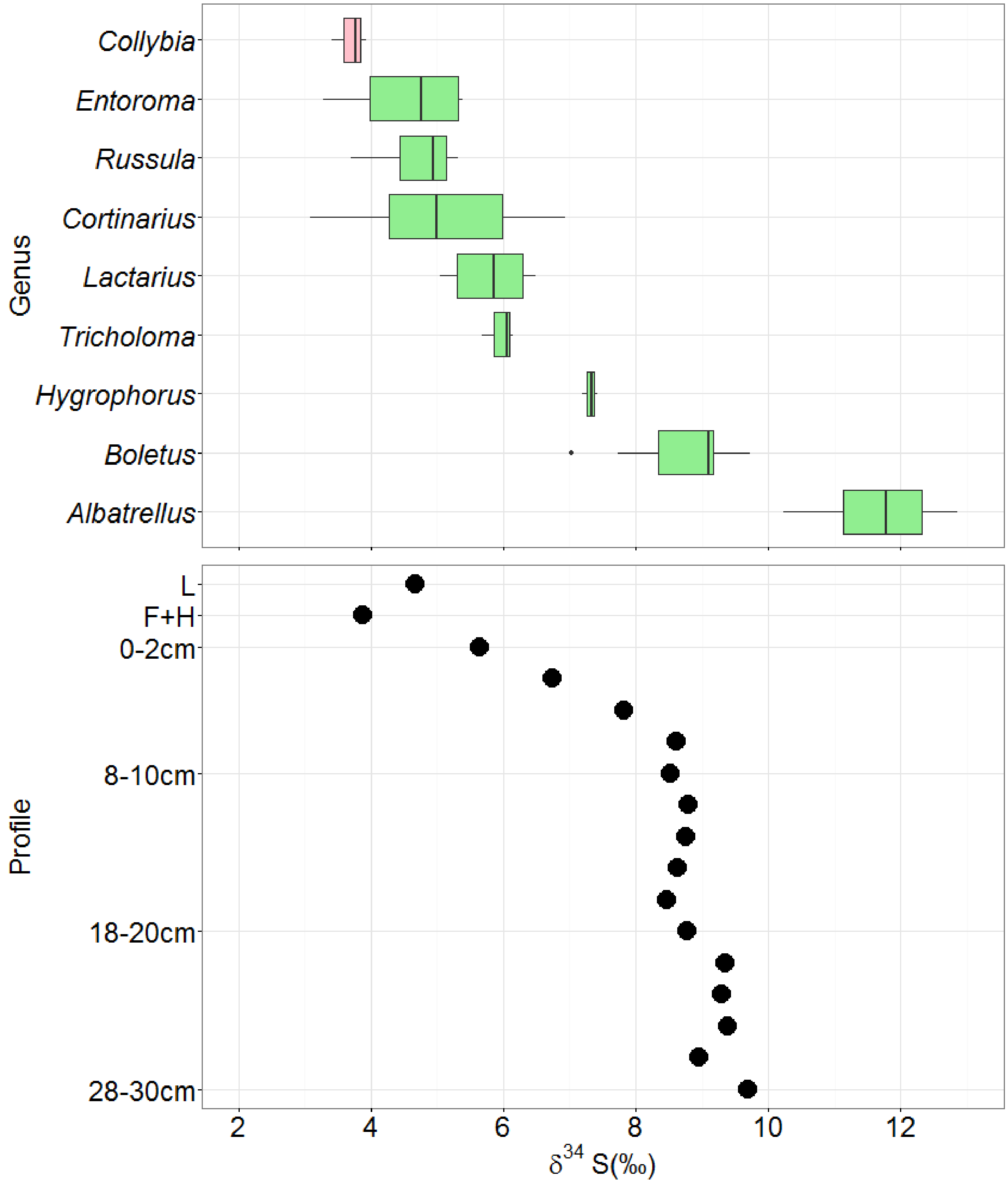
^{137}Cs concentration in saprophytic fungi was lower than that of ectomycorrhizal (ECM) fungi in average, but there was wide variation among genera and within genus in ECM fungi. Saprophytic fungi did not accumulate so much ^{137}Cs despite the high ^{137}Cs concentration in litter and humus layers. The vertical profiles of d^{13}C, d^{15}N and d^{34}S had a common trend; d values decreased with the depth. Saprophytic fungi showed most negative delta values for N and S isotopes, but most positive for C isotopes in the fungus groups. Genus-specific d values were observed for N and S isotopes, which variation was comparable to those observed for soil vertical profiles (figure).

Results of isotopes analysis suggested hyphal distributions of saprophytic and ECM fungi were completely different and that there was considerable difference in ECM fungi. Saprophytic fungi had d^{13}C and d^{15}N values close to those in organic layers after being adjusted based on the suggestions from previous studies about isotope fractionation. d^{34}S values in saprophytic fungi also were close to those in organic layers. The results of 3 isotopes indicated the hyphae of saprophytic fungi were restricted almost to soil organic layers. On the contrary, hyphal distribution of ECM had wide variations in mineral soil as indicated by genus-specific variations of d^{15}N and d^{34}S.

We did not observe significant relationships between hyphal distribution and ^{137}Cs concentration in fruit bodies. Saprophytic fungi showed lower ^{137}Cs concentration than ectomycorrhizal fungi regardless of shallow distribution of hyphae; and some genera of ECM fungi had similar values though they had different stable isotope ratios. These data are not consistent with the view that emphasized the relationship between hyphal distribution and ^{137}Cs concentration in fruit bodies. The view of soil-depth dependent ^{137}Cs accumulation by fungi needs to be re-examined.

Sulfur isotope seemed to be useful for estimating hyphal vertical distributions. Since the vertical profile of d^{34}S was similar to those of d^{13}C and d^{15}N, multi isotope approach will provide an effective tool for investigating biological processes in soil ecosystems. For further application to fungi study, isotope fractionation of sulfur and d^{34}S of available sulfur by fungi has to be studied.

Keywords: radioicesium, hyphae, stable isotope



δ^{34}S in fruit bodies and soil profile

Reconstruction of dog diet in Jomon period using carbon and nitrogen isotope analysis

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Dog is the earliest domesticated animal among other livestock. Dog’s role as companion animal and support for hunting is different from those of other livestock (e.g. goat, sheep, cattle and horse) which are mainly exploited for foods or labor as food. Because dog diet is similar to human diet in traditional subsistence, reconstructing dog diet is important to evaluate ancient human subsistence over time and space. A number of previous study show that isotope signature (carbon and nitrogen) obtained from ancient dog remains resemble to contemporary human remains.

Recently, a number of dog remain, especially for Jomon period, was excavated and identified in Japanese archipelago. However, the study of evaluating the isotope signature of Jomon dog is very little. Gakuhari et al. (2015) demonstrated the date of two buried dogs in Jomon period using directly radiocarbon dating of dog bones, so that two buried dog were dated to ca. 7300 calBP. In addition, the carbon and nitrogen isotope analysis of bone collagen indicated that the dietary characteristic of two buried dogs is similar to those of human remains from same site.

Here, we report new data of radiocarbon dating and stable isotope analysis from other dog bones in Kamikuroiwa rockshelter site and Higashimyo shellmound. We discuss the dietary characteristic of Jomon dog in Western Japan.

As results, other dogs in Kamikuroiwa rockshelter and 11 dogs in Higashimyo shellmoud showed very close ages to two buried dogs in Kamikuroiwa rockshelter, and assigned to the last Initial Jomon and the initial Early Jomon period. Although these results were not consisted with the previous archaeological interpretation, they are important because these dogs were one of the oldest evidence of certain Canis domestication in East Asia. In addition, based on carbon and nitrogen isotope analysis, we found that Kamikuroiwa and Higashimyo dogs could be discriminated to three populations with different isotopic patterns. This new finding is important to reconstruct ancient feeding culture of dog and human subsistence for Jomon period.

