Characteristics of hydrogen and oxygen stable isotope ratios in precipitation collected in a snowfall region, Aomori Prefecture, Japan

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Characteristics of δ2H and δ18O in precipitation samples collected at 16- to 17-d intervals at three stations at the northern end of Honshu Island, Japan, were investigated. Rokkasho and Ajigasawa stations are approximately 100 km apart, on the Pacific Ocean coast and the Sea of Japan coast, respectively, of Aomori Prefecture. Hakkoda station is between Rokkasho and Ajigasawa stations at 1334 m ASL. Precipitation samples were continuously collected during 2000–2011 at Rokkasho, and during 2003–2006 at Ajigasawa and Hakkoda. At all stations, δ2H and δ18O of the precipitation samples showed weak seasonality, with higher values in summer and lower values in winter. In the summer (June, July, and August), the intercept of the regression line between δ2H and δ18O, that is, the local meteoric water line (LMWL), was lower than that of the global meteoric water line (GMWL) with similar slope, but in winter (December, January, and February), the LMWL intercept was higher than the GMWL intercept with similar slope. Temporal variations of δ18O at the two coastal stations were similar, although the timing of precipitation events was different. At all stations, d-excess values showed clear seasonality, with high values in winter and low values in summer, indicating a seasonal change in the source of the water vapor in air masses arriving over northeast Japan. In winter, d-excess values were higher at Hakkoda than at the other stations, suggesting a precipitation at Hakkoda was derived from water vapor from a remote area of the Sea of Japan. The long-term precipitation data set for δ2H and δ18O obtained in this study will be useful for investigations of regional hydrology and validation of numerical models.

Keywords: hydrogen isotope ratio, oxygen isotope ratio, d-excess, seasonal variation, Aomori Prefecture

INTRODUCTION

Stable isotope ratios of hydrogen (2H/1H) and oxygen (18O/16O) in atmospheric water vapor and precipitation vary spatially and temporally because of equilibrium and kinetic isotopic fractionations associated with condensation and evaporation. Investigations of isotopic ratios of water have significantly contributed to our understanding of atmospheric circulation (e.g., Epstein and Mayeda, 1953; Fritz et al., 1987; Hoffman et al., 2000). Moreover, many hydrological studies have investigated 2H/1H and 18O/16O ratios in precipitation in relation to surface water (Gonfiantini, 1985; Telmer and Veizer, 2000; Gibson and Edwards, 2002), groundwater (Gat, 1981; Yonge et al., 1989), and their interaction.

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Mizota and Kusakabe (1994) mapped hydrogen and oxygen isotopic data for surface and shallow groundwater in Japan and the surrounding area using their own measurement data and data available in the literature, and others have examined the hydrogen and oxygen isotopic data of local meteoric waters in relation to the regional hydrological system of central Japan (e.g., Kusakabe et al., 1970). Data from northern Japan are relatively scarce, however; only a few data covering relatively short time periods have been reported from Aomori Prefecture, northern Honshu Island (Aoki, 1992; Toyama et al., 2010, 2011). No long-term data set is available for analysis of atmospheric circulation and the hydrological system in this region.

Therefore, the objectives of this study were (1) to document the characteristics of hydrogen and oxygen isotopic ratios in precipitation in Aomori Prefecture; (2) to estimate the important factors controlling water isotopes; (3) and thus to establish for this region a long-term data set of isotopic ratios suitable for hydrologic studies.

**MATERIALS AND METHOD**

**Study sites**

Situated at the northern end of Honshu Island, Aomori Prefecture is surrounded by ocean on three sides. The Ou Mountains, which are located in the center of the prefecture, divide it into two parts, creating a complicated topography of inland and coastal features and, consequently, highly varying climatic conditions in different areas of the prefecture (Fig. 1).

