

## Radon Concentration around Tachikawa Active Fault

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Characteristics of a radon concentration distribution around the Tachikawa active fault will be focused in this report.

According to the Headquarters for Earthquake Research Promotion, occurrence potentials of earthquake in active faults in Japan were updated after the Tohoku Earthquake (March 11, 2011). The report denotes the potential of the Tachikawa active fault while next 30 years was increased from 0.5-2% to 0.9-2%. In order to monitor a state change of the fault system by groundwater analysis, we have been surveying water qualities of spring water and hot-spring water.

Our survey of shallow spring waters around the fault in 2012 revealed that the radon concentration of shallow groundwater was affected by a cultivation process of groundwater on the ground surface around the northwest area of the Tama district. Therefore the shallow groundwater around the fault is not appropriate to get information on the state change of the fault. Our survey of deep hot-spring water around the fault in 2013 indicated the good relation between the radon concentration distribution and the location of the fault. The nearer the location of the hot-springs to the fault is, the higher the radon concentration becomes. Therefore the deep groundwater around the fault is useful to monitor the state change of the fault.

Keywords: Grounwater, Hot Spring, Radon, Active Fault

## Reconstruction of summer precipitation during last two millennia in central Japan by tree-ring oxygen isotope ratios

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### Introduction

To establish long-term plans of flood control by river management and/or water supply by dam construction, it is necessary to predict future change in precipitation due to global warming. Because precipitation can change at various time scales, it is important to understand the statistical probability of giant flood occurring once a century or millennium. However, due to the shortage of hydrological and meteorological records, it has been difficult to understand statistical characteristics of precipitation in very long time scales. Recently, we have found that tree-ring cellulose oxygen isotope ratios (d18O) record summer precipitation in the corresponding years very precisely and started reconstructions of long term summer precipitation changes in annual time resolution all over Asia monsoon area. Here, we present the two millennial data of summer precipitation in central Japan based on the tree-ring d18O of Hinoki cypress samples obtained from various periods and discuss its meanings in historical hydrology.

### Fundamental of precipitation reconstruction

Because cellulose is produced from carbohydrate originally photosynthesized in leaf, its d18O records changes in leaf water d18O. Leaf water d18O is controlled by two meteorological factors, precipitation d18O and relative humidity. It is positively correlated with precipitation d18O and negatively correlated with relative humidity due to the transpiration process. Because there are negative correlation between rain amount and precipitation d18O (amount effect) and positive correlation between rain amount and relative humidity, we can finally realize that there is distinct negative correlation between rain amount in growing season and tree-ring cellulose d18O.

### Tree ring samples for long d18O time series

Because trees can seldom live more than several hundred years, we must collect many tree-ring samples during various periods to establish reliable time series of tree-ring d18O beyond last two millennia, not only from living trees, but also from old architectures, archaeological remains, tree logs buried by landslides and so on. Tree-ring samples of Japanese cypress were collected from a certain wide area in central Japan, where we can find consistent tree-ring d18O variations among different trees at different sites and combine many d18O time series, according to the consistent d18O patterns, to establish statistically reliable two millennia length of tree-ring d18O time series.

### Application to historical hydrology

The time series of tree-ring d18O was first compared with various meteorological, historical and archaeological evidences on summer precipitation in Japanese history, including modern instrumental meteorological records, summer rainfall amounts quantitatively reconstructed from early modern diary weather notes, medieval documentary records on notorious flood and drought, and pit-house number in lowland plains of ancient Japan, and it was demonstrated that the tree-ring d18O coincides those records very well and successfully reconstruct past change in summer precipitation irrespective of reconstructed periods. However, the tree-ring d18O of Japanese cypress has a distinct age effect where it decreases gradually during its all life time, so that we cannot extract real precipitation trends more than 200 years periodicity from the cypress data. By comparing the tree-ring d18O time series with historical flood records in various rivers in central Japan, we found that there are very good coincidences in decadal time scale before 11th century, but the relation becomes diverse in 18th century, so that good coincidence disappeared in highly developed area where artificial flood control and/or excess logging might have influenced river hydrology. The summer precipitation records based on tree-ring d18O are useful to elucidate both of natural variability and human responses on the historical precipitation changes.

