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AHW24-01

Room:301A

Time:May 25 09:00-09:30

# Microbial methane production and denitrification in deep aquifer associated with the accretionary prisms

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To understand microbial potential of degradation of organic compounds, fermentation, methanogenesis, and denitrification in deep aquifer associated with the accretionary prism, the deep groundwater and natural gas samples were collected from 14 deep wells that were drilled to the aquifer. We performed a series of geochemical and microbiological analyses of the samples. Consequently, methane was the predominant component of the natural gas (51.4 to 99.4%). On the other hand, the natural gas samples containing  $N_2$  (0.6 to 48.6%) were found in some samples. Stable carbon isotopic analysis suggested that the methane was derived from both biogenic processes and thermogenic reaction. Phylogenetic analysis targeting bacterial 16S rRNA genes indicated the dominance of  $H_2$ -producing fermentative bacteria in the groundwater. In addition to the fermentative bacteria, 16S rRNA genes related to denitrifying bacterium were identified at the sites where  $N_2$  was detected from the natural gas. Phylogenetic analysis targeting archaeal 16S rRNA genes revealed the dominance of hydrogenotrophic methanogens in the groundwater. Anaerobic incubations using the groundwater amended with organic substrates indicated  $H_2$  accumulation and rapid methane production. These results suggested that methane in the deep aquifer is produced by a syntrophic consortium of  $H_2$ -producing fermentative bacteria and hydrogenotrophic methanogens in a wide range of deep aquifer associated with the accretionary prism in Southwest Japan. In addition to methane production, microbial denitrification using methane or organic matters as electron donors seems to be present in the deep aquifer associated with the accretionary prisms.

Keywords: accretionary prisms, groundwater, subsurface microorganisms, methane production, denitrification

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# Atmospheric circulation controls on the inter-annual variability in precipitation isotope ratio in Japan

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This study explored the primary driver of variations of precipitation isotopes at multiple temporal scales (event, seasonal and inter-annual scales) to provide a greater depth of interpretation for isotope data in Japan. Using a new one-year record of the isotopic composition of event-based precipitation and continuous near-surface water vapor at Nagoya in central Japan, we identify the key atmospheric processes controlling the storm-to-storm isotopic variations through an analysis of air mass sources and rain-out history during transport of moisture to the site, and then apply the identified processes to explain the inter-annual isotopic variability in the historical 17-year long Tokyo station record in the Global Network of Isotopes in Precipitation (GNIP).

An event-based one-year record of HDO in precipitation at Nagoya in Japan showed less seasonal variations, but there is large variability in HDO on a storm-to-storm basis. In summer, southerly flows transported moisture with relatively higher HDO from subtropical marine regions, and the warm rainfall type was relatively enriched in heavy isotopes compared with the other rainfall events. In contrast, low HDO were observed when northerly winds brought relatively cold air to the observation site. Some of the observed isotopic variability can be explained by changes in air mass sources, however this is not enough to have a large storm-to-storm isotopic range. The additional source of variability is attributed to rainfall amounts occurring both at the site and prior to the site. A clear decreasing trend in HDO with cumulative rainfall over nine-hour back trajectories demonstrates that rainout history plays a dominant role on the storm-to-storm isotopic variability in the summer. The more isotopically depleted precipitation is from large-scale weather systems accompanied by prolonged rainfall over wide areas. In winter, low HDO occurred when a cold frontal rainband associated with extra-tropical cyclones (Nangan cyclones) passed south of the Japan coast. Easterly or northeasterly winds north of the cyclone transport relatively cold air from the mid- or high-latitude regions to the site, and feed the cold frontal rainband. Therefore, the precipitation related to the Nangan cyclone is characterized by relatively lower isotopic values than those from another type of cyclone. It follows that the occurrence of Nangan cyclones is the most likely contributor to changes in winter mean precipitation HDO.

Using the historical record of monthly isotopes in precipitation at GNIP Tokyo station, we explored if factors controlling storm-to-storm isotopic variability can account for inter-annual isotopic variability. The 17-year variation of summer precipitation  $H_2^{18}O$  was independent of the variation in regional-scale summer precipitation, which is a substitute for cumulative rainfall along the trajectories. On the other hand, year-to-year variation of  $H_2^{18}O$  related closely to changes in air mass sources. The relatively higher  $H_2^{18}O$  in summer precipitation corresponded to the higher contribution of warm rainfall to the total summer precipitation, whereas the inter-annual variation of winter precipitation  $H_2^{18}O$  correlated to the relative ratio of the rainfall from Nangan cyclones to the total winter precipitation. The activity and storm track of intensified Nangan cyclones were responsible for increasing the contribution of cold frontal rainfall fed by northerly winds, and for further decreasing  $H_2^{18}O$  in winter precipitation. These indicate that inter-annual isotopic variability in winter and summer precipitation in the central Japan is primarily related to changes in meridional moisture transport due to the distinctive difference in isotopic composition between low- and high-latitude moisture.