Under typical winter conditions, a cold, wet air mass from over the Sea of Japan collides with the Ou Mountains, bringing heavy snowfall to the Sea of Japan side of the prefecture. However, the mountains shield the southeastern part of the prefecture on the Pacific Ocean side, with the result that many winter days in an area within several tens of kilometers of Hachinohe city are sunny and dry (Nibe, 1989). In summer, cool, humid easterly winds (known as Yamase) from the Okhotsk high cause many days on the Pacific Ocean side of the prefecture to be characterized by low temperatures and high humidity.

Precipitation samples were collected at three stations in Aomori Prefecture (Fig. 1). Rokkasho station is on the rooftop of the Institute for Environmental Sciences building (40°57'56" N, 141°21'38" E, 30 m ASL) in Rokkasho Village, approximately 2 km from the Pacific Ocean on the east and 12 km from Mutsu Bay on the west. Rokkasho Village experiences rather low temperatures in summer (Pacific Ocean side type summer) and heavy snowfall in winter (Sea of Japan side type winter). Ajigasawa station is at the summit of Mt. Tamoyachidake (40°40'33" N, 140°51'33" E, 1334 m ASL) in the Hakkoda Mountains, the northward extension of the Ou Mountains.

**Sample collection and analytical methods**

At Rokkasho and Ajigasawa stations, precipitation samples were collected with a wet and dry deposition sampler (US-750, Ogasawara Keiki Seisakusho Co. Ltd., Tokyo, Japan), which has an opening of 0.0314 m² and collects wet and dry deposition separately by means of an automatically moving lid. In winter, the collection area...
Hydrogen and oxygen isotope ratios in precipitation, Aomori 11

of the sampler is kept at 5–10°C by a circulating antifreeze solution that also melts any deposited snow. At Hakkoda station, precipitation samples were collected in summer by a total deposition sampler consisting of a polyethylene tank connected to a polyethylene funnel with an opening of 0.0798 m². In winter, snow samples were collected in an open polyethylene bag set in a vinyl chloride pipe with an opening of 0.0798 m².

At the three stations, samples were collected at a mean interval of 16–17 d (range, 9–28 d). Samples were collected around 09:00 local time (LT) on each collection day at both Rokkasho and Hakkoda stations, and at around 13:00 LT at Ajigasawa station. At Rokkasho station, samples were collected continuously from October 2000 to December 2011. At Ajigasawa and Hakkoda stations, continuous sampling was carried out from November 2003 to March 2006. Each collected sample was weighed and then passed through a filter with a pore size of 0.45 µm.

Hydrogen isotope ratios were measured using H₂ gas generated by reduction of the sample water with zinc metal at 490°C in a sealed quartz tube (Coleman et al., 1982). For determination of the oxygen isotope ratio, the water sample was equilibrated with CO₂ at 25°C in a plastic syringe (Yoshida and Mizutani, 1986), and then the isotope ratio of the CO₂ was measured. A dual-inlet stable isotope mass spectrometer (Optima, VG Isotech, UK, or MAT252, Thermo Quest, USA) was used to measure both ratios.

The conventional delta notation is used to express the stable isotope ratios of hydrogen (δ²H) and oxygen (δ¹⁸O) in the water samples: \( \delta = \left( \frac{R_{\text{sample}}}{R_{\text{standard}}} - 1 \right) \times 1000 \), where \( R_{\text{sample}} \) and \( R_{\text{standard}} \) denote the ²H/¹H or ¹⁸O/¹⁶O ratios in the sample and the standard material, respectively. Vienna Standard Mean Ocean Water (VSMOW) distributed by the IAEA (Vienna, Austria) was used as the standard for both ratios. The measurement system was calibrated by using IAEA reference materials, VSMOW, Greenland Ice Sheet Precipitation, and Standard Light Antarctic Precipitation (SLAP). Values of –428‰ and –55.5‰ were used for \( \delta²H \) and \( \delta¹⁸O \) of SLAP, respectively, following the guidelines of Coplen (1996). The estimated reproducibilities (1σ), which were evaluated from routine measurements of the laboratory standard water, were within 1.0‰ for \( \delta²H \) and within 0.1‰ for \( \delta¹⁸O \). Reported monthly averaged \( \delta²H \) and \( \delta¹⁸O \) in this paper are means of the measured isotope ratios weighted by the proportion of the total monthly precipitation amount.