Keywords: two millennia, precipitation, central Japan, tree ring d18O

## Water vapor origins in all over Japan in winter simulated by the regional isotope circulation model

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In this paper, water vapor origins in all over Japan in winter were simulated by using a regional isotope circulation model with stable isotopes in water ( $\delta^{18}\text{O}$  and  $\delta\text{D}$ ). Precipitation and stable isotopes were simulated for the period between December to February in 2001 — 2010, spatial distributions of them were reproduced observations well. Simulated daily sea-level pressure patterns were divided into two types: winter monsoon (WM) type and extratropical cyclone (EC) type. In the WM type, precipitation rate was high and low along the Japan Sea side and the Pacific Ocean side, respectively. Spatial distribution of  $\delta^{18}\text{O}$  in precipitation was recognized the latitude effect (values decrease with increasing latitude) on the Pacific Ocean and the Japan Sea. Spatial distributions of d-excess ( $=\delta\text{D}-8\times\delta^{18}\text{O}$ ) in precipitation and evaporation were above 16 ‰ around Japan, those were extreme high (above 22 ‰) especially on the Pacific Ocean and the Japan Sea. Simulated water vapor evaporated from the Japan Sea was predominant in all over Japan in the WM type without southwestern islands of Japan. Interestingly, a portion of this moisture moved eastward to the Pacific Ocean, however, the moisture was not contributed to total amount of precipitation along the Pacific Ocean side because it was little precipitation. In contrast, precipitation rate was high in all over Japan in the EC type. Spatial distribution of  $\delta^{18}\text{O}$  in precipitation was recognized the latitude effect on the Pacific Ocean and the Japan Sea and the amount effect (values decrease with increasing precipitation amount) across Japan. Spatial distributions of d-excess in precipitation and evaporation were below 14 ‰ around Japan without the western part of the East China Sea. Simulated water vapor evaporated from the Pacific Ocean was predominant in all over Japan. Comparing  $\delta^{18}\text{O}$  and d-excess in precipitation between the WM type and the EC type, those were 2 ‰ and 8 ‰ higher along the Japan Sea side in the WM type than in the EC type, respectively.

Keywords: stable isotopes in precipitation, water vapor origins, regional isotope circulation model, in all over Japan

## Preliminary results of the stable isotopes in precipitation throughout Japan observed in 2013

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Stable isotopes in precipitation are used for a proxy of climate change, which is related with temperature or precipitation amount. By using the atmospheric general circulation model with stable isotopes in water, water vapor origins (where the water vapor evaporated from?) can be estimated to determine the air mass transportation process. There are a lot of studies to observe stable isotopes in precipitation at only one or a few places in Japan. Tanoue et al. (2013) revealed seasonal variation and spatial distribution of stable isotopes in precipitation over Japan from the previous observational data at about 50 stations. However, spatial and temporal variations of stable isotopes in precipitation across Japan are still unknown, because a specific site and an observation period are different among the previous studies. Intensive observation of stable isotopes in precipitation over the whole Japan is required during the same period.

The Isotope Mapping Working Group of Japanese Society of Hydrological Sciences was conducted intensive observation of stable isotopes in precipitation throughout the year in 2013 (IOP2013). More than 2,000 precipitation samples are already collected at about 50 stations, and are analyzing its stable isotopic ratios by the Isotope Ratio Mass Spectrometer (Delta-V, Thermo Scientific) in Kumamoto University. Stable isotopic ratios in precipitation across Japan will be considered the relationships with locations (i.e. latitude, altitude, and distance from the coastline) and meteorological elements (i.e. temperature, precipitation amount, winds, specific humidity). Also, water vapor origins are estimated by using atmospheric general circulation model with stable water isotopes. Finally, the equations to reproduce stable isotopic ratios in precipitation at a specific place in Japan will be determined by its location and/or meteorological elements. It's a useful for the paleo-climate change as a proxy data of temperature and/or precipitation amount in the past.

In this study, we present the preliminary result of the IOP2013.

Keywords: Stable isotopes in precipitation, d-excess, Japan, IOP2013

## Tracking phosphorus sources and cycling in freshwater: stable isotope approach

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Stable isotope technique is increasingly used to provide ecological information to understand biological cycling and tracking environmental pollutants. The technique used for tracing phosphorus (P) in water is primarily based on the possibility of distinguishing the different P inorganic sources by phosphate oxygen isotopic signatures ( $\delta^{18}\text{O}_p$ ) [1]. To date, there are only few studies to examine P cycling on watershed scales using the phosphate oxygen isotope analysis.

Here we aim to characterize individual  $\delta^{18}\text{O}_p$  signatures of water, natural sources and potential anthropogenic sources in the Yasu River, the largest tributary river in the Lake Biwa Watershed. Special attention was paid to identify primary sources of P loadings in the Yasu River, associating with the land use pattern in its each catchment.