Keywords: Jomon dog, nitorgen

Food web analysis using stable carbon and nitrogen isotopic ratios: from the shallow to deep water in Toyama Bay

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The isotopic ratios of stable carbon and nitrogen are useful to clarify the influence that the environmental changes (e.g. climate changes) on marine food web. The main aim of this study was to explore the food web in Toyama Bay, which has different water mass structure in the water shallower than approximately 200m and the water deeper than that. The aquatic sample of fishes and zooplankton inhabiting in the water shallower than 200m and in the water deeper than 200m, sinking particles, sedimentary organic matter and POM (particulate organic matter) were collected in Toyama Bay. We also collected sample of zooplankton and POM in Yamato basin, Central Japan Sea, and analyzed the stable carbon and nitrogen isotopic ratios of these sample. From the analysis results, we reached following findings. Aquatic creatures of both the shallow water and the deep water in Toyama Bay are located on a food web starting from POM and it is considered that phytoplankton produced near the sea surface is the primary producer. The δ^{13}C values of zooplankton collected in Toyama Bay were higher than those from Yamato basin. It is inferred that the growth rate of the phytoplankton in Toyama Bay is faster. The δ^{15}N values of the fish inhabiting in the deep water are higher than the fish inhabiting in the shallow water. In the deep water, there were scavengers and zooplankton whose δ^{15}N values are higher than those in the shallow water suggesting that they raised the trophic level. Furthermore, the δ^{15}N level of POM collected in Toyama Bay and Yamato basin were low with the average of 3.3 ‰, it is suggested that the δ^{15}N value of primary producers is low.

Keywords: Stable isotope, carbon and nitrogen, Food web

Geochemical diagnosis of groundwater flow system in the Ohno basin, Fukui prefecture

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Shallow groundwater system is developed in the city area of the northwestern Ohno basin, Fukui prefecture. This groundwater, termed as city-GW, has been used for daily lives and industries, but the recharge area and flow system of city-GW remain unclear. For the sustainable use of city-GW, we determined multiple components for the surface water and well water collected by elementary pupils as environmental education.

The oxygen and hydrogen isotope ratios of the city-GW decrease from -8.1 ‰and -48 ‰to -9.1 ‰and -57 ‰toward the west. The city-GW is divided by an interval of 0.1 ‰in the d^{18}O value and 0.8 ‰in the d^{2}H value. As the groundwater level decreases from south to west, it is likely that each isotope envelope corresponds to the flow channel. This view is consistent with the spatial distribution of several dissolved ions such as Ca^{2+}; the high concentration of Ca^{2+} corresponds to the groundwater with a uniform d^{18}O-d^{2}H value (-8.9 - -8.8 ‰and -52 - -51.2 ‰). It is likely that the city-GW is recharged mainly from the southern alluvial fan where agriculture is active. This is consistent with the high concentrations of As and Mo in southeastern city-GW, suggesting the contribution of fertilizer.

The western city-GW is enriched in NO_3, indicating the source of N to be human activities such as sewerage. Several components (K, SO4) in the western city-GW is further subdivided into the north and the south by Mt. Kameyama, which is composed of granite. This result suggests that the geochemical nature of aquifer sediments in the north are different from that in the south by this mountain. This view is compatible with the analysis for the fluctuation record of water level at 14 monitoring sites in the city-GW area. We consider that Mt. Kamayama plays a role as a barrier for the southerly flow of the city-GW. Thus, the water quality mapping is effective for elucidating the flow system of groundwater.

Keywords: groundwater, Ohno basin, flow channel, stable isotope

Influence of volcanic ash in soils on Ca cycling in forest ecosystems - Using Sr isotopes to determine the contribution of volcanic ash to Sr and Ca in stream waters and plants in a granite watershed -

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Ca in soil serves as a neutralizer of acids and is an essential nutrient for plants. The supply of Ca from weathering of rocks and its subsequent cycling is a key factor that controls the tolerance of forest ecosystems to acid deposition. In studies of the Ca cycle, stable isotopes of Sr have been used to trace Ca, because the chemical and biological behaviors of Sr resemble those of Ca. The Sr isotope method has been successfully applied, not only to investigate Ca cycling in acid-impacted ecosystems, but also to evaluate soil development processes and water-rock interactions in groundwaters and surface waters.

Volcanic ash, composed of small grains and with a large specific surface area, is present as a tephra layer in soils in Japan; this ash releases base cations rapidly owing to relatively quick weathering. However, it is difficult to estimate the distribution of volcanic ash on steep slopes, because it blends into soil and cannot be visually identified easily. Sr isotopes may be used to detect volcanic ash blended with soil and volcanic ash-derived Sr in plants and stream waters, because the stable Sr isotope ratio of volcanic ash is, in most cases, different from the ratio of the parent materials.

The aim of this study was to examine the role of volcanic ash in the Ca cycle in Japanese forest ecosystems. Soils, plants, and stream waters from a ridge, slope, and a valley were collected in a granite watershed on Mt. Tsukuba, central Japan. Rocks and atmospheric precipitation were also collected to represent Ca sources in the watershed. The $^{87}Sr/^{86}Sr$ ratios and element compositions of samples were determined to support identification of the origins of Sr and Ca in plants and stream waters.

The $^{87}Sr/^{86}Sr$ ratios were determined using a thermal ionization mass spectrometer (TIMS) (Triton; Thermo Fisher Scientific) at the Research Institute for Humanity and Nature. Acid digests of soils and plants, and filtrates of stream waters and bulk precipitation, were transferred to Teflon vials, heated until dry, dissolved in 2 M HCl, purified by cation exchange (Na et al. 1995), and subjected to TIMS analysis.

The $^{87}Sr/^{86}Sr$ ratios of soils on steep slopes and the valley floor ranged from 0.7110 to 0.7139, which indicates that the major source of Sr was the granite substrate ($^{87}Sr/^{86}Sr$ ratio 0.7120–0.7131). Soils on a ridge had lower $^{87}Sr/^{86}Sr$ ratios (0.7068–0.7072), indicating that they were mainly composed of volcanic ash materials released around 30,000 years ago from Mt. Akagi (0.7069). The $^{87}Sr/^{86}Sr$ ratio decreased and the concentrations of Sr (and Ca) increased in stream water with increased elevation from the valley bottom to the ridge, indicating that volcanic ash was the dominant source of both cations in the upstream area. The contributions of volcanic ash-derived Sr to upstream and downstream water were 50 % and 0–1 %, respectively. The $^{87}Sr/^{86}Sr$ ratios of plants were between those of the soils in which the plants were growing and those of atmospheric precipitation (0.7100). More than 74 % of the Sr in plants on the ridge, but less than 17 % in plants in the valley bottom, was volcanic ash-derived. The origin of Sr (and thus Ca) in stream waters and plants varied depending on the volcanic ash content in soils, which was significantly influenced by the site elevation. These results confirm that information about the Sr isotopic composition is useful for determining the sources and contributions of Sr and Ca in stream waters and plants, even in complex systems containing volcanic ash and bedrock weathered products.

Keywords: Ca sources, Sr isotopes, Atmospheric precipitation, Soil, Plants, Stream water

Provenance analysis of aluminosilicate detritus in marine environment by Sr-Nd isotopes: terrigenous input from the Amur River to the Sea of Okhotsk and the western subarctic Pacific

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Provenance analysis of terrigenous material in marine sediment and suspended matter provides information about a paleo- and modern environment such as atmospheric and ocean circulations and climate change on continent. It also figures out a relationship between terrigenous input and biological productivity in ocean. Radiogenic isotopes such as strontium ($^{87}Sr/^{86}Sr$) and neodymium ($^{143}Nd/^{144}Nd$) in detrital (aluminosilicate) fraction of marine sediments and suspended particles are valuable indicators for identifying the geographical provenance of the terrigenous material (e.g. Dasch, 1969; Grousset et al., 1988; Asahara et al., 1999; Asahara et al., 2012). Because the isotope ratios in detritus, especially Nd isotope, are not significantly altered by the process of weathering, transportation and deposition.

Primary productivity is high in the Sea of Okhotsk (OS), most likely because of terrigenous input from the Amur River that includes dissolved matter and suspended matter. And the western subarctic Pacific (WSP) is one of High Nutrient Low Chlorophyll (HNLC) regions, and the most important source of iron in the WSP has been thought to be terrigenous matter from the Amur River together with the Asian dust (e.g. Nishioka et al., 2007). To reveal the transport and deposition processes of the fluvial materials in the OS, the Sr and Nd isotopes of the detritus in the surface sediments and suspended matter were investigated.