RESULTS AND DISCUSSION

Seasonal variation of \( \delta²H \) and \( \delta¹⁸O \) in precipitation

The observed \( \delta²H \) and \( \delta¹⁸O \) in precipitation at the three stations are shown in Figs. 2–4. At Rokkasho station during 2000–2011, \( \delta²H \) of all precipitation samples ranged

![Fig. 2. Time series of Rokkasho station measurement results for October 2000 to December 2011: (a) \( \delta²H \) and \( \delta¹⁸O \); (b) monthly precipitation amount (bars) and mean temperature (line).](image-url)
from $-91.1\%$ to $-15.4\%$, and $\delta^{18}O$ values varied from $-13.7\%$ to $-4.0\%$ (Fig. 2). Mean $\delta^2H$ and $\delta^{18}O$, weighted by precipitation, were $-56.2\%$ and $-8.9\%$, respectively ($n = 132$). During 2004–2005 ($n = 24$), when sampling was also being conducted at Ajigasawa and Hakkoda stations, the weighted means of $\delta^2H$ and $\delta^{18}O$ were $-52.5\%$ and $-8.6\%$, respectively.

At Ajigasawa station during 2003–2006, $\delta^2H$ and $\delta^{18}O$ in the precipitation samples ranged from $-70.2\%$ to $-25.0\%$ and from $-10.9\%$ to $-4.5\%$, respectively (Fig. 3). The weighted means during 2004–2005 were $-52.3\%$ for $\delta^2H$ and $-8.6\%$ for $\delta^{18}O$ ($n = 24$). Thus, they were almost equal to the means at Rokkasho station during 2004–2005.

At Hakkoda station, which is at the highest elevation (1334 m ASL; corresponding to the lower part of the free atmosphere) among the three stations, $\delta^2H$ and $\delta^{18}O$ in the precipitation samples varied from $-90.0\%$ to $-35.2\%$ and from $-14.0\%$ to $-6.9\%$, respectively, during 2004–2005 (Fig. 4). In this study, the minimum values of both $\delta^2H$ and $\delta^{18}O$ were observed at this station. Weighted means of all data at Hakkoda station were $-64.3\%$ and $-10.3\%$ for $\delta^2H$ and $\delta^{18}O$ ($n = 24$), respectively. These means are clearly lower than those at Rokkasho and Ajigasawa stations, but they are consistent with the $\delta^2H$ and $\delta^{18}O$ values of shallow groundwater in this region of Japan (Mizota and Kusakabe, 1994).

In general, the $\delta^2H$ and $\delta^{18}O$ values in precipitation at the three stations do not show clear seasonal changes. The $\delta^2H$ and $\delta^{18}O$ values show only weak seasonal variation, being generally higher in summer and lower in winter. Araguas-Araguas et al. (1998) reported similar weak seasonality in $\delta^2H$ and $\delta^{18}O$ of precipitation at Tokyo and Ryori, Japan, and at Pohang, South Korea.

At Rokkasho station, however, 11-year monthly averaged $\delta^{18}O$ values show clear seasonality, with higher values in summer and lower values in winter (Fig. 5).