### Materials & Methods

We collected river waters from 19 sites across the mainstream of Yasu River and its branches, whose catchment areas greatly vary in land use pattern. We also gathered water samples from 8 sewage treatment plants, 2 agricultural waste water plants and one livestock farm as point sources of anthropogenic P. We regarded phosphate fertilizers and sewage treatment plant waste waters as indicators for agricultural and domestic non-point P sources, respectively. We also collected sand from the riverbed of 5 headwaters as natural P sources. The sand samples were acid extracted to desorb dissolved inorganic phosphates [2]. These samples were treated with magnesium-induced coprecipitation (MagIC) method for phosphate extraction and then converted to silver phosphate after purification through the sequence of resin separation and precipitation [1,3]. We determined  $\delta^{18}\text{O}_p$  for each of these silver phosphate samples using a thermal conversion elemental analyzer coupled to a continuous flow isotope ratio mass spectrometer via a helium stream.

We constructed an isotopic mixing model to estimate the relative contribution of individual P sources in each catchment.

### Results & Discussion

A wide range of  $\delta^{18}\text{O}_p$  in river water was detected. This indicates that this technique is a promising tool to trace P sources in the watershed ecosystems.

The isotopic mixing model showed that urban land use accounted for spatial variation in the relative contribution of domestic P loadings though there were some uncertainty in the model simulation.

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[3] McLaughlin et al. (2004) Limnol. Oceanogr. : Methods 2, 204-212

Keywords: Biological recycling, Eutrophication, Land use, Non-point phosphorus loading, Phosphate oxygen isotope analysis

## Stable isotopic map of spring water and surface water in the Shirakami Mountains, Japan

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The Shirakami Mountains is the general name given to an extensive mountainous region of 130,000 hectares ranging from the southwest of Aomori to the northwest of Akita prefecture. Within this area are 16,971 hectares of land, enclosing virgin forests of Japanese beech, which were registered as a world heritage region in December 1993. However, environmental impact by acid rain at the Shirakami Mountains is becoming an issue these days. Acid rain deposits nitrates that can lead to increases in nitrogen in forests. So we have studied about the chemical and isotopic compositions of river and spring waters in the Shirakami Mountains area, to clarify origin and geochemical characteristics since 2011.

The result of the investigation was that  $\delta\text{-}^{18}\text{O}$  and  $\delta\text{-D}$  of water samples showed -8.8 to -11.5 permil and -48.8 to -64.8 permil, respectively. Stable isotope composition of the samples roughly resemble those of meteoric water ( $\delta\text{-D} = 8\delta\text{-}^{18}\text{O} + 20$ ), thereby indicating that these are local meteoric water. The least-squares regression line for all data is:  $\delta\text{-D} = 5.7\delta\text{-}^{18}\text{O} + 1.7$  ( $R^2 = 0.88$ ). On the other hand, the regression line calculated with data from NW-part is:  $\delta\text{-D} = 6.0\delta\text{-}^{18}\text{O} + 0.5$  ( $R^2 = 0.98$ ). Characteristics of two regression line suggest that these water origin were brought by rain from different air mass, respectively.

Keywords: The Shirakami Mountains, Stable isotope map

## Source of spring water and nitrate in northern foot of Mt.Fuji

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Water chemistry of spring water in Northern foot of Mt. Fuji is discussed with special reference to its source of water and nitrate. Monthly spring water and river water samples were collected from 8 springs and 7 locations of the 3 rivers, from June 2013 to January 2014. Land use of the study area are urban located about <1000m, forest distributed >1000m and forest limit is about 2500m. The oxygen isotope range of all spring water samples shows temporal variation (>1.0 permil), which suggests the possibility of the water changes of groundwater water recharge elevation. The nitrate-nitrogen concentration ranges from 0.2 to 1.8 mg/L and from 0.1 to 2.2 mg/L in river water and spring water samples respectively. Similarly, nitrate-nitrogen isotope values ranges from 2.7 to 9.9 permil and 1.4 to 10.4 permil in river water samples and spring water samples respectively. Although nitrate concentration was low, nitrogen isotope values overlaps with forest soil nitrogen and sewage or manure nitrogen. This trend suggests that the recharge elevation of the spring water might spread across a wide area. This presentation will discuss about recharge processes of the spring water including temporal variation of the isotopic values and water quality.