Keywords: Stable water isotopes, East Asian monsoon, Baiu, Southern coastal cyclone

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AHW24-03

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#### Stable isotopes in precipitation all over Japan observed in 2013

ICHIYANAGI, Kimpei<sup>1\*</sup>; TANOUE, Masahiro<sup>2</sup>; YOSHIKUNI, Tsukasa<sup>1</sup>

Stable isotopes in precipitation (Oxygen-18 and Deuterium) are useful natural tracers for understanding the hydrological cycle and reconstructing paleo-climates. There are a lot of observational studies of stable isotopes in precipitation in Japan. However, most observations on the local scale were conducted only one point or less than several points. The Isotope Mapping Working Group of the Japanese Association of Hydrological Sciences (JAHS-IMWG) conducted the intensive observation of stable isotopes in precipitation across Japan throughout 2013 (IOP2013). In this study, seasonal variation and spatial distribution of Oxygen-18 and d-excess at 57 stations across Japan were shown from the preliminary result of the IOP2013. Annual mean values in Oxygen-18 show the strong altitude effect from -13permil in the north part to -6permil in the southwest part of Japan. The Oxygen-18 values along the Pacific coast and the Japan Sea coast of Japan are more and less than -8permil, respectively. Annual mean values in d-excess ranged from 7permil in the southwest part to 22permil in the northeast part of Japan. The d-excess values in the north part and along the Japan Sea coast are relatively higher than those along the Pacific coast of Japan. The differences in d-excess values between the Pacific coast and the Japan Sea coast are mentioned in the previous studies. Most of the monthly Oxygen-18 values ranged from -15permil to -5permil observed in the north part of Japan, and those values were relatively high in the spring months (March-June) and low in the winter months (December to February). Most of the monthly Oxygen-18 values ranged from -10permil to -5permil in the southwest part of Japan, the seasonal variation is small. On the other hand, the monthly d-excess values show strong seasonal variations observed both in the north and southwest parts of Japan. Those values ranged from Opermil to 40permil in the north part and from Opermil to 30permil in the southwest part of Japan. Especially in the north part of Japan, monthly d-excess values were extremely high in the winter month (December to February). The high d-excess values are caused by the strong evaporation from the Japan Sea in the winter months.

Keywords: stable isotope, precipitation, d-excess, all over Japan, Isotope Mapping Working Group

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AHW24-04

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# Comparison of stable precipitation isotopes between regional isotope simulation and intensive observation around Japan

TANOUE, Masahiro<sup>1\*</sup>; ICHIYANAGI, Kimpei<sup>2</sup>

Stable isotopes in precipitation have been used as tracer of hydrological cycle. However, there are few or no observation studies throughout Japan at the same time. Here, we organized Isotope Mapping Working Group (IMWG), and conducted the intensive observations for the period from January to December in 2013 at 124 sites. These observational data with high spatial resolution are vulnerable on a global scale. In this study, the observational data was compared with isotopic compositions derived from isotope regional circulation model. By comparing spatial distribution of isotopic compositions of precipitation, it was seen that the model captured not only the latitude effect (values decrease with increasing latitude), but also the intensity of the effect around Japan, except for June to September. The spatial distributions from June to September were uniform around Japan. Then, in order to compare seasonal variation of isotopic compositions of precipitation between simulation and observation, we calculated regional averaged seasonal variations according to regional partition of Japan Meteorological Agency. As a result, the model captured observed seasonal variation, such as high values from March to July in the northern part of Japan. However, model overestimated isotopic compositions in January in the western part and the eastern part of Japan. The overestimation was due to an intensity of the low pressure system in the south coast of Japan around 14 January 2013, because the simulated isotopic compositions by the low pressure system were higher compared with observed that. We will introduce the result of comparisons.