Relation between $\delta^2H$ and $\delta^{18}O$, and deuterium excess in precipitation

Craig (1961) reported that the stable isotope ratios of hydrogen and oxygen in global precipitation were related as follows: $\delta^2H = 8 \times \delta^{18}O + 10$. This relationship was later defined as the Global Meteoric Water Line (GMWL).
The relationship between $\delta^2$H and $\delta^{18}$O at each station was compared with the GMWL (Fig. 6). The slope of the regression line for summer (June, July, and August; $\delta^2$H = $7.5 \times \delta^{18}$O + 4.2, r = 0.98) at Rokkasho station was similar to the GMWL slope, but its intercept was lower than the GMWL intercept. The slope of the regression line for winter (December, January, and February; $\delta^2$H = $7.9 \times \delta^{18}$O + 22.6, r = 0.96) was also similar to the GMWL slope, but its intercept was higher. The $\delta^2$H and $\delta^{18}$O values in precipitation collected during spring (March, April, and May) and autumn (September, October, and November) were distributed between the summer and winter regression lines. The results obtained for Ajigasawa and Hakkoda stations are essentially in agreement with the Rokkasho station results.

The different intercepts can be explained by reference to the deuterium excess (d-excess) concept, where d-excess = $\delta^2$H – 8 $\times \delta^{18}$O (Dansgaard, 1964). The d-excess value has been used as a tracer of conditions affecting evaporation in oceanic moisture source regions, such as sea surface temperature and the relative humidity of the overlying air mass (Merlivat and Jouzel, 1979). Several studies have reported that d-excess values in precipitation in Japan vary seasonally (e.g., Waseda and Nakai, 1983; Satake et al., 1984; Uemura et al., 2012), and Uemura et al. (2008) have shown that d-excess values of water vapor above the ocean surface correlate negatively with the relative humidity above the ocean and positively with sea surface temperature. In general, higher d-excess values indicate lower relative humidity in the oceanic moisture source region. In this study, the d-excess values of precipitation show clear seasonal variation at all three stations, with lower values in summer and higher values in winter (Fig. 7).
Fig. 7. Temporal variation of monthly averaged (a) d-excess, (b) $\delta^{18}O$ in precipitation, and (c) monthly precipitation amount at Rokkasho (solid line), Ajigasawa (dashed line), and Hakkoda (chain line) during November 2003 to March 2006.

Relation to local meteorological parameters

At mid-latitudes, both temperature and the rainfall amount are important factors controlling the final isotopic compositions of precipitation (Dansgaard, 1964; Lawrence and White, 1991). The slopes of the linear regression lines between $\delta^{18}O$ and surface air temperature, the monthly precipitation amount, and the logarithm of monthly precipitation (Log P) at each station are shown in Table 1. $\delta^{18}O$ and surface air temperature showed a weak positive correlation at Rokkasho and Ajigasawa stations, but at Hakkoda station, the correlation was not significant ($p > 0.05$). Monthly precipitation amounts did not show a clear correlation with $\delta^{18}O$ at any of the three stations. When we compared the slopes of the regression lines between $\delta^{18}O$ and surface air temperature, the monthly precipitation amount and the logarithm of monthly precipitation (Log P) among seasons at Rokkasho station (Table 2), however, we found a positive correlation of $\delta^{18}O$ with air temperature in spring, and a negative correlation of $\delta^{18}O$ with the two precipitation parameters in winter.

Johnson and Ingram (2004) applied a multiple regression model to the analysis of spatial and temporal variations of stable isotopes in modern precipitation in China, as follows:

$$\delta^{18}O = \beta_0 + \beta_T T + \beta_{\log P} \log P$$

where $\beta_0$ is the $y$-intercept, and $\beta_T$ ($‰/°C$) and $\beta_{\log P}$ ($‰/\log \text{mm}$) are the partial regression coefficients for temperature and the logarithm of precipitation, respectively. We applied this multiple regression model to the data at the three stations (Table 3). As a result, at Rokkasho station, we obtained a regression equation ($R^2 = 0.37$) with $\beta_T = 0.14$ and $\beta_{\log P} = -2.64$. These partial regression coefficients are similar to those at Yantai, on the coast of northeastern China ($\beta_T = 0.11$, $\beta_{\log P} = -3.26$), where the $\delta^{18}O$ time series is controlled by both air temperature and precipitation amount (Johnson and Ingram, 2004). The $\delta^{18}O$ data at the other two stations, however, were not significantly correlated with air temperature or monthly precipitation amount. The higher coefficient of determination ($R^2 = 0.37$) of the multiple regression analysis compared with the linear regression analysis ($R^2 = 0.03–0.21$) suggests that $\delta^{18}O$ in precipitation at Rokkasho station may be controlled by both local meteorological factors (temperature and precipitation amount). To assess the seasonality of both $\beta_T$ and $\beta_{\log P}$, we applied the multiple regression model to the $\delta^{18}O$ data of each season at Rokkasho station (Table 4). We found significant correlations in three seasons. In spring and autumn, $\delta^{18}O$ was positively correlated with air temperature and negatively correlated with $\log P$. These correlations are consistent with well-known effects (temperature effect, amount effect; e.g., Dansgaard, 1964). In summer, $\delta^{18}O$ did not show any clear relation with air temperature or precipitation amount, but in winter, the results of the model analysis show that $\delta^{18}O$ was negatively correlated with both $T$ and $\log P$. The liquid-vapor equilibrium isotope fractionation factor increases with decreasing tempera-
Hydrogen and oxygen isotope ratios in precipitation, Aomori

If the condensation temperature is assumed to be correlated with the surface air temperature, condensed precipitation should be isotopically lighter under the lower air temperature condition (Dansgaard, 1964). The observed negative correlation between air temperature and $\delta^{18}O$ is contrary to this expectation. According to our observations in this study, monthly precipitation and the monthly averaged air temperature were positively correlated in winter. There is a possibility that precipitation amount is more effective factor controlling $\delta^{18}O$ of winter precipitation at Rokkasho station. However, the relationships between $\delta^{18}O$ and local air temperature or monthly precipitation amounts are not very clear. In addition, the expected isotope variation of approximately 2‰ (based on the observed summer-winter air temperature difference of 22°C at Rokkasho station), calculated from the temperature dependency of equilibrium fractionation (0.1‰/°C; e.g., Majoube, 1971), was less than the observed summer-winter variation of $\delta^{18}O$ (10‰) at Rokkasho station. These facts suggest that the important factor controlling monthly averaged $\delta^{18}O$ in precipitation was synoptic-scale vapor circulation rather than local meteorological parameters.

Relation to air mass pathways

Figure 7 shows time series of monthly averaged d-excess, $\delta^{18}O$ in precipitation, and monthly precipitation amounts at the three stations from November 2004 to March 2006. At Hakkoda station, d-excess values tended to be higher and $\delta^{18}O$ in precipitation tended to be lower than at the other two stations. Even though Rokkasho and Ajigasawa stations are 100 km apart and precipitation events were not synchronous between them, temporal changes in $\delta^{18}O$ in precipitation and the d-excess value were remarkably similar at these two stations. As shown in Figs. 2–4, the relationships between $\delta^{18}O$ in precipitation and local air temperature or monthly precipitation amount were not very clear. In addition, the expected isotope variation of approximately 2‰ (based on the observed summer-winter air temperature difference of 22°C at Rokkasho station), calculated from the temperature dependency of equilibrium fractionation (0.1‰/°C; e.g., Majoube, 1971), was less than the observed summer-winter variation of $\delta^{18}O$ (10‰) at Rokkasho station. These facts suggest that the important factor controlling monthly averaged $\delta^{18}O$ in precipitation was synoptic-scale vapor circulation rather than local meteorological parameters.