Keywords: Mt.Fuji, spring water, oxygen and hydrogen isotopes in water, nitrogen and oxygen isotopes in nitrate

## Spatial distribution of vanadium concentrations and water isotopes in lake bottom water from Lake Kawaguchi

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Spatial distribution of vanadium concentrations was examined in bottom water from Lake Kawaguchi, on the northern foot of Mt. Fuji, in order to test the hypothesis that the water outflow from underwater springs in Lake Kawaguchi could affect the vanadium concentrations of lake water. The samples were collected from the east lake basin and the Funatsu lake basin on July 14-August 2, 2005, and from the west lake basin on November 1 and October 31, 2013. Vanadium concentrations in the bottom water from Lake Kawaguchi range from 0.66  $\mu\text{g/L}$  to 3.18  $\mu\text{g/L}$ . Because vanadium concentration in precipitation is generally  $<0.1 \mu\text{g/L}$ , the variations are most likely attributed to the dilution of the lake water due to precipitation, and/or the input of water masses with high vanadium content. Although the concentrations of vanadium are significantly lower than those in the groundwater of the Mount Fuji, we found a relatively vanadium-enriched water mass along the southern coast of the west lake basin, off coast of Higashiken lava flow from Mount Fuji. The area matches well with the potential location of underwater springs in Lake Kawaguchi, and the lack of any riverine input around the area suggests that a water mass with relatively high vanadium concentration is likely provided from underwater springs that are located in off coast of the basaltic lava flow of Mount Fuji. We also plan to discuss the source of underwater springs based on stable water isotope ratios in the presentation.

Keywords: Kawaguchiko, vanadium, stable water isotopes, Mount Fuji

## Contribution of volcanic gas to spring waters in the Mt. Yotei.

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The Mt. Yotei is a stratovolcano in southwest Hokkaido with an altitude of 1893 m. There are a lot of springs in the foot of the mountain and most of these are found from the altitude of 200 to 260 m. This elevation is consistent with that of boundary between volcanic rocks of the volcano and basement consists of tuff and pumice. Northern, eastern, and southern slopes of the mountain are covered by pyroclastic fall and reworked deposits of that, while the western slope is covered by lava erupted from summit and parasitic volcanos in western slope (Katsui, 1956).

It is revealed that springs in eastern and southern slopes tend to show larger discharge and lower dissolved components, while those in the western slope tend to show smaller discharge and higher dissolved components (Yamaguchi and Sato, 1971; Yamaguchi, 1972; Tsurumaki, 1989). Because of higher  $\text{HCO}_3^-$  and free  $\text{CO}_2$  gas concentrations and higher temperature of springs in the western slope, addition of volcanic gas to those is estimated (Yamaguchi and Sato, 1971; Tsurumaki, 1989). However, contribution of volcanic gas to groundwater has not been evident. In this study, contribution of volcanic gas to spring waters will be discussed based on dissolved components and isotopic compositions.

Spring water samples were collected on August 2013, and were analyzed for major dissolved components, isotopic compositions ( $\delta\text{D}$ ,  $\delta^{18}\text{O}$ ,  $\delta^{13}\text{C}$ ,  $^3\text{He}/^4\text{He}$ ), and groundwater-age indices (CFCs and  $\text{SF}_6$ ). Dissolved components of spring waters are  $\text{Ca-HCO}_3$  type or  $\text{Na-HCO}_3$  type. Spring waters in the western slope tend to show higher  $\text{HCO}_3^-$  and free  $\text{CO}_2$  gas concentrations. These results are consistent with those of previous studies.  $\delta^{13}\text{C}$  of dissolved inorganic carbon (DIC) in spring waters in eastern and southern slopes are -21.7 to -17.7 ‰, while those of the western slope are -18.1 to -3.0 ‰. This result suggests that spring waters in western slope contain DIC of which origin is different from that contained in springs in eastern and southern slopes. Dissolution of marine carbonates is unlikely because groundwater flow in volcanic rocks. Relationship between  $\delta^{13}\text{C}$  and inverses of concentrations of total carbon are plotted in mixing zone of volcanic  $\text{CO}_2$  and soil  $\text{CO}_2$ , and spring water showing higher  $\delta^{13}\text{C}$  and total carbon concentration were correspond to higher contribution of volcanic  $\text{CO}_2$ . These results suggest that spring waters in western slope showed higher  $\delta^{13}\text{C}$  and total carbon concentrations because of addition of volcanic  $\text{CO}_2$ .  $\delta\text{D}$  and  $\delta^{18}\text{O}$  of spring waters are plotted along the meteoric water line (GSJ, undisclosed data), suggesting contribution of magmatic water is quite small. Effect of parasitic volcanos in the western slopes is suggested by Yamaguchi and Sato (1971) as a possible factor for higher contribution of volcanic gas to springs in the western slope. Further discussions based on dissolved components, isotopic compositions, and groundwater-age indices will be performed to reveal relationship between contribution of volcanic gas and groundwater flow system.

