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# Relationship between nitrate in river waters and land use in a hilly and mountainous area: stable isotope-based analyses

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Declining quality of waters arises from an imbalance of the inflow of pollutant loads such as nutrient versus the self-purification of waters. For urbanized areas where the population density is high, the sewage effluent can be a dominant source contributing to the increased nitrate concentration in rivers and often leads to the increased eutrophic levels in downstream waters. Also, for hilly and mountainous areas where the population is declining, the nitrate concentration in rivers can be increasing every year even though its cause remains unclear. One such example is Hii River, which is the largest river flowing into Lake Shinji and Lake Nakaumi in Shimane prefecture. The nitrate concentration in the Hii River tends to be increasing every year despite depopulation, an increased percentage sewered population and improved sewage disposal in the basin. That could cause algal blooms in Lake Shinji and Lake Nakaumi and thus immediate countermeasures are required.

Land use within a river basin is one of the most important factors influencing the nitrate concentration in the river water. However, little information is available about how land use affects river nitrate concentration in hilly and mountainous areas. This may be because the nitrate concentration varies within a relatively narrow range and thus appears to differ little among river basins with different land uses. Also, there is not always a constant relationship between the nitrate concentration and percentage of land use in the river basin because the nitrate concentration varies according to river discharge. In this study, to examine the effects of land use on river nitrate concentration in a hilly and mountainous area, river water samples were collected at the end of nine subbasins in the Hii River basin and concentrations and isotopic compositions of nitrate were analyzed. We are going to report those results.

Keywords: diffuse pollution, forest, hydrological condition, agricultural land, enclosed waters



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# Understand seasonal variation of the stalagmite growth rate with the hydrochemical parameters of dripwater

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Carbon dioxide ( $CO_2$ ) degassing is given for a main factor to control the stalagmite growth recently. The seasonal variation (high p $CO_2$  in summer and low p $CO_2$  in winter) of the CO<sub>2</sub> partial pressure (p $CO_2$ ) of the cave air by ventilation of the cave air which occurred with a temperature difference between the cave outside and the cave air was observed. Because a change of this p $CO_2$  changes easiness (difference (dp $CO_2$ ) cave air p $CO_2$  and dripwater equilibrium p $CO_2$ ) of the CO<sub>2</sub> degassing, the seasonal variation of the stalagmite growth rate is supposed (Spotl et al., 2005; Baldini et al., 2008). In addition, there is the study (Genty et al., 2001) that was going to see the cyclic variation of the stalagmite growth rate by the theory equation found from an annual growth thickness and chemical kinetics of the dripwater, and stalagmite growth rate increase is supposed regardless of a season when the Ca<sup>2+</sup> concentration of dripwater is high in this study. The common understanding of the mechanism of the short-term variation of the stalagmite growth rate is not provided in this way. Therefore we built technique to estimate the stalagmite growth rate from the hydrochemical parameter of the dripwater and observed a stalagmite growth rate (Calcite deposition rate) monthly in Inazumi limestone cave in mie-machi, bungo-ono-shi, Oita-ken, Japan. As a result, we report that we got the seasonal variation and the control factor of the growth rate.

As Spotl et al. (2005) and Baldini et al. (2008) guessed, the stalagmite growth rate of the Inazumi limestone cave showed a slow tendency in the summertime and fast tendency in the wintertime, but we got that growth became slow in winter by taking it finely. Genty et al. (2001) insisted that the  $Ca^{2+}$  concentration of dripwater controlled stalagmite growth, but there was not the correlation between  $Ca^{2+}$  concentration and the stalagmite growth rate, and this observation result did not support the claim. Many researchers believe (e.g. Spotl et al. (2005) and Baldini et al. (2008)) that a difference between cave air pCO<sub>2</sub> and dripwater equilibrium pCO<sub>2</sub> controls stalagmite growth but the variation is not seasonal variation. Furthermore, dripwater dissolves a stalagmite grows up even at the time of plus pCO<sub>2</sub> value of the summertime. The observation result of the Inazumi limestone cave does not support the claim of Spotl et al. (2005) and Baldini et al. (2008). The control factor of the stalagmite growth has calcite saturation index (CSI) and dripwater quantity besides dpCO<sub>2</sub>. Because CSI value of the dripwater always shows pluses (supersaturation for calcite), stalagmite is going to always grow up. Furthermore, the variation of CSI resembles the variation pattern of the stalagmite growth rate closely. However, the variation of the stalagmite growth rate in the Inazumi limestone cave cannot explain a variation of the winter only by CSI. The fall of this growth rate is caused by the decrease of the dripwater quantity, and this observation result support a claim of Genty et al. (2001) that the dripwater quantity controls the stalagmite growth most when the dripwater quantity is very few.