Keywords: stable isotopes in precipitation, isotope regional circulation model

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### A two-year record of stable isotope characteristics of monthly rainfall at the Douala and Yaounde urban cities, Cameroon

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The stable isotopes of oxygen (<sup>18</sup>O) and hydrogen (<sup>2</sup>H) in precipitation are useful tools in environmental studies including hydrological and climatological investigations. Like in most of tropical Africa, stable isotope data of rainfall is regrettably limited in Cameroon. As a contribution to desired data, 43 monthly rainfall samples have been collected from January 2013 to December 2014 in the urban cities of Douala and Yaounde (in the tropical evergreen forest of Cameroon). The objectives were to produce local meteoric water lines (LMWLs), define the spatial and temporal variations of the stable isotopes and controlling factors. The conventional delta ( $\delta$ ) <sup>18</sup>O- $\delta$ <sup>2</sup>H diagram for the two-year data gave the regression lines:  $\delta$ <sup>2</sup>H= 7.92 $\delta$ <sup>18</sup>O + 12.99 (R<sup>2</sup>=0.97) and  $\delta^2 H = 8.35\delta^{18}O + 15.29$  (R<sup>2</sup>= 0.99) for Douala and Yaounde, respectively. These lines represent the LMWLs for the two cities. The similarity of slopes to the Global Meteoric Water Line (GMWL) of 8 indicates that rain formation processes in both areas occurred under conditions close to isotopic equilibrium with insignificant evaporation effect during precipitation. Douala precipitation showed a wide range of  $\delta^{18}$ O from -5.22 to -0.75 %, mean of -2.71 % (2013); and -5.26 to -1.28 %, average of -3.09 % (2014). A similar range was observed in Yaounde for  $\delta^{18}$ O from -5.20 to 1.81 %, mean of -2.49 % (2013); and -5.86 to -0.66 %, average of -3.37 % (2014). The large range suggests varied controls on precipitation in both localities. Despite the closeness of the Douala sampling point to the Atlantic Ocean (35 km), the weighted mean d-excess value of 13.12 % was higher relative to 10 % of the Atlantic moisture. Further inland in Yaounde (191 km), a relatively higher weighted mean d-excess value (14.55 %) was also observed. The high d-excess values in both cities reflect an addition of recycled continental moisture to precipitation. The additional moisture is likely from the evergreen rainforest and a network of rivers in the areas. Weighted mean  $\delta^{18}$ O and  $\delta^{2}$ H values for the sampling period were -3.27 ‰ and -13.01 ‰, and -3.07 ‰ and -10.04 ‰ in Douala and Yaounde, respectively. The slight increase in weighted mean  $\delta$  values from Douala to inland Yaounde reflects a lack of continental effect probably due to the additional supply of inland recycled moisture to rain. Monthly weighted mean  $\delta$  values showed a definite seasonal variability in both areas. Isotopically enriched and depleted values were observed during the pre- and post-monsoon low rainfall and heavy monsoon rains, respectively, in agreement with the amount effect of tropical low latitude rains. This effect is probably controlled by northward and southward oscillation of the Intertropical Convergent Zone and associated air masses. The  $\delta^{18}$ O and  $\delta^{2}$ H of rainfall in the tropical cities of Douala and Yaounde show a similar temporal variability that is mainly controlled by the addition of inland recycled moisture and amount effect. The generated isotope data and LMWLs can be used as tools for groundwater recharge and atmospheric moisture circulation studies in the region.

Keywords: Stable isotope variation, Local meteoric water line, Amount effect, Moisture recycling, Douala-Yaounde, Cameroon

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AHW24-06 Room:301A Time:May 25 11:00-11:15

#### Saline groundwaters in and around the Osaka Basin and those origins

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Many saline waters including high amounts of Fe and occasionally CO<sub>2</sub> and bicarbonate ions are issuing in the mountainous areas surrounding Osaka Basin. Among those waters, Arima hotspring is the most famous and the hot saline water has been called Arima-type brine, which is characterized by heavy oxygen isotope shift and high 3He/4He ratio similar to the mantle derived component (e. g., Nagao et al. 1981). High CO<sub>2</sub> and <sup>3</sup>He/<sup>4</sup>He saline waters similar to the Arima-type brine have been known at Ishibotoke at southern mountainous area of Osaka (e. g., Matsumoto et al., 2003). Saline groundwaters were found in the Quaternary sediments and underlying basement rocks of Osaka Basin. Those waters occasionally contained high <sup>3</sup>He/<sup>4</sup>He ratios and were suggested the similarity to the Arima-type brine (Morikawa et al., 2008). In this study, saline waters containing >500 mg/L chloride ions and >400 mg/L HCO<sub>3</sub><sup>-</sup> ions were three-dimensionally mapped assuming the well depth was the sampling depth of groundwater to see the relationship between the occurrence of saline waters and geological structure. Then, the origins of saline waters were estimated from the relationships among hydrogen and oxygen isotope ratios and chloride ion concentrations.