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## Estimation of recharge elevation and residence time for springs in Mt. Yotei

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Mt Yotei is a stratovolcano located on southwestern part of the Hokkaido. 17 springs with flow rate over 20 L/s distribute on the foot of the volcano body. We used the multi tracer method to estimate the recharge elevation and the residence time for the springs. Water samples were collected from 14 springs at August 2013, and were measured in stable isotope, Tritium, CFCs and SF<sub>6</sub>. In this presentation, we discuss the relationships between the scale of groundwater flow system and the hydrogeology of volcano body, based on the recharge elevation and the residence time for springs.

Keywords: Mt. Yotei, spring, recharge elevation, residence time, transient tracer

## Isotope characteristics of groundwater in and around Mt. Sambe, an active volcano in western Japan

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A study using a coupled hydrological and geochemical (water chemistry,  $\delta D$ - $\delta^{18}O$ ,  $\delta^{13}C$ , and helium isotope) approach is in progress to elucidate the groundwater system in and around Mt. Sambe, a 1126-m high active volcano located in western Japan. Groundwater in its south to east flanks proved to be characterized by high  $\delta^{13}C$  values (in the range between -9 - -5 ‰  $\delta^{13}C$ ) as well as an elevated concentration of total carbon compared to those in other flanks. These observations clearly indicate an admixture of magmatic fluid into shallow groundwater of the south to east flanks, being also in accordance with its water chemistry with an increased chloride ion concentration.

Keywords: active volcano, groundwater, water chemistry, isotopes, volcanic fluid

## Origin of the high-chloride groundwater in the central part of the Kanto Plain from the viewpoint of noble gas hydrology

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There exist three regions in the Kanto plain, central Japan, whose artesian groundwater is characterized by a high Cl<sup>-</sup> concentration: 1) central parts of the Kanto plain (Saitama Prefecture), 2) south-east parts of Gunma Prefecture), 3) floodplains and deluvial uplands along the lower reaches of Kokai and Tone rivers (Ibaraki and Chiba Prefectures).

As for that in the central parts of the Kanto plain, confined groundwater with a high Cl<sup>-</sup> concentration of up to 216 mg/l is obtained from the productive boreholes of 200-430 m depth. The area of Cl<sup>-</sup>-rich groundwater, spreading from the northwest to southeast, corresponds with the so-called Motoarakawa tectonic zone (ca. 10 km wide by 35 km long) bounded by the fault on its longer sides. The <sup>36</sup>Cl/Cl results of these high chloride groundwaters imply that admixture of meteoric water and sea water in the period of the Shimosueyoshi transgression (peak period at around 125,000 yrs. BP) is likely to account for its elevated Cl<sup>-</sup> concentration (Yasuhara et al., 2011). Morikawa et al (2006) conducted dissolved noble gas analyses for the groundwaters in the Motoarakawa tectonic zone. The results of helium isotopes are as follows; (1) there is a tendency of high <sup>4</sup>He concentration in the groundwaters inside the tectonic zone, (2) helium isotopic ratios (<sup>3</sup>He/<sup>4</sup>He) are relatively homogeneous with an end member of 0.8-1.1 x 10<sup>-6</sup>. Helium-4 concentration show clear positive correlation with chloride concentration. Combined with this correlation and characteristics of helium isotopic ratio, it is inferred that the groundwater from the tectonic zone is a mixture of meteoric water and high chloride saline water bearing with high <sup>4</sup>He or is stagnant old groundwater and that the groundwater flow system is distinct from those from outside of the tectonic zone.

In this study, we investigated the noble gas in the deeper groundwaters (hot springs) around the Motoarakawa tectonic belt and the high-chloride groundwaters from south-east parts of Gunma Prefecture to elucidate the origin of water and chloride component in the central parts of the Kanto plain. Low <sup>3</sup>He/<sup>4</sup>He ratios in the hot springs indicate that there is no interconnectivity between the high chloride groundwater and hot spring water around the Motoarakawa tectonic zone. In contrast, both <sup>3</sup>He/<sup>4</sup>He ratio and correlative <sup>4</sup>He and Cl concentration in the high-chloride groundwaters from south-east parts of Gunma Prefecture are similar to those in the groundwaters in the Motoarakawa tectonic zone.