Table 1. Slopes of regression lines between $\delta^{18}O$ and air temperature (T), precipitation amount (P), and logarithm of the precipitation amount (logP) during the study period

<table>
<thead>
<tr>
<th>Station</th>
<th>n</th>
<th>$\beta_T$</th>
<th>$\beta_P$</th>
<th>$R^2$</th>
<th>p</th>
<th>$\logP$</th>
<th>$R^2$</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rokkasho</td>
<td>132</td>
<td>0.10</td>
<td>0.21</td>
<td>0.00</td>
<td>-0.005</td>
<td>0.03</td>
<td>0.03</td>
<td>-1.40</td>
</tr>
<tr>
<td>Ajigasawa</td>
<td>29</td>
<td>0.07</td>
<td>0.15</td>
<td>0.04</td>
<td>-0.006</td>
<td>0.04</td>
<td>0.29</td>
<td>-0.81</td>
</tr>
<tr>
<td>Hakkoda</td>
<td>29</td>
<td>0.06</td>
<td>0.10</td>
<td>0.09</td>
<td>0.006</td>
<td>0.00</td>
<td>0.78</td>
<td>0.77</td>
</tr>
</tbody>
</table>

Table 2. Slopes of regression lines between $\delta^{18}O$ and air temperature (T), precipitation amount (P), and logarithm of precipitation amount (logP) during each season at Rokkasho station

<table>
<thead>
<tr>
<th>Season</th>
<th>n</th>
<th>$\beta_T$</th>
<th>$\beta_P$</th>
<th>$R^2$</th>
<th>p</th>
<th>$\logP$</th>
<th>$R^2$</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spring</td>
<td>33</td>
<td>0.22</td>
<td>0.31</td>
<td>0.00</td>
<td>-0.01</td>
<td>0.09</td>
<td>0.09</td>
<td>-2.13</td>
</tr>
<tr>
<td>Summer</td>
<td>33</td>
<td>0.08</td>
<td>0.02</td>
<td>0.41</td>
<td>-0.006</td>
<td>0.08</td>
<td>0.10</td>
<td>-1.96</td>
</tr>
<tr>
<td>Autumn</td>
<td>34</td>
<td>0.12</td>
<td>0.10</td>
<td>0.07</td>
<td>-0.009</td>
<td>0.15</td>
<td>0.03</td>
<td>-2.45</td>
</tr>
<tr>
<td>Winter</td>
<td>32</td>
<td>-0.28</td>
<td>0.13</td>
<td>0.04</td>
<td>-0.02</td>
<td>0.35</td>
<td>0.00</td>
<td>-2.89</td>
</tr>
</tbody>
</table>

Table 3. Summary of the multiple regression analysis results for the three stations

<table>
<thead>
<tr>
<th>Station</th>
<th>n</th>
<th>$\beta_T$</th>
<th>$\beta_P$</th>
<th>$R^2$</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rokkasho</td>
<td>132</td>
<td>0.14</td>
<td>-2.64</td>
<td>0.37</td>
<td>0.00</td>
</tr>
<tr>
<td>Ajigasawa</td>
<td>29</td>
<td>0.06</td>
<td>-0.94</td>
<td>0.17</td>
<td>0.09</td>
</tr>
<tr>
<td>Hakkoda</td>
<td>29</td>
<td>0.06</td>
<td>-0.31</td>
<td>0.10</td>
<td>0.24</td>
</tr>
</tbody>
</table>

Table 4. Summary of the multiple regression analysis results for each season at Rokkasho station

<table>
<thead>
<tr>
<th>Season</th>
<th>n</th>
<th>$\beta_T$</th>
<th>$\beta_P$</th>
<th>$R^2$</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spring</td>
<td>33</td>
<td>0.25</td>
<td>-2.36</td>
<td>0.50</td>
<td>0.00</td>
</tr>
<tr>
<td>Summer</td>
<td>33</td>
<td>0.08</td>
<td>-1.94</td>
<td>0.11</td>
<td>0.17</td>
</tr>
<tr>
<td>Autumn</td>
<td>34</td>
<td>0.15</td>
<td>-2.79</td>
<td>0.28</td>
<td>0.01</td>
</tr>
<tr>
<td>Winter</td>
<td>32</td>
<td>-0.11</td>
<td>-2.61</td>
<td>0.37</td>
<td>0.00</td>
</tr>
</tbody>
</table>