A key result is that a stalagmite grew up because dripwater is always a supersaturation condition, and the seasonal variation of the stalagmite growth rate is controlled by the seasonal variation of CSI. In the Inazumi limestone cave, there is the peculiarity that quantity of the dripping water decreases in winter and tends to depend on dripping quantity of water than CSI in the time. We are conscious of contribution to the study of the paleoclimate reconstruction that used the striped pattern to appear in the growth direction of the stalagmite and want to clarify the reason of seasonal variation of CSI of the dripwater and the variation of the drip rate of the dripwater in future.

Keywords: Stalagmite growth rate, Hydrochemical parameters of dripwater, Seasonal variation of growth rate



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### Altitude effect of precipitation samples at Kusatsu area, Gunma Prefecture

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The stable isotopes of oxygen and hydrogen in precipitation are formed with complex factors, i.e. precipitation amount, elevation, altitude and air mass, and the change pattern of isotopes is different in various sites. There is a lot of study about isotopes in precipitation, but many of them target the isotopes at one-slope. Thus the investigation of the isotopes in precipitation at the mountainous area which has a complex topography is few, there are uncertain points for the mechanism of isotopes in precipitation. The isotopes in precipitation are useful tool to understand the recharge area of spring water and groundwater. In this study, the study site is Kusatsu area which is located to northwest at Gunma Prefecture and spread out the mountainous region. The objective of this study is to male clear the characteristics of isotopes in precipitation around Kusatsu area.

An annual mean air temperature is 7.4 degree and annual precipitation amount is about 1700 mm at Kusatsu area. The precipitation sampler is settled at 6 points with 3 different slopes. The monthly precipitation samples have been sampled since March 2010. The pH, EC, water quality and stable isotopes of oxygen and hydrogen were analyzed for all precipitation samples.

The stable isotopes of oxygen and hydrogen in precipitation show a similar change. The isotope values are relatively high in June and relatively low in August 2010. Thus the isotope values are relatively low with the high elevation and relatively high with the low elevation, the altitude effect is confirmed in this area. The d-excess values are high at the high elevation and low at the low elevation. And there is the obvious seasonal variation of d-excess in precipitation; that the d-excess are relatively low in summer period and relatively high in winter period. It is cause of the air mass which is source of the precipitation. The altitude effect using the data of 6 observation sites is -0.26 per mill / 100 m for  $d^{18}$ O (r<sup>2</sup>=0.980) and -1.9 per mill / 100 m for dD (r<sup>2</sup>=0.988). The altitude effect is slightly different on three slopes and also different according to the month. It is assumed that the reason of the difference of altitude effect is cause of the difference of precipitation amount and transportation process of the water vapor which is source of the precipitation. The local meteoric water line in Kusatsu is  $dD = 8.0d^{18}O + 9.6$ , which is almost same the Craig's meteoric water line. In future, the observation and sampling of the precipitation will be continued, and the characteristics of isotopes in precipitation will be clarified in detail.

Keywords: Kusatsu, precipitation, stable isotope, altitude effect



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# Elevation effects for hydrogen and oxygen isotope values of rainwater within the Kofu Basin

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To elucidate the factor controlling elevation effects in hydrogen and oxygen isotope values, 150 rain water samples were collected from 6 points in different altitude from 250m to 1300m of 25 rainfall events. The water oxygen and hydrogen isotope values were analyzed with these samples. And the rainfall feature in each event was investigated by the observation of the X-band multi-parameter radar of University of Yamanashi.