Keywords: Kanto Plain, groundwater, chloride ion, Noble Gas, Helium isotope

## Geochemical study of hot spring waters and gases in Jozankei area, Hokkaido, northern Japan

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Jozankei hot spring area is located in the western part of Sapporo city, Hokkaido, northern Japan. In this area, hot spring waters >85 degree C. are springing out from the entire Toyohira river valley. Total discharge rate of hot spring waters is about 10 tons/min and the "heat energy index" is rank V (Fukutomi, 1965). Hydrogen and oxygen isotopic study clarified that the major origin of hot spring water is local meteoric water (Matsubaya et al., 1978)

To understand the other origin of Jozankei hot spring, analyses of chemical compositions, hydrogen and oxygen isotope ratios were carried out for hot spring waters from Jozankei area and its adjacent area. From the good correlation among chloride ion content, oxygen and hydrogen isotope ratio of hot spring waters in Jozankei area, it is clarified that one of the end member of hot spring waters of Jozankei area is originated from magmatic fluid and chemical composition of magmatic fluid is 3-5 NaCl wt%.

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Keywords: Jozankei, magmatic fluid, hydrogen and oxygen isotopic ratio

## Genesis of Kashio brine: slab-derived fluid

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In this study, we show the results obtained by investigation of a very saline spring named Kashio brine, central Japan and discuss its genesis and cause of chemical and isotopic features. The brine water is spouting out through the cracks of metamorphic rocks close to Median Tectonic Line (MTL) at 700m asl. The Cl concentration of water is greater than that of the seawater with <sup>18</sup>O-shifted isotopic composition. The brine is thought to originate from slab-derived fluid with the chemical and isotopic composition of water like the Arima hot spring, in spite of its low temperature and CO<sub>2</sub>-less features. The cause of this chemical character might be explained by a reaction with ultramafic rocks existed close to Kashio area which causes high pH condition to deposit all the CO<sub>2</sub> as CaCO<sub>3</sub>. The low temperature may be interpreted by the low upwelling rate because of the low <sup>3</sup>He/<sup>4</sup>He ratio (lower than 2Ra) due to the addition of crustal <sup>4</sup>He. The amount of NaCl is finally supplied to the river and is estimated to be 2000kg/day.

Keywords: Kashio brine, isotopic ratio, flow rate, slab-derived fluid

## GC/C/IRMS as a tool to evaluate the degradation of chlorinated organic compounds in groundwater

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The in-situ remediation techniques by microbial activity are used for reduction of chlorinated organic compounds in polluted groundwater. However, the evaluation of microbial activity for decomposition of chlorinated organic compounds is not clear by only the concentration analysis. So, we focus on the usage of the stable carbon isotope analysis of chlorinated organic compounds such as trichloroethylene (TCE), and its daughter products; cis-dichloroethylene (cis-DCE) and vinyl chloride (VC).

The stable carbon isotope ratio ( $\delta^{13}\text{C}$ ) of these organic compounds were analyzed by a GC (Agilent : 7890A) coupled to an isotope ratio mass spectrometer (SerCon : 20-22) with combustion interface (SerCon : GC-CP) (GC/C/IRMS). The system was equipped with a purge-and-trap concentrator (GL science : AQUA PT 5000J PLUS) connected to the GC. In the  $\delta^{13}\text{C}$  value measurement, the measurement limit was usually 200ng-C, and standard deviation in TCE, cis-DCE and VC were  $\pm 0.08$ ,  $\pm 0.37$ ,  $\pm 0.11$  permil, respectively.

Groundwater samples were collected at polluted site with and without bioremediation treatment.

In samples from without treatment site, even the concentration decreasing were detected, the  $\delta^{13}\text{C}$  values of TCE are almost same as original one. On the other hands,  $\delta^{13}\text{C}$  values of not only TCE but also the daughter products increase with a decrease in concentration of TCE. These results show the usefulness of GC/C/IRMS for distinguishing the reason of concentration reduction by in-situ microbial activity of decomposition of these organic pollutants from physical factors such as dilution, diffusion, and adsorption.

Keywords: GC/C/IRMS, chlorinated organic compounds, biodegradation

## Isotope characteristic of rain water and atmospheric vapor in Hiratsuka, Japan

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### Introduction

The stable isotope ratios of hydrogen and oxygen in meteoric water ( $\delta D$  and  $\delta^{18}O$ ) are affected by geological and climatic conditions. Global meteoric water line (GMWL) describes the average isotopic compositions in the world. According to Craig (1961), the relationship between  $\delta D$  and  $\delta^{18}O$  was expressed as

$$\delta D = 8\delta^{18}O + 10 \quad (1)$$

However those intercept are not always 10 in each area. In Japan, the meteoric water originates in both Pacific Ocean and Japan Sea. The effects of two seas vary due seasonally. The isotope ratio of atmospheric vapor is important for study of atmospheric circulation, however, the number of published paper is not so much. In this study, we investigate the d-excess ( $d = \delta D - 8\delta^{18}O$ ) of rain water and atmospheric vapor in Hiratsuka, Japan.