The saline waters distribute characteristically along the boundary between sedimentary basin and surrounding mountains and the bottom of the basin including the lowermost sedimentary formation and basement rocks. The former is usually along active faults; Arima-Takatsuki Tectonic Line at the north and Ikoma faults system at the east and south. These faults would work as recharging paths of deep groundwater and seawater might inflow using these faults. The latter does not have clear relationship to the tectonic structure.

Hydrogen and oxygen isotope ratios of saline waters are plotted on a mixing line of seawater and local meteoric water. If seawater is one of the end members, relationships of chloride concentration and those isotope ratios also give mixing lines. Although these relationships of saline waters at <100 m depths show the mixing lines, the those of deeper ones do not give simple mixing lines; chloride concentration and oxygen isotope ratio of saline waters at 100-500 m depths are on the mixing line while hydrogen isotope ratios are smaller than those affected by seawater, and the saline waters >500 m depth have smaller isotope ratios than those contributed by seawater. Compared with the Arima-type brine, oxygen shift is not large for these saline waters, however, such an isotope characteristics would be on the formation process of Arima-type brine.

Keywords: Deep groundwater, Arima-type brine, active faults, hydrogen and oxygen isotope ratios

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AHW24-07

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On deep-seated salty groundwater mixed into shallow groundwater flow system in Sagae district, Yamagata, Japan

YASUHARA, Masaya<sup>1\*</sup>; MORIKAWA, Noritoshi<sup>1</sup>; INAMURA, Akihiko<sup>1</sup>; TAKAHASHI, Hiroshi<sup>1</sup>

<sup>1</sup>Geological Survey of Japan, AIST

On deep-seated salty groundwater mixed into shallow groundwater flow system in Sagae district, Yamagata, Japan

Keywords: deep-seated groundwater, salty water, shallow groundwater flow sytem, mixing ratio, isotopes

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AHW24-08

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#### Chemical properties of deep groundwater at coastal area

 $MACHIDA, Isao^{1*}; ONO, Masahiko^1; KOSHIGAI, Satoru^1; IKAWA, Reo^1; MARUI, Atsunao^1$ 

Two drilling surveys up to a depth of 150m have been carried out close to shoreline at Suruga bay, Shizuoka Prefecture. The properties of groundwater flow can be discussed by the chemical and isotopical data from geological core.

Keywords: Borling survey, Pore water, Coastal Area, Boundary of saline and fresh water, Isotopes, Water quality

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Vertical profiles of stable isotopic composition of groundwater in Abukuma granite: comparison with radiocarbon dates

TAKAHASHI, Hiroshi $^{1*}$ ; TAKAHASHI, Masaaki $^{1}$ ; TSUKAMOTO, Hitoshi $^{1}$ ; YASUHARA, Masaya $^{1}$ ; KAZAHAYA, Kohei $^{1}$ ; INAMURA, Akihiko $^{1}$ 

Groundwater in hard rock aquifer mainly circulates along fractures and fissures, and the flow is mainly governed by the physical properties of the rock heterogeneities such as opening, spacing and connectivity. The present study reported stable isotopic compositions of groundwater in granite fractures. The study site is situated at the northern part of the Abukuma Mountains in Fukushima Prefecture, northeastern Japan. To collect groundwater in granite fractures, three drillings were performed. Groundwater sample in the present study were taken from fractures in the weathered-fissured zone and fracture zone of granite. The radiocarbon dates of groundwater ranged from modern to ~16 ka. The vertical profiles of radiocarbon dates indicate a relatively constant age of 10-16 ka for groundwater deeper than 100 m.

Stable isotopic composition suggests that all groundwater originated from meteoric water. The shallow around-groundwaters indicated wide isotopic range, since the monsoon climate will produce the seasonal isotopic variations of precipitation. The range of stable isotopic compositions of stream waters around Mt. Utsushiga-take showed slightly lower than that of around-groundwater. This suggested that around-groundwater is unlikely originated from recharge area in Mt. Utsushiga-take, although the local groundwater flow was not clarified. Groundwater flow from the mountain to the drilling sites might be blocked off, owing to the faults and tectonic line between Mt. Utsushiga-take and the drilling sites.