The elevation effects were not well understood on windward side. Hence, these results were different from the previously reports of Friedman and Smith (1970). Elevation effects are well defined on the rainfall amount. Hence, to clarify the factor of elevation effects, it must identify the distribution of rainfall amount. In this presentation, we examine the relationship between rainfall amount and the rainfall distribution in rain clouds advected to the Kofu Basin.

Keywords: water hydrogen and oxygen isotope, elevation effects, rainfall, X-band multi-parameter radar



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### Stable isotopic composition of rainwater and soil water of Kathmandu Valley, Nepal

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The present study examined the hydrogen and oxygen stable isotopic composition of rainwater and soil water and of Kathmandu Valley, Nepal.Ten stations were selected for rainwater sampling and two sites were selected for soil water samplings. Bulk rainwater samples were collected in biweekly basis whereas soil water samples were collected in a weekly basis during monsoon season (June - September) of 2010. Soil water samples were collected from 4 different depths (50, 70, 120 and 150cm) using tension lysimeter. Both the rainwater samples and soil water samples were analyzed water oxygen and hydrogen isotope values using Cavity Ring-Down Spectroscoyy (Picarro, L1102-1).

The preliminary results of stable isotopic composition are used to construct the Local Meteoric Water Line (LMWL), and found to be delta D (permil) = 8.17 delta180 + 10.8 (r2= 0.98). This meteoric line was then compared with the Global Meteoric Water Line (GMWL), where the slope and intercept of LMWL are close to that of GMWL ((i.e. delta D (permil) = 8delta180+10) as described by Craig (1961).

The isotopic composition of soil water varies according the depth of soil. In upper surface (50cm and 70cm depth), most of the soil water samples contain lighter delta180 composition where the heavier values are observed in the greater depth (120 and 150cm). However some of the samples showed the similar isotopic composition of in all depths.

The variations and differences in isotopic composition of rainwater and soil water of Kathmandu Valley could provide the estimates of evaporation, infiltration processes which are very useful for the groundwater management perspective.

Reference

Craig (1961)

Keywords: rainwater, soil water, stable isotopes, Kathmandu



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## Isotopic composition of throughfall in coniferous forest plantation

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A few studies reported the changes of isotopic composition of rainwater during its passage through the vegetation canopy (Brodersen et al., 2000; Liu et al, 2008). The selection processes, the forest canopy structure and resulting evaporation processes seemed to be the main factors influencing the isotopic composition of open rainfall while passing the canopy, however the effect of rainfall redistribution by the vegetation canopy on the isotopic composition of rainfall has often been neglected. In this study, we conducted field measurement of throughfall using 10m long and 10m wide interception plots and a set of 20 tipping bucket rain gauges and throughfall collectors for isotopic analysis in Japanese cypress and Japanese cedar plantations.

Keywords: Coniferous forest plantation, Throughfall, Isotopic composition



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### Recharge and flow processes of groundwater on the outer rim slope of Hakone caldera

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The purpose of this study is to understand recharge and flow processes of groundwater on the slope of outer rim of Hakone caldera, central Japan from using environmental tracer methods. 99 stream and 38 spring water samples were collected from study area in middle of August to early September 2010, and analyzed for oxygen isotopic composition ( $d^{18}O$ ).

The stream and spring waters in the western slope of study area have higher  $d^{18}O$  value compared with that in other slopes as well as precipitation data presented by Miyashita (2009). The isotopic difference between western and other slopes are probably caused by a rain shadow effect of mountain, because the dominant wind directions during summer period, when intensive groundwater recharge occurs, is from SW to NE. In other words the precipitation of northeastern slope, which is leeward side, is more isotopically depleted than that of western slope, which is windward side. A comparison of spring water samples at the same elevation (about 1000m a.s.l., near the top of outer rim) indicate that  $d^{18}O$  of those on the leeward northeastern slope are more isotopically depleted and about 1.0 per mil less than those from the windward west slope.