### Sampling methods

Samples were collected on the roof of a No.17 building at Shonan campus, Tokai University from May to December 2013. Rain water samples were collected based on a method described by Negrel et al. (2011) and Yoshimura (2002). The collection duration was days or hours in scale. Rain water samples were percolated through 0.2  $\mu m$  filter, and kept into a 100 ml low-density polyethylene bottle. Atmospheric vapor samples were collected through a trap cooled with ethanol and dry ice mixture. Samples were 42 of rain water and 11 of atmospheric vapor.  $\delta D$  and  $\delta^{18}O$  of samples were measured by a Cavity Ring-Down Spectrometer analyzer (model L2120-I from PICARRO). Some data of rain water, which were sampled several times in a day, were processed to be the average value.

### Results and discussion

Rain water shows a wide variation in  $\delta D$  and  $\delta^{18}O$  from -86.4 to +6.2 ‰ and -12.6 to -2.6 ‰, respectively. Atmospheric vapor shows a variation from -223.5 to -98.6 ‰ and -31.2 to -14.7 ‰, respectively. The  $\delta D$ - $\delta^{18}O$  relationship of rain water gives a regression line:  $\delta D = 9.2\delta^{18}O + 24.0$  ( $R^2 = 0.95$ ) and that of atmospheric vapor gives a regression line:  $\delta D = 7.3\delta^{18}O + 7.9$  ( $R^2 = 0.96$ ). The d-excess values show a variation from 4.4 ‰ to 33.2 ‰. In Japan, origin of meteoric water affects to d-excess (Waseda and Nakai, 1983). In case of Pacific Ocean, d-excess is low ( $10 \geq d$ ). In case of Japan Sea, d-excess is high ( $20 \leq d$ ). In this study, the d-excess was low in summer when southern winds were blown from Pacific Ocean as the seasonal wind, and that value was high in winter when northern winds were blown from Japan Sea. Samples of atmospheric vapor show also this trend. Suggesting that atmospheric vapor is influenced by the same effect of meteoric water. The meteoric water line of rain samples was affected by d-excess which reflects variations of moisture sources, which is the reason why the slope of this line would be bigger than GMWL.

Keywords: rain water, isotope

## Exploring the sources of sulfur ion deposition and runoff in forest watersheds on the northern side of Lake Biwa

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To clarify the sources of sulfur ions in precipitation and runoff from forest watersheds, bulk deposition and stream water samples were collected within the small Kutsuki and Surumi forest watersheds on the northern side of the Lake Biwa basin, central Japan. Samples were analyzed for major ions and  $\delta^{34}\text{S}$ . Continuous monitoring from 1990 to 2010 showed that the average  $\text{SO}_4^{2-}$  concentration in stream water samples was  $1.62 \pm 0.31$  (0.76, 3.58)  $\text{mg l}^{-1}$  at Kutsuki and  $6.59 \pm 1.54$  (3.68, 16.1)  $\text{mg l}^{-1}$  at Surumi (t-test,  $p < 0.01$ ). However, the average  $\text{SO}_4^{2-}$  concentration in bulk deposition samples was similar in both watersheds:  $2.15 \pm 1.31$  (SD)  $\text{mg l}^{-1}$  (range, 0.202-10.2  $\text{mg l}^{-1}$ ) at Kutsuki and  $2.24 \pm 1.29$   $\text{mg l}^{-1}$  (0.350-6.07  $\text{mg l}^{-1}$ ) at Surumi (t-test,  $p > 0.05$ ). The  $\delta^{34}\text{S}$  values in bulk deposition samples fluctuated from +8.86 to +9.14 ‰ at Kutsuki and from +9.87 to +11.1 ‰ at Surumi, whereas non-sea salt (nss-)  $\delta^{34}\text{S}$  in stream water samples varied from +6.89 to +12.0 ‰ and from +4.64 to +5.11 ‰, respectively (t-test,  $p < 0.01$ ). It is said that the  $\delta^{34}\text{S}$  values in coal products from northern China varied from -3 to -1 ‰ and Japanese oil varied from +5 to +18 ‰.

Our findings suggest that the difference in the ability of canopies in the watersheds to catch  $\text{SO}_4^{2-}$  dry deposition is the reason for the significant difference in nss-  $\delta^{34}\text{S}$  values in stream water samples. The more open canopy in the Kutsuki watershed consists of young conifers and deciduous broadleaf trees, whereas the canopy in the Surumi watershed consists of mature conifer trees at a high density. Therefore, it appears that the Kutsuki watershed was only affected by  $\text{SO}_4^{2-}$  deposition from the Asian continent. There were not any domestic air pollution sources on the north side of Kutsuki. In contrast, the dense canopy of the Surumi watershed was affected by local  $\text{SO}_4^{2-}$  pollution sources of national roads which were located 1 km northwest from Kutsuki, rather than continental sources. Overall, this investigation suggests that the difference in stream water  $\text{SO}_4^{2-}$  concentrations in both watersheds is caused by the canopy differences.