The isotopic values of borehole-groundwater beneath the depth of 100 m showed significantly lower values than those of around-groundwaters: the isotopic depletions are 5-10 ‰ in hydrogen isotopic ratio and 1.5-2.0 ‰ in oxygen isotopic ratio. These indicate that they were recharged under colder climate conditions and/or at higher altitudes. The stable isotopic compositions of stream water of the Mt. Utsushiga-take did not represent the same ranges with those of borehole-groundwater. Radiocarbon dates of groundwater beneath the depths of 100 m ranged 10-16 ka, corresponding to be the last glacial period, Oldest, Older and Younger Dryas periods. The colder climate enhances the isotopic fractionation during the air mass transportation and rainfall. Some studies reported the low values of isotopic compositions of groundwater due to recharge under the clod climate such as last glacial period.

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AHW24-10

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Time:May 25 12:00-12:15

#### Isotopic characteristics of acid springs in Shunomata river basin in Mt. Chokai volcano

HAYASHI, Takeshi<sup>1\*</sup>; YASUHARA, Masaya<sup>2</sup>; MORIKAWA, Noritoshi<sup>2</sup>; TAKAHASHI, Hiroshi<sup>2</sup>; ASAI, Kazuyoshi<sup>3</sup>; KUSANO, Yukiko<sup>4</sup>

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There are some rivers that have low pH and deposition of Iron oxides on the east side of Mt. Chokai volcano. Shunomata river basin has some limonite ore deposits on the river bed of tributaries (EL: approximately 900m). These ore deposits are considered to have made by acid springs that are characterized by low pH (about 3 to 5) and relatively high temperature (about 20 to 25 degree Celsius). Also, previous studies pointed out that pH of the acid springs have decreased after a volcanic activity of Mt. Chokai in 1974. However, there are quite few studies on the origin and groundwater quality evolution process of the acid springs. Therefore, this study aims to clarify these points. We will show the results of measurements of major dissolved ions and environmental isotopes.

Temperature and pH of the acid springs showed 15.5 to 19.5 degree Celsius and 2.8 to 3.0, respectively. As for major dissolved ions, groundwater quality was characterized by high concentrations of SO42- and Cl (117 to 181 mg/L and 66 to 106 mg/L). Comparing these results with that of "Detsubo" spring that is located on the north side of Mt. Chokai volcano and is famous for low temperature (about 7 degree Celsius) and low pH (about 4.5), groundwater quality was largely different. Stable isotopic ratios of oxygen and hydrogen were about -11.5 per-mil and -68 to -69 per-mil, respectively. Elevation of recharge area of the acid springs was estimated more than 1,600 masl.

The results of this study suggest that there are some groundwater quality evolution systems of acid springs in Mt. Chokai volcano.

Keywords: acid springs, environmental isotopes, limonite ore deposits, Mt. Chokai Volcano

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AHW24-11 Room:301A

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## Groundwater age determination by using <sup>85</sup>Kr and its verification by other hydrogeochemical tracers

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Krypton 85 (<sup>85</sup>Kr) is a man-made trace gas from reprocessing plant origin whose atmospheric concentrations have been increasing over the past few decades. As it is soluble in water, it can be used as groundwater age indicators over timescales ranging from a few years to a few decades. In this study, <sup>85</sup>Kr specific activities in groundwater were measured with an on-site dissolved Kr gas extraction system using an external flow through type hollow fiber membrane modified after Ohta et al. (2009).

 $^{85}$ Kr specific activities in groundwater were confirmed at 3 sites in Miyakonojo basin, south-western Japan, considering regional groundwater flow system. Estimated groundwater age were 2 years, 11 years and 60 years in the recharge, intermediate and stagnant discharge areas along the groundwater flow line, respectively. In order to verify these  $^{85}$ Kr ages, we also measured other age tracer gases such as Sulfur hexafluoride (SF<sub>6</sub>) and Chlorofluorocarbons (CFCs) at the same sampling wells of  $^{85}$ Kr measurement. The result of the SF<sub>6</sub> age dating in the three locations were well harmonized with the  $^{85}$ Kr dating results; the SF<sub>6</sub> age were 1 year, 23 years and over 60 years. However, CFCs could not show reasonable groundwater age due to the local contamination by the urban and industrial origin CFCs.

The seasonal fluctuation of the stable isotopes ( $\delta^{18}O$  and  $\delta D$ ) in groundwater were also measured to evaluate the comprehensive groundwater age tendency. Relatively high seasonal fluctuation of the stable isotopes were measured only in the shallow unconfined well site at the recharge area, which reflect the seasonal isotopic fluctuation in the precipitation. This is another evidence of the relatively young groundwater characteristics to support the  $^{85}Kr$  and  $SF_6$  age in the recharge area.

Keywords: Krypton-85, Groundwater age, Groundwater flow system, Sulfur hexafluoride, Groundwater age tracer, Miyakonojo basin

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