Therefore, it is necessary to evaluate the altitude effect of water samples in each slope (west, southeast and northeast), dividing from the spatial distribution patter of  $d^{18}O$ . Based on the relationship between  $d^{18}O$  values and mean elevations of catchment for several selected springs, recharge-water lines were obtained in each slope. Altitude effect of recharge water in each slope was different: -0.2 per mil/100m for the western slope, -0.18 per mil/100m for the southeastern slope and -0.14 per mil/100m for the northeastern slope. CFCs (chlorofluorocarbon) and SF6 (sulfur hexafluoride) have been used to determine the average residence time for two spring waters, located on the middle (360m a.s.l.) and lower (45m a.s.l.) part of the northeastern slope. Consequently, the residence times of both spring waters were < 20 years (probably about 10 years). On the basis of mean recharge elevation obtained by recharge-water line and distribution pattern of springs and result of the CFCs and SF6 datings, conceptual model of groundwater flow system in the outer rim slope of Hakone caldera are proposed.

Miyashita (2009) reported that the  $d^{18}O$  of stream and spring waters was about 2 per mil higher than weighted mean  $d^{18}O$  of precipitation at the same elevation. It has also been found that the difference in  $d^{18}O$  between recharge water and precipitation was 1.5~2.0 per mil in this study. The difference may be responsible for the evaporation of precipitation in the recharge process, and we calculated annual evaporation rate using the Rayleigh-type equation. The evaporation rates are about 15 percent of the annual precipitation in each slope.

Keywords: Hakone volcano, Oxygen isotope composition, Rain shadow effect, Altitude effect, Groundwater flow system, Evaporation rate



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Groundwater flow system of the Yiluo River Basin from the tritium and stable isotopes ratios

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Groundwater flow system and the water quality in Yiluo River Basin, China # Pei Zhao[1] [1] Graduate School, Univ.Rissho

The author has carried out the analysis of tritium and stable isotopes of surface water and groundwater to evaluate the groundwater flow system in the Yiluo River basin, China.

1) Tritium concentrations of the groundwater in the slope of the southern and northwestern mountain range were higher than 10 T.U. and suggested that the residence time of these groundwater was younger than 50 years. On the other hand, tritium concentrations of the deep groundwater along the Yiluo River and in the lower area of the basin were very low. Therefore, the residence time were estimated to be longer than 60 years.

2) The stable isotopic ratios of hydrogen and oxygen of the groundwater showed relatively low value in the southern peripheral part of the basin and showed relatively high value in the central part. In the eastern part, down-stream part, and the southeastern part of the basin, groundwater with low dD and  $d^{18}$ O flows toward the lower area of the basin.

Keywords: tritium concentration, stable isotopic ratios, groundwater flow system, The Yiluo River Basin



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# Water quality, environmental isotopes and subsurface temperature of high Cl groundwater area in the northern Kanto plain

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Previous studies point out that there are some areas of groundwater with high  $Cl^-$  concentration (dozens to about 200 mg/l) in the inland part of Kanto plain that is the largest sedimentary plain in Japan (i.e. Ikeda, 1984; METI, 1975). As for the high  $Cl^$ groundwater area in the central part of the plain, from the view point of isotopic characteristics, residence time of the groundwater is probably longer than the groundwater in the surroundings (i.e. Yasuhara et al., 2008; Hayashi et al., 2003). However, origin of  $Cl^-$  in this area has not been revealed yet. With regard to the high  $Cl^-$  groundwater areas in the northern part and northeastern part of the plain, Miyakoshi et al. (2003) estimates that subsurface temperature in these areas are higher than the surroundings. However, information about groundwater quality, environmental isotopes and relation of distributions between the high  $Cl^-$  groundwater and subsurface temperature are quite limited. Therefore, we collected groundwater samples from these two areas and measured major dissolved ions and environmental isotopes. Also, we measured subsurface temperature profiles to reveal subsurface temperature distribution.