The regional variations of the isotopes indicate that the detritus has three main sources: Amur River detritus, with a high $^{87}Sr/^{86}Sr$ ratio (0.711–0.715) and relatively low ε_{Nd} value (–8 to –7); volcanic detritus derived from the Okhotsk-Chukotka volcanic belt to the north of the OS, with a low $^{87}Sr/^{86}Sr$ ratio (0.703) and high ε_{Nd} value (+7 to +8); and detritus from the sedimentary rocks north of Sakhalin Island, with a high $^{87}Sr/^{86}Sr$ ratio (0.709–0.710) and low ε_{Nd} value (–10 to –9). The results indicate that the Amur River detritus is dispersed across the northwestern continental shelf and further transported southward along the east coast of Sakhalin while flowing out of the shelf. The Amur River detritus flowing out of the shelf is mixed with the Okhotsk-Chukotoka volcanic material transported from the northern area of the OS by the Okhotsk Sea intermediate water (OSIW). The transport processes of the Amur River detritus in this area are supported by previous studies of turbidity that indicated that the dense shelf water (DSW) entrains resuspended sedimentary particles on the northwestern continental shelf and transports them to the OSIW. It is possible that the OSIW entrains the Amur River material, which then circulates in the OS and partly flows out to the WSP.

[Reference]

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Keywords: strontium isotope, neodymium isotope, marine sediment, the Sea of Okhotsk, Amur River

Spatial variation of neodymium and strontium isotope ratios of shellfish soft bodies in the coastal sea of eastern Tohoku District

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Sr isotope ratio ($^{87}Sr/^{86}Sr$) has been used as a geographical index of water and vegetation and a tracer of animal migration in terrestrial ecosystem. However, it has little power in marine ecosystem because of extremely homogeneous $^{87}Sr/^{86}Sr$ ratio in seawater. In contrast, the Nd isotope ratio ($^{143}Nd/^{144}Nd$) has a potential to become effective tracer of marine organisms because the $^{143}Nd/^{144}Nd$ ratio of seawater is known to show a variation in the area and depth in the ocean (Amakawa et al., 2004). Nonetheless, there is little report on the Nd isotope ratio for marine and even terrestrial organisms, largely due to the extremely low content of Nd in organisms, and the resultant poor awareness of the element in ecological fields.
In order to explore the potential of Nd isotope as a biogeographical tracer in marine ecosystem, we determined the isotope ratios of Nd and Sr in soft bodies of shellfishes, mainly oysters and mussels in the coastal sea of eastern Tohoku district. Our results show that the $^{143}Nd/^{144}Nd$ ratio of the shellfish has a wide variation (0.5123-0.5128), whereas the $^{87}Sr/^{86}Sr$ ratio lies in the narrow range (0.70912-0.70921), which is almost equivalent to the value of seawater. Although the shellfish has lower $^{143}Nd/^{144}Nd$ ratio than the exchangeable fraction of river sediments in the watershed of the coast, both ratios show a strong positive relationship ($r^2 = 0.75$). Further, the shellfishes have higher $^{143}Nd/^{144}Nd$ ratios than seawater in the northern Pacific, and the exchangeable $^{143}Nd/^{144}Nd$ ratio of river sediments has a weak negative relationship with the $^{87}Sr/^{86}Sr$ ratio of the associated river water ($r^2 = 0.37$). These results demonstrate that the shellfish contains Nd derived from the watershed rocks of the adjoining river and Nd from oceanic water. It is concluded that the Nd isotope ratio has promise as a geographical index of habitat and foraging site of marine organisms and an effective tracer of marine animal migration at least in coastal sea zones.

Keywords: Nd isotope ratio of organisms, soft bodies of shellfish, marine ecosystem

Geochemistry of Mesopotamian clay tablets and strontium cycle in West Asia

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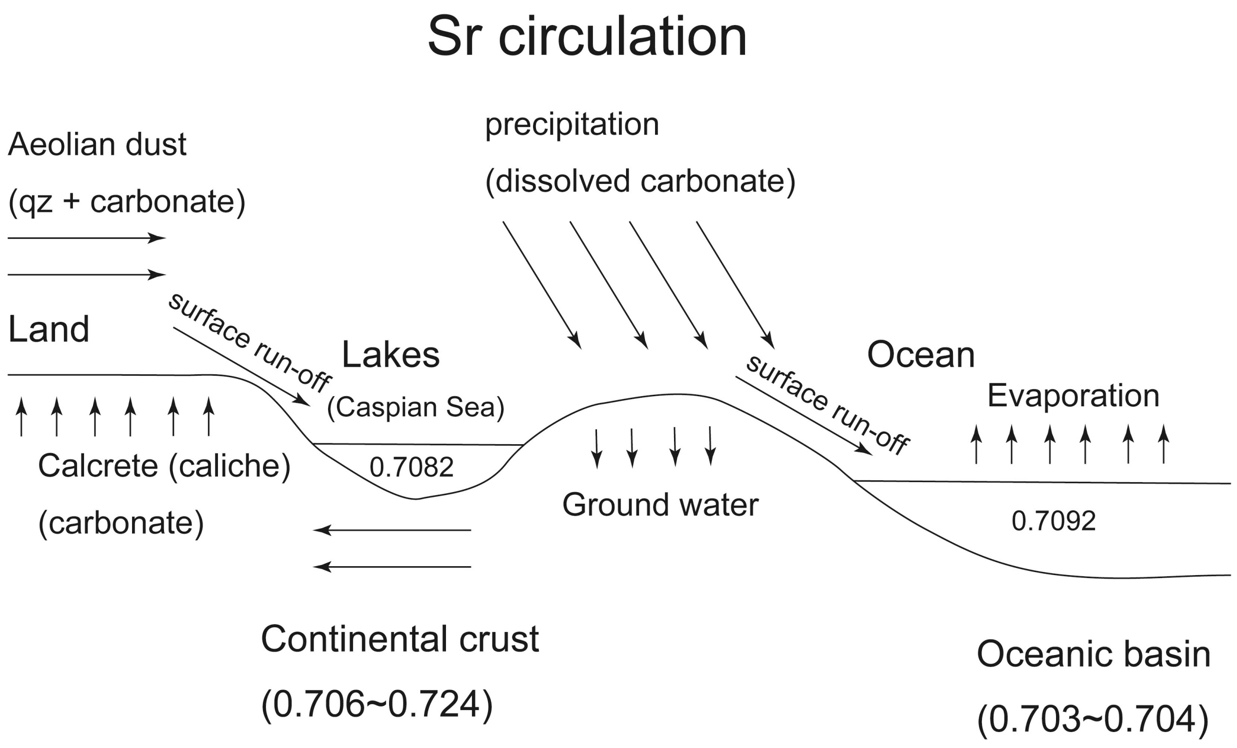
In this study, we present bulk chemical compositions and Sr-Nd isotopic ratios of cuneiform clay tablets ubiquitously excavated from Mesopotamia aiming to pin the place of origin by comparing characteristic physical feature of tablet clay and sediments from Meosopotamia. The obtained data were further compared with the compositions of meteoric water and river water in West Asia to discuss circulation of strontium and other elements, that involves interaction between rock/sediments, water and atmosphere.

Bulk chemical analyses on tablet clay samples revealed that they contain ~1% Na_2O, whereas CaO contents reached up to ~10%, implying that secondary Ca-carbonates were accumulated in pore spaces in original sediments during diagenesis or during preservation of the clay tablets. Ca-carbonate fills the pore-space under the optical microscope and was also detected by XRD analyses. The Ca-carbonate may have added artificially as a cementing material to achieve and maintain appropriate material properties for writing. TiO_2 (0.6%), Ni and Cr contents (> 100 ppm) are high as a felsic magmatic rock and imply some influence of mafic rocks, such as ophiolites. Samples from Nuzi (upstream of Tigris) has higher Ti/Fe ratio compared to Euphrates samples.