## Geochemical and isotope systematics of Asahi and Yoshii rivers

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We have undertaken a detailed geochemical and isotopic analysis of water samples collected from the Asahi and Yoshii rivers of the Okayama Prefecture, Japan. More than 300 samples were collected from the rivers and their tributaries during March 2011 to January 2014. In several locations, samples were collected periodically in order to monitor the long-term fluctuation of the geochemical properties. All samples were filtered with 0.2  $\mu\text{m}$  filter prior to the analyses for major dissolved constituents (F, Cl, NO<sub>3</sub>, SO<sub>4</sub>, Br, PO<sub>4</sub>, Ca, Mg, Na, K), trace elements and O-H-Sr-S isotopes.

Based on the results obtained so far, we have constructed a high-resolution geochemical map of the rivers covering central to eastern Okayama Prefecture. The map shows that the concentrations of most major dissolved constituents, as well as the O-H-Sr isotope ratios changes systematically from the upstream towards the downstream. For example, the deuterium excess (DE) is generally high in the upstream ( $>20$ ) and gradually decreases towards the downstream ( $<12$ ). This difference in the DE exceeds the seasonal variation observed in locations where the long-term monitoring was conducted. Thus, the regional change in the DE is interpreted to be the result of different air mass contributing to the meteoric water of different locations.

The Ca, Sr, and Ba concentrations are generally low in the upstream and gradually increase towards the downstream. The <sup>87</sup>Sr/<sup>86</sup>Sr also changes from approximately  $\sim 0.705$  and  $\sim 0.706$  in the headwaters of the Asahi and Yoshii rivers, respectively, to  $\sim 0.708$  in the areas close to the Seto inland sea. The shift in the Sr isotope ratio seems to correlate well with the change in the ages of the rocks exposed in the river basin. This implies that these variations are likely related to the water-rock interactions.

The concentrations of other important dissolved constituents such as the NO<sub>3</sub> and SO<sub>4</sub> tend to increase abruptly in the densely populated regions. This is also accompanied by a shift in the S isotopic composition. These variations may reflect a change in the degree of human influence such as breakdown of fertilizers used for agricultural activities.

Keywords: Geochemical map, Asahi River, Yoshii River

## Chracteristics of groundwater discharge around western foot of Mt.Chokai

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To clarify the total discharge rate of the submarine spring from Mt. Chokai, base-flow observation was conducted on 15 streams around western foot of Mt. Chokai at the end of January 2014. In this presentation, based on the distribution of specific discharge rate, we discuss the characteristics of the groundwater discharge and the hydrological balance in this area.

Keywords: Mt.Chokai, submarine spring, base-flow, hydrological balance, groundwater age

## Estimation of groundwater recharge area at the south foot of Mt. Bandai using the observation data in 2013

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The Mt. Bandai (1,816 m a.s.l.) which is an active volcano of the Quaternary period is located at Fukushima prefecture. Because of the geological characteristics in the volcanic area, it is estimated the groundwater recharge is large around Mt. Bandai. The groundwater and spring water around Mt. Bandai is used as the source of the public tap water, so it is important to comprehend the groundwater flow and groundwater recharge ratio at Mt. Bandai. To make clear the groundwater flow systems at Mt. Bandai, the investigation was carried out.

The EC value is under 10 mS/m and water quality indicates the Ca-HCO<sub>3</sub> type at most points in the south slope of Mt. Bandai. The oxygen isotopic ratios ( $\delta^{18}\text{O}$ ) are -11.1 to -10.8 ‰ (Site 1) and -11.2 to -11.1 ‰ (Site 2), and hydrogen isotopic ratios ( $\delta\text{D}$ ) are -67 to -65 ‰ (Site 1) and -68 to -67 ‰ (Site 2). The altitude effect is recognized in these areas. The average recharge area (altitude) in the south slope at Mt. Bandai is estimated from 1,150 to 1,270 m (Site 1) and from 1,360 to 1,420 m (Site 2). These recharge area (altitude) in the south slope is lower rather than the north and west slopes. In future, the investigation at more low altitude area (about 520 to 550 m) will be carried out, and estimate the groundwater flow in the south slope at Mt. Bandai.

Keywords: Mt. Bandai, recharge area, spring water, water quality, stable isotopes