For  $Cl^-$  concentration, the highest value was 538 mg/l in the northern area and was 221 mg/l in the northeastern area. However, in the northeastern area, only one sample showed high  $Cl^-$  concentration higher than 20 mg/l. For delta<sup>13</sup>C, these two areas showed relatively high values than surroundings: -8.5 to 0.3 permil in northern area and -7.3 to 2.0 permil in northeastern area. However, correlations between  $Cl^-$  concentration and delta<sup>13</sup>C were not clear. As for the subsurface temperature distribution, subsurface temperatures of the two areas were higher than the surroundings. Especially, the northern area was one of the highest temperature areas in the Kanto plain. In a larger sense, distribution patterns of  $Cl^-$ , delta<sup>13</sup>C and subsurface temperature were consistent.

Keywords: kanto plain, high Cl groundwater, environmental isotopes, subsurface temperature



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An isotopic study on the origins of water and chloride ion in artesian groundwater of the Kanto plain, central Japan

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There exist three regions in the Kanto plain, central Japan, whose artesian groundwater is characterized by a high  $Cl^-$  concentration: 1) central parts of the Kanto plain (Saitama Prefecture), 2) floodplains and deluvial uplands along the lower reaches of Kokai and Tone rivers (Ibaraki and Chiba Prefectures), and 3) south-east parts of Gunnma Prefecture). An isotopic study has been under way to make it clear the origins of both water and  $Cl^-$  of these  $Cl^-$ -rich artesian groundwaters. As for that in the central parts of the Kanto plain, confined groundwater with a high  $Cl^-$  concentration of up to 216 mg/l is obtained from the productive bores of 200-430 m depth. The area of  $Cl^-$ -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 faults on its longer sides. Taking all isotopic data (delta-D, delta-<sup>18</sup>O, <sup>13</sup>C, <sup>14</sup>C, <sup>4</sup>He, <sup>36</sup>Cl/Cl) obtained so far into account, with regard to the origin of groundwater in the Motoarakawa tectonic zone, a potential source is assumed to be precipitation of low stable isotopic composition in the Last Glacial Maximum when the sea level of the Tokyo Bay was lower than the present by more than 100 m. Admixture of sea water in the period of and/or after the Shimosueyoshi transgression (peak period at around 125,000 yrs. BP) is likely to account for its elevated  $Cl^-$  concentration. The Ayasegawa and Kuki faults, and other unknown faults could act as a geologic barrier to the modern regional groundwater flow system, preventing mixing of groundwater between in and out of the tectonic zone. The results from the similar isotopic study now in progress on the other two  $Cl^-$ -rich groundwater regions will help an overall understanding of the long-term groundwater system development in the Kanto plain in these 500,000 yrs.

Keywords: Kanto plain, artesian groundwater, chloride ion, delta-180 & delta-D, 14C, 36Cl/Cl ratio



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A multiple-isotope apporach to reveal the coastal hydrogeological system and its temporal changes.

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Multiple-isotope data and the results of the diffusion-sedimentation model were used to reveal the coastal hydrogeological system and its temporal change at a small catchment and its offshore extension, the Uto Peninsula, Kumamoto, Japan. Chloride concentration decreases gradually downwards, and the profile of the stable chlorine isotopic ratio showed the typical pattern formed by diffusion-controlled mass transport process. One-dimensional diffusion/sedimentation model explained the measured chloride profile and the fractionation of chlorine isotopes sufficiently well. The apparent residence time of groundwater below inter-tidal zone is on the order of 100 years while that below sea bottom is about 2000 years, suggesting that groundwater situated below the inter-tidal zone constitutes a part of present-day groundwater flow system while the fresh groundwater below sea bottom was separated from the present-day flow system. The coincidence among the apparent residence time of groundwater below sea-bottom, the age of the start of the deposition of marine clay, and the age of the start of the diffusion process strongly suggests that the deposition of marine clay controlled the hydrogeological system and resulted in the reduction of the extent of the groundwater discharge.