All the clay tablet samples show a weak Eu-anomaly and LREE enriched pattern, but they have different REE concentrations. Bulk Sr-Nd isotopic ratios were plot in a narrow range: 0.70835 to 0.70895 for ^{87}Sr/^{86}Sr isotopic ratio, 0.51237 to 0.51243 for ^{143}Nd/^{144}Nd. To evaluate the influence of Ca-carbonates in the tablet, we leached clay samples using acetic acid. Sr isotopic ratios of the leachate were approximately 0.7080 and plot in much narrower range. The obtained Sr isotopic ratio is similar to that of average Turkish meteoric water and Caspian Sea water (0.7082), but significantly lower than marine Sr isotopic ratios (0.7092). Iranian meteoric water has slightly higher value than Turkish meteoric water, but still lower than the marine Sr isotopic ratio. Sr isotopic ratio in the meteoric water is generally controlled by composition of aerosol (loess), especially soluble material such as carbonates. Our results indicate that Ca-carbonate cements (caliche or calcrete) formed at the surface condition have a great influence on the strontium circulation in the atmosphere of West Asia.

Keywords: Mesopotamia, Strontium cycle, clay tablet, cement, meteoric water, sediments


Sr circulation
Aeolian dust
(qz + carbonate)
precipitation
(dissolved carbonate)
Land
surface run-off
Lakes
(Caspian Sea)
0.7082
Calcrete (caliche)
(carbonate)
Ground water
Ocean
surface run-off
Evaporation
0.7092
Continental crust
(0.706~0.724)
Oceanic basin
(0.703~0.704)

Usefulness of strontium isotope composition for determining the geographical origin of Japanese horseradish (*Wasabia japonica*) (II)

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Originally, the use of strontium isotope ratios ($^{87}Sr/^{86}Sr$) was restricted to geology and petrology, but they have been used as a particularly effective tracer of geographical food origin, as plants reflect the strontium isotope composition of the regionally based rock, soil, and water.
Japanese horseradish (*Wasabia japonica*) is cultivated in springs and streams located at the top of rivers, where dissolved element and isotopic compositions would reflect the geological characteristics with the exception of effects of atmospheric deposition and human activity. This study aims to evaluate the usefulness of $^{87}Sr/^{86}Sr$ for determining the geographical origin of Japanese horseradish.
We collected 97 horseradish samples and 95 water samples from 34 locations of major production areas in Japan: Shizuoka, Iwate, Nagano, Tokyo, and Shimane. 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 horseradishes collected from Izu and Fuji regions in Shizuoka, where young volcanic rocks such as Quaternary basalts are distributed, had low $^{87}Sr/^{86}Sr$ (below 0.7040), whereas those from Nagano and Tokyo, where older rocks such as Mesozoic granites and accretionary complex are distributed, showed higher ratios (over 0.7095). We conclude that $^{87}Sr/^{86}Sr$ value of the horseradish allowed us to distinguish the production area clearly.

Keywords: discrimination of the geographic origin, Japanese horseradish (Wasabia japonica), trace element, strontium isotopic ratio, Shizuoka Prefecture

Geochemical and isotopic analyses of river waters from the Okayama Prefecture, Japan.

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We have undertaken detailed geochemical and isotopic analyses of river waters from the Asahi, Yoshii, and Takahashi River systems of the Okayama Prefecture, Japan. A total of 537 samples were collected from 381 locations in Okayama and neighboring prefectures. In some locations, samples were collected periodically over a period of 4~5 years to monitor the long-term fluctuation of the geochemical signatures. All samples were filtered prior to the analyses of the major dissolved components, trace element concentrations, and O-H isotopes. For some samples, S and Sr isotopes were also determined.

Using the data obtained in this study, we constructed high-resolution geochemical maps of the rivers, which were then used to explore the geochemical and isotopic nature of the river systems. The results show that O-H isotopes display a regional variation that exceeds the seasonal variation. The deuterium excess is high in the upstream of the river systems near the Chugoku-mountains, and gradually decreases towards the downstream.

The concentrations of the major dissolved components and trace elements also display a regional variation. For example, the Ca concentration is highest in the areas where limestones are exposed, indicating that breakdown of $CaCO_3$ has a significant impact on the Ca concentration of the river waters. The Sr concentration follows a similar pattern. The Sr isotopic signature is low in the area where the Quaternary volcanic rocks are exposed, and high in the areas where the Carboniferous to Jurassic sedimentary rocks are exposed. Such a geographical correlation between the Sr isotopic signature and rocks exposed in the area imply that the breakdown of Ca, Sr bearing minerals has an important impact on the geochemistry of the river waters.

The concentration of SO_4 is lowest in the northern region of the Okayama prefecture near the Chugoku-mountains, but gradually increases in the areas south of Tsuyama and Kuse basins. The increase in SO_4 is accompanied by shift in sulfur isotopic composition towards ~0‰. This correlation may imply that decomposition of fertilizers used in agricultural activities is, at least in part, responsible for the change in the SO_4 concentration.

Keywords: Okayama Prefecture, River Water, Geochemistry

The geochemical and stable isotope characteristics of the Chikusa River in Hyogo Prefecture

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The simultaneous survey of water temperature in the Chikusa River was performed by Chikusa River Conservation Committee on August 9, 2015. The activity of the simultaneous survey has been putting it into effect by cooperation with a hundred of areal residents on Sunday of the beginning of August every year since 2002. In the survey, water samples were collected at 74 sites in total from Chikusa-cho Mimuro in the upstream and Amako seller to the Ako City coastal part in downstream. The cooperative geochemical study for the Chikusa River between the Chikusa River Conservation Committee and Research Institute for Humanity and Nature with Kobe University and Museum of Nature and Human Activities, Hyogo has started from the 2015's survey. In this study, we analyzed the water samples collected in 2015 by the Chikusa River Conservation Committee and areal residents in order to construct an environmental map for conservation in the Chikusa River area. The hydrogen and oxygen isotope composition and an ion chromatography analysis were performed about the water samples.

Monthly monitoring of multiple water-quality to diagnose the linkage between surface water and groundwater in the Saijo plain, Ehime prefecture

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Coastal plain of Saijo city in Ehime prefecture is divided into eastern Saijo plain and western Shuso plain, and is known to be rich in groundwater irrespective of low precipitation in the Setouchi district. This is ascribed to be the presence of Ishizuchi mountainous area in the back of the plain, where the amount of precipitation increases with elevation. For the sustainable use of the groundwater, we collected samples of groundwater and river water over the whole plain area in cooperation with residents of Saijo city. Spatial analysis for the concentrations of 54 elements and stable isotope ratios of H, O, S, and Sr of these water showed that (1) the groundwater is divided into several areas dependent on the watershed and flow areas, and (2) the unconfined groundwater in the eastern plain flows in shallow aquifer recharged from the head of alluvial fan of the Kamo river, whereas the artesian water, which is representative and good quality of Saijo citizens, flows in deeper aquifer recharged from more mountainous area. Based on this finding, we started to monitor water-quality monthly at two Kamo river sites (Funagata bridge and Isonohashi bridge), Ohmachi elementary school site for shallow groundwater, Uchinuki hiroba site for artesian groundwater, and Tokumasu house site for a mixture of both waters. Temporal analysis of water quality data obtained during 2007 to 2015 is summarized as follows:
(1) The concentrations of most elements (Sb, As, V, Rb, Si, Al, Ti) in river water become high in summer and low in winter or spring, whereas those in groundwater at the Ohmachi site are high in winter and low in spring or summer and those in groundwater at the Tokumasu site are high in autumn and low in winter or spring. From the temporal analysis of these water-quality data, we can estimate the flow rate of the shallow groundwater to be about 10 cm per day.
(2) This seasonal variation is not so distinct for most elements in the artesian water at the Uchinuki site; the concentrations of Cl, B, Na, Sb, and As increase, whereas those of Ca and Sr decrease monotonously with weak seasonal change. Other elements (Si, V, Mg, Ti, SO4) show more complex variation. This result indicates that the recharge area and flow rate of the artesian water differ significantly from that of shallow one, and other water source in addition to the Kamo river is involved.
It is concluded that the monthly monitoring of water quality is effective to evaluate the linkage of the surface water and groundwater system, but further continuous monitoring is required especially for elucidating the flow system of artesian water.