Hydrogen and oxygen isotope ratios in precipitation, Aomori
Fig. 8. Results of the backward air mass trajectory analysis for summer (left panels) and winter (right panels) at the three stations: (a) Rokkasho, (b) Ajigasawa, and (c) Hakkoda.

demonstrated a relationship between air mass origins and isotopic signals (e.g., He et al., 2006; Pfahl and Wernli, 2008). Therefore, we calculated 72-h backward air mass trajectories starting at the altitude of 500 m (1500 m at Hakkoda) using the NOAA HYSPLIT model (Draaxler and Rolph, 2013; Rolph, 2013) for the three stations in summer (August 2005) and winter (December 2004) (Fig. 8). In winter, most trajectories passed over the Sea of Japan,
whereas in summer, they did not pass primarily over any particular area. In winter, the pathways by which the air masses arrived at the two coastal stations did not differ very much, and they transited east coast of Russia. The pathway of the air masses arrived at Hakkoda station transited through the northeast of Korean Peninsula. In summer, however, though the pathways of air masses arriving at Rokkasho and Ajigasawa were almost the same, the pathways of those arriving at Hakkoda frequently crossed over the Asian continent.

Yoshimura and Ichiyanagi (2009) used an atmospheric general circulation model that incorporates isotope data (IsogSM; Yoshimura et al., 2008) to analyze the mechanism of the remarkable seasonal change in δ-excess values in precipitation observed in East Asia. In winter, the East Asian seas and western Pacific Ocean are much warmer than the overlying air, which causes the effective relative humidity (h*, vapor pressure normalized by the saturated vapor pressure at sea surface temperature) to be low and the water vapor to have high δ-excess values. The model results showed a higher δ-excess value in the western Sea of Japan (east coast of Korean Peninsula) and the Pacific Ocean adjacent to south coast of Japan. In contrast, the vapor distributed over the central Sea of Japan and the Pacific Ocean adjacent to northeast coast of Japan showed lower δ-excess values.

The results of backward air mass trajectory analysis suggest that the vapor arrived at Hakkoda station have different moisture source. The higher δ-excess values at Hakkoda station than at the other stations reflect the water vapor arrived at the station derived from relatively remote ocean areas. Thus, the precipitation that falls at Hakkoda station in winter is mainly derived from water vapor from the western Sea of Japan areas (east coast of Korean Peninsula). In contrast, the relatively lower δ-excess values observed at the two coastal stations indicate that the winter precipitation was derived from water vapor that evaporated from the central or eastern parts of the Sea of Japan.

CONCLUSIONS

We reported the measurement results of δD and δ18O values in precipitation samples collected at Rokkasho, Ajigasawa, and Hakkoda stations. Values were generally lower at Hakkoda station than at the two coastal stations, and they showed weak seasonal variation at all three stations. In particular, the 11-year monthly averaged δ18O in precipitation at Rokkasho station showed clear seasonality, with higher values in summer and lower values in winter. However, δ18O was not clearly correlated with local meteorological parameters. Temporal variations of δ18O at the two coastal stations were similar, although they face different oceans, and the timing of precipitation events differed between them. Thus, conditions in the source area of the water vapor are a more important control on δ18O values at the three stations than local climatic conditions.

The δ-excess values at all stations showed clear seasonality, being high in winter and low in summer, reflecting seasonal changes in the water vapor sources of the air masses arriving over northeast Japan. Higher δ-excess values were observed at Hakkoda station in winter than at the other two stations, however, suggesting that precipitation at Hakkoda was derived from water vapor from a remote area of Sea of Japan.

To clarify in more detail the relationship between the water isotopic system and climatic parameters, event-based observations and numerical model-based analyses are required.

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**SUPPLEMENTARY MATERIALS**

URL (http://www.terrapub.co.jp/journals/GJ/archives/data/46/MS279.pdf) Table S1