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## Occurrence and formation mechanism of Harazuru hot spring, Fukuoka, Japan

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Harazuru hot spring is located to the eastern part of Chikugo Plain in Fukuoka-ken, Kyushu, Japan, where is just beside of the Chikugo River, the longest river in Kyushu. There two types of hot springs, chemically; One is Na-HCO3 type, and the other Na-Cl/HCO3. The former is located in the central part of Harazuru extending to the NE-SW direction, and the latter distributing to the northwestern and southeastern part of the former type. The delta D of the Na-HCO3 and Na-Cl/HCO3 types are delta D =-49 to -51 per mill and -54 to-57 per mill, respectively. The delta D of the Chikugo River shows -50 per mill. Such chemical relations suggests Na-HCO3 type of water is formed by mixing of Na-Cl/HCO3 with meteoric water. However, the delta D is so small compared to that of Chikugo-gawa river nearby, the water is probably migrated through a big geological structures extending EW direction from the upper part of the Chikuko-gawa river.

Keywords: hot spring, chemistry, idotope, formation mechanism, Harazuru, Fukuoka



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# Origin of dissolved inorganic carbon of hot spring waters discharged from the non-volcanic region of central Kyusyu

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For elucidation of carbon sources of dissolved inorganic carbon (DIC) in the hot spring waters discharged from the nonvolcanic region between Aso and Kirishima volcanoes of Kyusyu District, Japan, and also in order to search a deep-seated aqueous fluid derived from subducting oceanic plate in the region, we analyzed major chemical components, dD and  $d^{18}O$  of water and  $d^{13}C$  and concentration of DIC, rare gas isotope concentrations of dissolved gases of the hot spring waters. Although water of every hot spring is originated from meteoric water shown by water isotopic data (dD and  $d^{18}$ O), relationships between d<sup>13</sup>C and concentration of DIC suggests that the DIC should be formed by mixing of soil and two kinds of deep-originated CO<sub>2</sub>. This idea is supported by isotopic date of rare gases (<sup>3</sup>He/<sup>4</sup>He vs. <sup>4</sup>He/<sup>20</sup>Ne) and relation between concentrations of Ca and  $HCO_3$  ions, and it is confirmed that the two kinds of deep-originated  $CO_2$  are mantle-derived  $CO_2$  and  $CO_2$  originated from subducted marine carbonate by a calculation of contributions of source carbons of selected hot spring waters. Moreover, we calculated respective contribution ratios of deep-originated CO2 to DIC of all the hot spring waters on the basis of the linear relation observed between contribution ratios of deep-originated CO<sub>2</sub> and d<sup>13</sup>C values of DIC of selected hot spring waters, and expressed hot springs showing high contribution ratios (more55percent) on a published map showing crustal resistivity structure of this studied area, thereby it appears that hot springs rich in DIC derived from the subducted marine carbonate are roughly concentrated on low electrical resistivity zone extending NE direction from Kirishima volcano. This result seems to suggest the possibility that an associated aqueous fluid of dehydrated fluid from subducting oceanic plate forms the low electrical resistivity zone. On the other hand, distributions of high contributions of mantle-originated CO<sub>2</sub> of hot spring waters are concentrated in the Hitoyoshi Basin which is thought to be a tectonic basin formed by fault movement, and this result may suggest that a passageway for rising of mantle-derived CO<sub>2</sub> must be formed in the crust under this area.