Keywords: groundwater, surface water, monitoring, flow rate, water quality diagnosis, recharging area

Spatiotemporal variations in oxygen-hydrogen and strontium isotopic and trace elemental compositions of precipitation, Saijo, Ehime

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We collected monthly precipitation at 6 sites with different altitudes in Saijo city of Ehime prefecture over 9 years from 2007 to 2015, and determined their $\delta^{18}O$, δD and Sr isotopic and elemental compositions and to elucidate the precipitation process.
The values of deuterium excess (d-excess) ranged from 20 to 30‰ during winter and from 3 to 10‰ during summer in precipitations. The values of d-excess at the location “Jojusha (1,280 m above sea level)” were 3~10‰ higher than those of another sites during summer from 2009 to 2012. On the other hand, the values of d-excess at Jojusha had same as those of another sites during summer from 2013 to 2014. These results indicate heterogeneity of the isotope ratio within the investigation area and were ascribed to the origins of clouds, process of condensation from clouds and massive heavy rain.
The enrichment of As, Cu, Pb, Sb and Zn concentrations in the precipitation at lowest altitude site (20 m on the roof of city office) suggests the input of industrial anthropogenic activities from the urban area in the Saijo city and/or surrounding Setouchi region.
The concentrations of Al, Mn, Fe, Zn, V, Cu, As, Rb, Sr, Mo, Cd, Sb, Cs and Ba were high at city office site, Fujinoishi (700 m) and Jojusha from winter to spring. The Sr isotope ratio of precipitation at Jojusha also showed a seasonal variation with high in spring, indicating the contribution of the eolian dust from the Asian continent.

Keywords: precipitation, oxygen-hydrogen isotpe, Sr isotope, trace element

Seasonal and Secular Change in Sulfur, Hydrogen, Oxygen and Strontium Isotopic Ratios of Precipitation Across Chugoku District.

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We have sampled precipitations at 7 sampling points in Tottori and Okayama prefectures. Sampling method is bulk sampling by which both wet- and dry-deposition are sampled together. We measured major chemical composition, sulfur isotopic ratio of sulfate ion, hydrogen, oxygen and strontium isotopic ratios of water, and considered the origin and seasonal and secular change of precipitation and its dissolved components from 2011 to 2015.

Sulfur isotopic ratio of non-sea-salt sulfate ion decreases with increase in the distance from the Sea of Japan in most seasons. It shows seasonal change, high in winter and low in summer. Therefore, cross-border pollution from mainland China seems to affect strongly at the Sea of Japan side than at the Seto Inland Sea side, and affect stronger at winter season. The maximum and minimum sulfur isotopic ratio of non-sea-salt sulfate ion seems to increase gradually from 2011 to 2015. It is necessary to monitor sulfur isotopic ratio of non-sea-salt sulfate ion continuously in order to find whether this increase is due to the increase of cross-border pollution or not.

The d-index of precipitation calculated from its hydrogen and oxygen isotopic ratio shows seasonal variation, high in winter and low in summer, at every sampling point. Furthermore, d-index of the Sea of Japan Sea side precipitation is lower than that at Seto Inland Sea side precipitation in summer season, and the former is higher than the latter in winter season. It indicates that winter precipitation is caused by the air mass coming from the mainland China and that summer precipitation is caused by the air mass coming from the Pacific Ocean. It supports that the cross-border pollution indicated by high sulfur isotopic ratio of non-sea-salt sulfate ion is brought by the air mass coming from mainland China.

Non-sea-salt Sr isotopic ratios in Yurihama and Misasa, both close to the Sea of Japan, show seasonal variation with higher ratio (>0.7010) at spring and lower ratio (~0.7070) at summer and autumn. The higher ratio at spring reflect the contribution of yellow sand from the mainland China. On the other hand, low isotopic ratio at summer and autumn seems to be affected by the low isotopic ratio material derived from local rock, which accords with the low material transfer from the mainland China indicated by sulfur isotopic ratio of nss-sulfate and d-index of precipitation. The moderate Sr isotopic ratio at winter seems to result from the small yellow sand transport during winter season.

Keywords: precipitation, Chugoku district, non-sea-salt sulfate, S isotope ratio, H and O isotope ratio, Sr isotope ratio

Characteristics of River Water Quality in Shodo-shima Island –Drawing a material Map in Rivers -

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[Introduction]

In Kagawa Prefecture, where water resources are low, the accumulation of organic matter in the rivers downstream has become a problem (Yamada et al. 2015). For the cause analysis of organic contaminant, it is necessary to clarify its origin and load process and is essential to make a material map as the database of substance concentration.

So far, we have reported a material map in rivers in Shikoku Island. This time, by drawing a material map of Kagawa Prefecture, including Shodo-shima Island, the relationship between the river water quality and the river’s basin environment will be analyzed, especially regarding Shodo-shima Island.

[Methods]

In 27 rivers and 125 points of the entire area of Shikoku Island (Kagawa Prefecture) and Shodo-shima Island, water sampling was performed in August 2011. The items analyzed were main dissolved ions, chlorophyll *a* (Chla + Pheo), particulate organic carbon (POC) and particulate organic nitrogen (PON), and dissolved silicate (D-Si).

[Results and Discussion]

In Shikoku Island, POC was from 26 mg L^{-1} to 3232 mg L^{-1}. As POC and Chla have a correlation (r^2= 0.54, $p < 0.001$), it can be said that the main components of POC in the rivers are algae. Additionally, when multiple regression analysis was performed by setting the population density, domestic animal wastes, the density of reservoir and the amount of precipitation as explanatory variables, as the effects of the density of reservoir were shown to be especially significant (standard partial regression coefficient 3.54, $p = 0.001$), it can be considered that the algae produced in the reservoirs are the origin of POC in the rivers.

In Shodo-shima Island, POC was from 90 mg L^{-1} to 5277 mg L^{-1}, which was higher compared to Shikoku Island, especially in the southern part. Furthermore, as POC and Chla have no correlation and C/N was as high as 14 on average, it can be said that higher forms of life contribute greatly to the organic matter within the rivers of the southern part of Shodo-shima Island. When comparing the ratio of forest in the basin of the river with C/N of the downstream end, both the ratio of forest and C/N ratio were high in most of the points in the southern part of Shodo-shima Island. Accordingly, it seems that the ratio of forest has a great influence on the composition of POC. Additionally, examining the relationship between the quantity of river water and POC, POC became high at the points when the quantity of the river water was small.

The D-Si of Shodo-shima Island became higher compared to that of Shikoku Island (Shikoku Island average: 7.53mg L^{-1}, Shodo-shima Island average: 13.6 mg L^{-1}). In particular, it tended to be high at the points of granites, while it became low at the points where there is basalt in the upstream that contains Hyp-Aug-Ol basalt containing a small amount of silicon. When comparing the D-Si of the source of the rivers in Shikoku Island and Shodo-shima Island, which include granite with the amount of precipitation of the basin, it was found that there was a negative correlation. It was found that, as Shodo-shima Island has a small amount of precipitation, D-Si become high. Furthermore, by setting up the prediction formula for precipitation at the granite area points in Kagawa Prefecture, the D-Si of the other nature of the soil of Shodo-shima Island was corrected by the amount of precipitation. As a result, it was found that D-Si was higher at the points of andesite compared to the points of granite. It is considered that much silicate has been eluted because the geological age of andesite of Shodo-shima Island is new.

[Acknowledgments]
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Keywords: Shodo-shima Island, River, Water Quality

Temporal and spatial variation of anthropogenic sulfur deposition in Japan by using sulfur isotopic ratio

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[Introduction]
It is well recognized that non-sea salt (nss) sulfur isotopic ratios are useful to identify the sources because the sulfur isotopic ratio have source specific values. We have measured sulfur isotopic ratio in precipitation by using Japanese monitoring sites for the Acid Deposition Monitoring Network in East Asia (EANET). In this study, we investigated the spatial and temporal distribution of anthropogenic sulfur deposition and evaluated the contribution from transboundary transport from the Asian continent to Japan,

[Observation and analysis]
We collected precipitation samples at 12 sites (Rishiri, Tappi, Ochiishi, Sadoseki-misaki, Niigata-Maki, Niigata-Kajikawa, Oki, Happo, Tokyo, Ijira, Hedo, Ebino, Ogasawara) after 2014. The sampling interval was 2 weeks, 1 month, and seasonally depending on the sampling schedule at each site. The sulfur isotopic ratios were measured by stable isotope mass spectrometer. Canyon Diablo Troilite (CDT) was used as the standard material. The analytical precision was ±0.16‰. The nss-sulfur isotopic ratio was calculated assuming that Na^+ is originated from sea water.