Keywords: non-volcanic region, dissolved inorganic carbon, mantle, deep-originated  $CO_2$ , low electrical resistivity zone, dehydrated fluid from subducting plate

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### Distribution of the helium isotope ratios in Kyusyu district

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Kyusyu island is located at the intersection of the SW Japan arc and the Ryukyu arc. The Philippine Sea plate is subducting beneath the Eurasian plate at the Ryukyu trench and the Nankai Trough along these arcs. The subduction angle of the Ryukyu trench is larger than that of the Nankai Trough. The former angle is almost orthogonal and the subduction of the Philippine Sea plate is seismically detected to the depth of 150-180 km (Nakajima and Hasegawa, 2007). The Okinawa Trough is to the west of the Ryukyu trench and extends as a back-arc basin of the Ryukyu arc. The Beppu-Shimabara Graben in central Kyusyu is regarded as an extension of Okinawa Trough. Strong low-velocity anomalies are distributed extensively along the volcanic front and extend to the back-arc side in the crust and upper mantle. Volcanisms are supposed to result from the fluid supplied by the dehydration processes of the descending Philippine Sea plate. In addition, the hot upwelling materials related to the back-arc opening have contributed to the Unzen volcanism in Beppu-Shimabara Graben (Wang and Zhao, 2006). Thus two different mechanisms for volcanism exist in north Kyusyu.

In this study, we measured the helium isotope ratios ( ${}^{3}\text{He}/{}^{4}\text{He}$  ratios) of hot springs around the area of the prefectural boundary of south Fukuoka and north Kumamoto in order to study the precise geographical distribution of helium isotope ratios in this region and to compare them with tectonic data. This area covers the Beppu-Shimabara Graben and its northern and southern areas. We collected 13 samples of hot spring waters in the above area. High  ${}^{3}\text{He}/{}^{4}\text{He}$  ratios were observed at the Beppu-Shimabara Graben, and low  ${}^{3}\text{He}/{}^{4}\text{He}$  ratios were observed in the northern and southern area of the Beppu-Shimabara Graben. It is very peculiar since the observed area belongs to the back-arc region where  ${}^{3}\text{He}/{}^{4}\text{He}$  ratios are generally higher than the atmospheric value as commonly seen in NE Japan. We indicated that the high  ${}^{3}\text{He}/{}^{4}\text{He}$  ratios simply reflected a high velocity region of about 25-30 km in depth under the sampling region (Xia et al., 2008), and was not due to the addition of fossil pore water drawn from impermeable marine clay layers as suggested by Mahara and Kitaoka (2009). Thus,  ${}^{3}\text{He}/{}^{4}\text{He}$  ratios could be closely related with the presence of deep fluid at the basement of the crust.

Keywords: helium isotope ratio, Kyusyu, Beppu-Shimabara Graben



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# Application of <sup>36</sup>Cl to deep fluid systems in Japan: Implications for the sources and residence time of chlorine

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This study applied the long-lived radionuclide <sup>36</sup>Cl to better elucidate the sources of chlorine in deep fluids in Japan. Several regions with different tectonic/geological settings were selected for the collection of deep fluid samples from hot spring wells: e.g., a coastal sedimentary basin in Aomori, surroundings of volcanic calderas in Hokkaido, and vicinity of tectonic faults in western Japan. Concerning the samples obtained from a coastal sedimentary basin, the <sup>36</sup>Cl/Cl ratios mostly fall on the seawater-shallow groundwater mixing trend line, with a few samples deviating upward possibly due to the build-up of nucleogenic <sup>36</sup>Cl in the subsurface. The calculated <sup>36</sup>Cl/Cl ratios of assumed seawater fractions were positively correlated with crustal <sup>4</sup>He concentrations, associated with increasing residence time of the fluids in the subsurface. This trend suggests that the source of deep fluids in this area is probably old seawater. In the case of the samples nearby major tectonic faults, the delta <sup>18</sup>O-delta D relationship depicts a shift to Arima-type thermal brine (Matsubaya et al., 1973) or magmatic water (Giggenbach, 1992). These samples tend to show low <sup>36</sup>Cl/Cl ratios close to the seawater value (1-2 x 10<sup>-15</sup>) especially for the samples with high <sup>3</sup>He/<sup>4</sup>He ratios similar to that of the upper mantle. It implies a deep-seated source of these fluids, such as mantle- or magma-derived components, and also suggests a relatively short residence time in the crust without significant production of nucleogenic <sup>36</sup>Cl.

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Keywords: deep fluid, chlorine, origin, residence time, chlorine-36