[Results and discussion]
At the monitoring sites located in the coast of the Sea of Japan, temporal variations of nss-sulfur isotopic ratio were characterized by seasonal variation with high in winter and low in summer season (e.g. Niigata-Kajikawa, +2-+4.9‰). Similar seasonal variations were also found in Tokyo and Ijira, where locates the Pacific coast sites. However, nss-sulfur isotopic ratio in Tokyo and Ijira was lower with smaller seasonal variation in comparison with those at the coastal site of the Sea of Japan (e.g. Tokyo, -0.73-+4.0‰). It was reported that sulphate in the air mass in China were strongly affected by coal combustion (0-15‰, Xiao et al., 2011), whereas that in Japan were derived from oil combustion, which was negative values (-2.7‰, Ohizumi et al.,1997). Furthermore, it was reported that nss-sulfur isotopic ratio in aerosol in central China is 4.5±1.3‰ (Li et al., 2013). The difference of nss-sulfur isotopic ratio and these seasonal amplitude between coastal site of Sea of Japan and Pacific coast suggests that transboundary transported sulphate were mixed with the emitted sulphate from domestic (Japan) sources.

At Happo, mountain monitoring site (1850 m asl), nss-sulfur isotopic ratio were almost constant value (4.7±1.2‰) except for several samples in spring. Contribution of Asian dust will cause the higher values. Almost constant value at Happo suggests that nss-sulfur isotopic ratio is minor contribution from local anthropogenic sources, indicating the transboundary transport from the Asian continent.

At Ogasawara, remote marine site, nss-sulfur isotopic ratios were high in spring-summer and low in winter, which were opposite seasonal variation. The higher nss-sulfur isotopic ratio in

spring-summer was possibly the biogenic emissions.
Relative contributions of sulfur with different sources have been evaluated with mass balance model. It was clear that the transboundary transported sulphur deposition in the coastal site of Sea of Japan (e.g. Niigata-Kajikawa 1.1-20 mg m^{-2} day^{-1}) is two-three times larger than those in the Pacific coast (e.g. Tokyo 0.03-6.7 mg m^{-2} day^{-1}).

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Keywords: sulfur isotopic ratio, precipitation, transboundary transport

Annual and vertical variation of strontium isotopic ratio in two forest catchment in Japan

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[Introduction]

In the case of light elements such as sulfur, when it is used by plants in the ecosystem, isotopic ratio is varied by isotopic fractionation. It is caused by that light isotope is easier taken up to plants than heavier isotopes. On the other hand, in the case of heavy elements such as strontium (Sr), isotopic fractionation is almost negligible level, and isotopic ratio is only varied by component mixture from different sources. Therefore, such as elements are more suitable to estimate the contribution of some sources in the ecosystem. In this study, we discuss about annual and vertical variation of Sr isotopic ratio in two forest catchment, Kajikawa catchment in Shibata city, Niigata Pref. and Ijira catchment in Yamagata city, Gifu Pref.

[Methods]

We collected rainfall, streamwater and soil solutions in the slope soil in each catchment and determined Sr isotopic ratio of samples which collected from Dec. 2013. Soil solutions were collected at the three plot, upper, middle and lower slope and two depth, 20 and 60 cm. Sr isotopic ratio was determined by thermal ionization mass spectrometry (TIMS) of Research Institute for Humanity and Nature (Kyoto city, Kyoto Pref.). Sr isotopic ratio is shown as ^{87}Sr/^{86}Sr calculated based on the standard substance NBS987.

[Results and discussion]

Figure shows analysis results of ^{87}Sr/^{86}Sr in both catchment. In the case of rainfall as input side, annual variation range is comparative larger than streamwater as output side. In Kajikawa catchment, the highest value was shown in spring season when the amount of yellow sand increased. Second highest season was winter when sea salt contribution and amount of Sr derived from the continent increased by monsoon. These variation of Sr isotopic ratio indicate the contribution from different sources. In winter season, sea salt contribution rate became high level, and ^{87}Sr/^{86}Sr value became close to seawater which value is about 0.709. In spring season, dust including soluble mineral (^{87}Sr/^{86}Sr: 0.711±0.001) derived from the continent made ^{87}Sr/^{86}Sr value higher than winter season. On the other hand, ^{87}Sr/^{86}Sr values of streamwater were stable throughout the year, and so far from rainfall value in both catchment. Sr concentration of streamwater was higher than rainfall over than one digit, and suggests correlation between calcium and magnesium. In addition, streamwater of Ijira catchment which geology include middle Paleozoic marine sedimentary rocks indicated higher ^{87}Sr/^{86}Sr value than Kajikawa catchment which include rich granite. These results suggests that major part of Sr which flowed out to stream in both catchment was not derived from atmospheric deposition, probably from geologic origin.

In this study, we also discuss about the vertical variation including soil solution value and comparison with sulfur (S) isotopic ratio.

[Acknowledgements]

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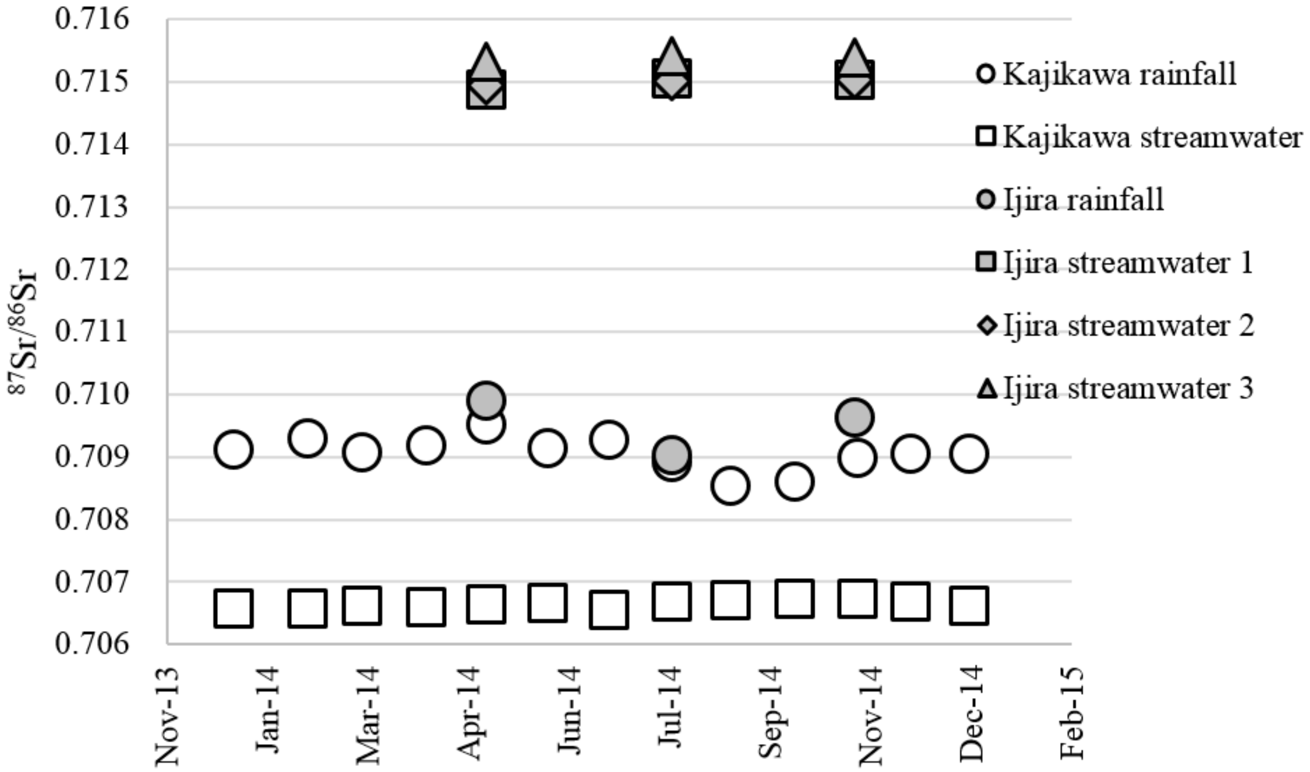
organizations.

Keywords: strontium isotopic ratio, rainfall, stramwater



Chemical separation of environmental materials for Stable Isotope Analysis

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Stable Isotopes (SI), especially Sr-Nd-Pb isotopes, had been widely used as powerful geochemical tracers in Earth sciences. In recent, these isotope ratios have been applied to the biogeochemical tracer in traceability research. Since its invention, multi-collector ICP-MS (MC-ICP-MS) has been widely used for isotope analysis of metal and metalloids, including Sr and Pb because of their higher ionization efficiency of the ICP ion source, the enhanced sample throughput, and the flexibility of sample introduction. However, the chemical separation is still adapted from those developed for TIMS, and not modified for the use of MC-ICP-MS. The higher ionization efficiency makes almost all introduced component to ionize, causing matrix effect to the analyzed data and deterioration of the machine. Diverse environmental samples, especially organic materials cause a problem during chemical separation that could not expect. It should need that the element for MC-ICP-MS analysis should be clearly separated from matrix component not only cation elements but also organic materials.

The purpose of this study is to describe a newly developed sequential separation of Sr, Pb, and Nd with Sr resin and nitric acid solution reducing the processing time, the amount of acid reagent solution, and evaporation steps.

Two type of separation methods has developed depending on the elements. One is single separation method and the other is multi-separation ones. Single separation method separates a small amount of Sr (~1ug Sr) or Pb (~1ug Pb) only with restricted volume of the resin. This method merits fast recovery of the target element and direct analysis of Sr isotope ratios with MC-ICP-MS. The Pb solution recovered by hydrochloric acid evaporated and dissolved in nitric acid for isotope analysis. Multi-separation method uses a large amount of the resin to separate REE, Sr, and Pb in a row. The REE solution follows further separation of matrix elements with a cation exchange resin to purify rare earth elements. Nd elements purified from REEs with Ln resin.

To reduce organic materials from the Sr resin, a polymeric adsorbent resin is used in the lower layer of the Sr resin. For water samples, it is recommended to use hydrofluoric acid during sample evaporation, because natural water contains silica as a major component and poor separation in the resin. During these methods, the elements recovered up to 95% of Sr and 99 % of Pb of the loaded samples, respectively. The blank levels of Sr and Nd in the separation process were ~10pg and ~20pg, respectively.

Keywords: Stable isotope, MC-ICP-MS, chemical separation, Sr resin

Evaluation for information on paleoclimate and event recorded in stable isotopic compositions of an annually-laminated tufa, Asama volcano, central Japan

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Here we present analytical results of $\delta^{18}O$ and $\delta^{13}C$ in an annually-laminated tufa Asama volcano, deposited from AD. 1999 to 2012, and discuss its implication of paleoclimate and volcanic activity around Asama volcano.

Keywords: Tufa, Stable isotope, Volcano

Tracing of the freshwater fish movement patterns and water dynamics by strontium stable isotope ratios and the elements concentrations analysis

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In coastal area of Otsuchi, Iwate prefecture, spring water is abundant and thereby the rare freshwater three-spined stickleback (Gasterosteus aculeatus), which prefer cold water environment, inhabit here. This area was damaged by the tsunami caused by the 2011 off the Pacific coast of Tohoku Earthquake. During the reconstruction and recovery works, the understanding of the habitat range and the environmental conditions of the three-spined sticklebacks contributes to the appropriate ecosystem management. Strontium stable isotope ratio ($^{87}Sr/^{86}Sr$) has been used as a powerful indicator to trace the past movement patterns of fish because of the same $^{87}Sr/^{86}Sr$ value between fish and the water of its habitat. Therefore, we used $^{87}Sr/^{86}Sr$ indicator to elucidate the three-spined stickleback movement patterns.

First, we investigated the relationship between the $^{87}Sr/^{86}Sr$ of three-spined stickleback whole otoliths (ear bone) and the water in isolated water bodies. We confirmed that both water and fish have almost same $^{87}Sr/^{86}Sr$.

Second, by analyzing the water of different rivers and tributaries in Otsuchi, we found that, just in one tributary of Kozuchi mainstream, $^{87}Sr/^{86}Sr$ and many element concentrations (e.g. Ca, Sr) were remarkably different between the upstream and downstream. Using $^{87}Sr/^{86}Sr$ and Sr concentration values of water, the downstream water was estimated to be mix of the upstream water, mainstream water and seawater (Contribution ratio; upstream: 82.6%, mainstream: 16.7%, seawater: 0.6%).

Then, using $^{87}Sr/^{86}Sr$ and elemental concentration of otoliths of fish captured in the upstream and downstream, we estimated their past movement patterns. We compared the water $^{87}Sr/^{86}Sr$ between the upstream and the downstream and the $^{87}Sr/^{86}Sr$ of otolith. As a result, it was possible to identify that stickleback lived in either upstream or downstream within single tributary; three-spined sticklebacks captured in the upstream (40 individuals in total) were not likely to move from downstream, while the fish caught at downstream (13 individuals in total) were estimated to have moved from the upstream or the mainstream. Furthermore, in the downstream, some fish otoliths showed a larger value of Sr/Ca than the others, which means that those fishes lived in high salinity environment in the past suggesting that the downstream population of three-spined stickleback was able to live in both fresh and brackish water environments.

Keywords: strontium stable isotope ratio, otolith, fish, water

The apterous winter stonefly fauna and its feeding on seasonal snowfields in Japan
- Analysis of the food web using C and N Stable Isotope –

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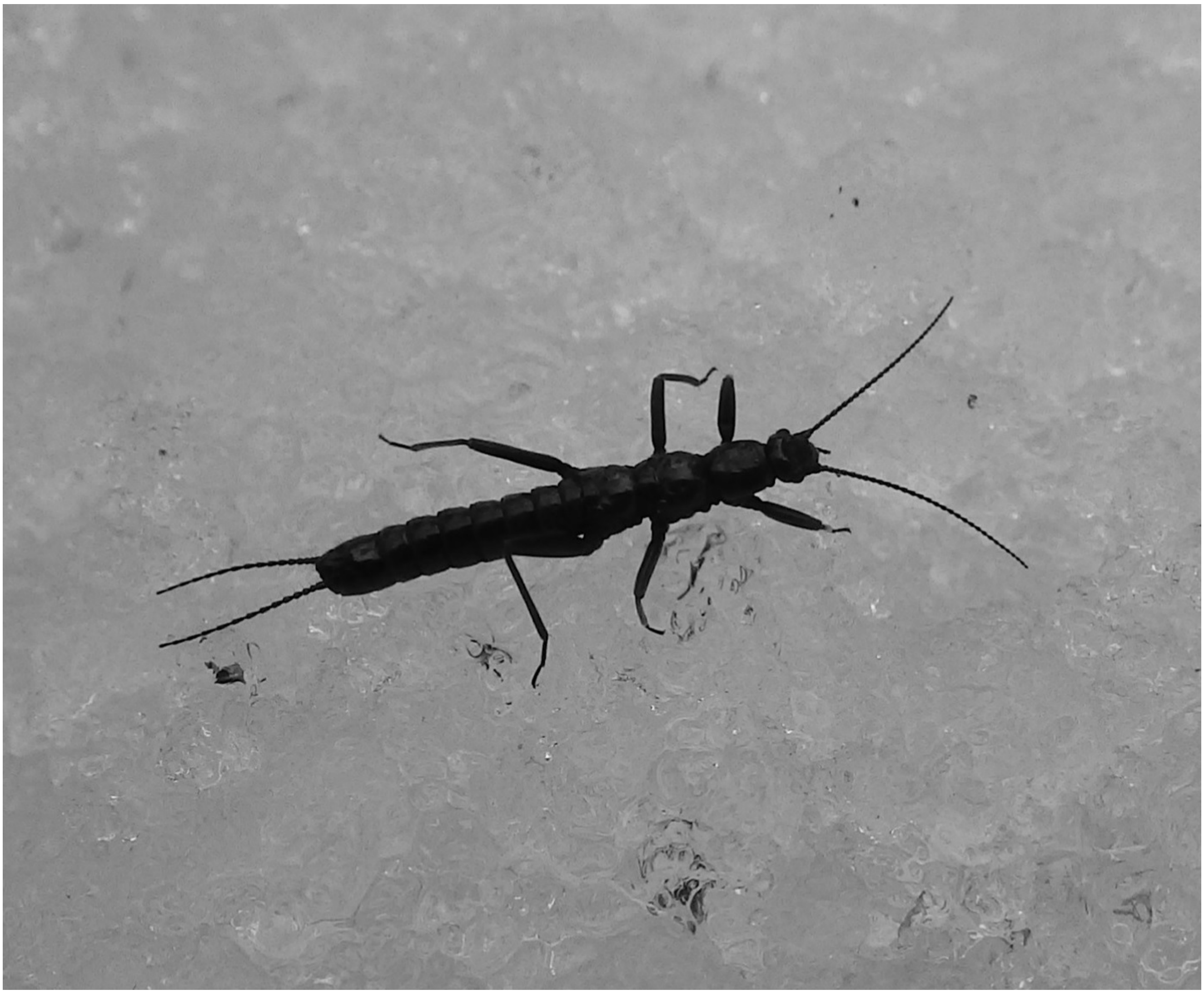
In snow and ice environments, there are diverse living organisms that can be active and grow in cold temperature. For example, snow algae, heterotrophic bacteria, and insects can be found on snow fields in Japan. An apterous winter stonefly is one of the typical insects that appear on snow surface in mountainous regions during winter or spring in Japan. However, their food web, particularly food resources of the stoneflies on snowfields, is still not well-known. It is important to clarify the food web of the organisms living on snowfields to understand carbon and nitrogen cycles quantitatively in the snow and ice environments. The stable isotope ratios of carbon and nitrogen of the specimens provide a means of analyzing their food resources. In this study, we analyze the food webs of the winter stoneflies and other organisms on Japenese snow fields using C and N stable isotope analysis.

The stonefly specimens were collected on the snow surface at five sites in Japan during 2015. The study sites included snow fields in Shinjo in Yamagata (altitude: 150 m), in Ushitakeonsen in Toyama (490 m), in Uonumakyuryo in Niigata (210 m), in Mt. Gassan (1300m altitude), in Tateyama Mountains (2300 m). Furthermore, other specimens including springtails, spiders, leaves, snow algae and particulate organic matter in snow were collected. The stable isotopes showed that potential food sources of apterous winter stoneflies are springtails, aphids, small spiders, snow algae, particularly dead leaves.

Keywords: stonefly, food web, stable isotope



Effects of additional nitrogen input and different river slope on the outflow from upstream nitrogen-saturated forests

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Non-point source nutrients, such as excess fertilizers from agriculture and human sewage water, are main cause of eutrophication of lakes, streams, and coastal areas. The upstream areas of suburbs of Fukuoka are covered with nitrogen-saturated forests, and supplied high concentration NO_3^- river water to the downstream. Such nitrogen-saturated forests have large impact on the downstream water quality (Chiwa et al., 2012), but less information was available of the mechanism of the changes in NO_3^- concentration from upstream to the downstream.

The objectives of this study were to clarify (1) whether the nitrogen dynamics (the concentration of NO_3^-, $\delta^{15}N_{NO3}$ and $\delta^{18}O_{NO3}$) in the downstream were affected by the nitrogen-saturated forests in the upstream, (2) the influence of nitrogen runoff from agriculture and urban area in the downstream, and (3) the influence of the river gradient on denitrification.

NO_3^- concentration and the stable isotopes, $\delta^{15}N_{NO3}$ and $\delta^{18}O_{NO3}$ were measured in the Tatara River Basin, located in northern Kyushu, western Japan. Water samples were mainly collected from branch rivers; Ino River, Sue River and Umi River. Deciduous mature forest dominates the ridges in the upstream, with paddy, farmland and urban dominating in the middle and downstream. Topographic Index (TI) was calculated by using Digital Elevation Model (DEM) with ArcGIS software.

Our results showed that the NO_3^- concentration in the upstream of three rivers (33.9-82.8 μmol/L) were higher than the other non-saturated forests. Lower $\delta^{18}O_{NO3}$, higher NO_3^- and $\delta^{15}N_{NO3}$ in the downstream were be found in Sue River and Umi River. On the other hand, Ino River showed different trend that lower NO_3^- concentration, higher $\delta^{15}N_{NO3}$ and $\delta^{18}O_{NO3}$ concentration in the downstream. The higher NO_3^- concentration in the downstream in Sue River and Umi River was affected by the increasing agriculture and urban areas in the downstream. However, the NO_3^- concentration in the downstream of Ino River decreased with the increasing agriculture and urban areas.

The average TI value showed no difference between rivers. However, the distribution of TI values in Ino River was different from other rivers. Ino River has smaller changes in gradient from upstream to downstream and fewer agriculture and urban areas in the downstream than the other two rivers. These differences could result in the higher probability of denitrification occurring and the lower NO_3^- loading in the downstream in Ino river.

The nitrogen-saturated upstream forests in this watershed has large impact on the nitrogen sources to the downstream. In the areas with steeper gradient, the high concentration of nitrogen runoff was supplied to the agriculture and urban areas. On the other hand, in those areas with gentle gradient, the nitrogen loading in the downstream also decreased due to the gentle gradient of the watershed and it was suggested that the rate of denitrification and decrease NO_3^- concentration in the downstream was also high.

Keywords: nitrogen saturation, stream water quality, nitrogen isotope, land use, Topographic Index

Trial application of oxygen, carbon and strontium isotope analysis in tooth enamel for identification of past-war victims for discriminating between Papuan, Japanese and US soldiers

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Stable isotope analysis has undergone rapid development in recent years and yielded significant results in the field of forensic sciences. In particular, carbon, oxygen and strontium isotopic ratios in tooth enamel obtained from human remains can provide useful information for prescreening for identification of remains. The aim of this study is to evaluate this method for discriminating remains between local people, Japanese and US soldier in the battlefield of New Guinea of World War 2.

In this study, the carbon, oxygen and strontium isotopic ratios in the tooth enamel of the examined Papuan (South highland, East New Britain and Bougainville) and Japanese (Tottori) individuals is compared to previously reported data for US individuals, and statistical analysis is conducted using a discriminant analysis.

US populations can be extracted from these groups accurately. The discrimination between the Papuan and Japanese is found to be partly accurate. Thus, the present method has potential as a discrimination technique for these populations for use in the examination of mixed remains comprising Papuan, Japanese and US fallen soldiers.

Keywords: Stable isotope analysis, War victim, Geographic origin

Stable isotope analysis and radiocarbon dating on human skeletal remains from the Yoshigo shell mound of the Jomon period

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The author investigated radiocarbon ages and carbon and nitrogen stable isotope ratios in human skeletal remains excavated from the Yoshigo shell mound in the Aichi Prefecture. Although a large number of skeletal remains has been excavated from the Yoshigo shell mound, ages of these skeletal remains are still unclear. Radiocarbon dating on human bone collagen can estimate the ages of the individuals, but the precise estimate on marine food dependence is needed to correct marine reservoir effect on radiocarbon dates. This study investigated radiocarbon ages and carbon and nitrogen stable isotope ratios of human skeletal remains from the Yoshigo shell mound during the Late-Final Jomon period. By using the Bayesian mixing model on the carbon and nitrogen isotope ratios, the dietary dependences on marine food of each individual were estimated. Then, the results of radiocarbon dating were corrected. The results of this calibrated ages of human skeletal remains exhibited the ages during the Late-Final Jomon period, and it can enable us to analyze the dietary difference through time.

Keywords: Human skeletal remains, Stable isotope, Diet

Building an ontology of isotope environmental science

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In its Midterm Master Plan Phase 3 (FY 2016-2021), the Research Institute for Humanity and Nature (RIHN) will promote research and development in the visualization of isoscapes, or spatial information on environmental isotope ratios, including the disclosure and sharing of measured isotope data. As a preliminary study in this area, the authors built an ontology of isotope environmental science using the following method. First, in a brainstorming session with researchers of informatics and isotope environmental science, participants sorted the relationships of vocabularies extracted from lectures in the RIHN citizen seminars by posting sticky notes on a wall. This brainstorming session successfully organized the ontological relationships of terms in isotope environmental science, particularly in hydrology, ecology, geochemistry, and the social sciences. Then, the ontology was structured and visualized by using an analytical program called Hozo (http://hozo.jp). To continue this approach, the ontological model should be extended to other related fields of research, such as archaeology and geography. Furthermore, the authors plan to apply the RDF (Reference Description Framework) for describing the ontological relationships within the triple of subject, predicate, and object.

Keywords: Isotope environmental science, ontology, knowledge base, terminology