



**Isoscape of
streamwater across
the Japanese
archipelago**

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Spatial distribution of oxygen-18 and deuterium in stream waters across the Japanese archipelago

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Abstract

The spatial distribution of oxygen and hydrogen isotopic composition ($\delta^{18}\text{O}$ and $\delta^2\text{H}$) of stream waters across Japan was clarified with a data set compiling sample data obtained from 1278 forest catchments during the summer of 2003. Both $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values showed positive correlations with the mean annual air temperature and annual evapotranspiration, and negative correlations with latitude and elevation. Deuterium excess (d excess) values in stream waters were higher on the Sea of Japan side, and lower on the Pacific Ocean side, of the Japanese archipelago. The d excess in precipitation was generally higher in winter and lower in summer in Japan. The Sea of Japan side experiences a great deal of snowfall, and seasonal changes in monthly precipitation are rather small. In contrast, the Pacific Ocean side experiences a large amount of rainfall during summer with low levels of precipitation during the winter. Therefore, the lower d excess in stream waters on the Pacific Ocean side reflects summer precipitation, and the higher values on the Sea of Japan side are affected by delayed recharge from snowmelt. The isoscapes of stream water connote not only spatially integrated but also temporally integrated isotope signals of precipitation, and provide a framework for addressing applied hydrological, ecological, or meteorological research questions at regional scales, such as the effects of climate change.

1 Introduction

The importance of “isoscapes”, that is, the mapping of large-scale spatiotemporal distributions of stable isotope compositions in various environments (West et al., 2010), is being recognized as providing a framework for fundamental and applied research questions at large scales. The Global Network for Isotopes in Precipitation (GNIP) database has been applied, for example, to monitor climate-change impacts on the character and intensity of precipitation (Aggarwal et al., 2012), as well as

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wide and spatially dense stream water isoscape for Japan, which has a wide range of climatic and geographical conditions over a small area, may provide spatial isotope information fundamental for the application of isotopes in hydrological studies.

Here we present the stream water $\delta^{18}\text{O}$ and $\delta^2\text{H}$ isoscape of the Japanese archipelago, and provide multivariate regression analyses using key environmental and geographical parameters to determine which variables are the key drivers of stream water isotopic patterns. The identification of key parameters is essential in evaluating the vulnerability of hydrological responses in the watershed to climate change. Moreover, by comparing the data with existing precipitation-isotope data, we consider the advantage of using stream water isoscapes as an integrated indicator of precipitation for future isotopic hydrology studies.

2 Methods

2.1 Stream water sampling and measurement

The sampling campaign “Japan-Wide Stream Monitoring (JWSM) 2003” (Konohira et al., 2006) was conducted during the summer, from 1 July to 11 October of 2003 by 11 researchers. All samples were collected from forested headwater streams to avoid the influence of anthropogenic impacts such as agriculture or urban effects. The sampling points covered 45 prefectures; only two prefectures, Chiba and Okinawa, were excluded from the campaign. Samples were collected from approximately 30 catchments in each prefecture, and finally, 1278 forested headwater catchments were selected (Fig. 1). The catchment areas ranged from 0.05 to 136.8 km², and were 5.3 km² on average. In total, 95.3 % of the catchments were smaller than 15 km². The sampling procedures were unified between all 11 researchers prior to sampling. During the campaign, grab samples of stream water were collected once in each catchment under baseflow conditions to avoid the effects of rainstorms. The collected samples were immediately filtered and preserved by freezing.

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The stream water samples were analyzed by the Colorado Plateau Stable Isotope Laboratory using an Off-Axis Integrated Cavity Output Spectroscopy liquid water isotope analyzer (Los Gatos Model 908-0008).

2.2 Isotope data on precipitation and climate conditions

5 We collected isotope data on precipitation from the published literature and unpublished data kindly offered by many researchers, in addition to our original data. The policy for collecting data was that both $\delta^{18}\text{O}$ and $\delta^2\text{H}$ were to be measured monthly or more over 1 year to calculate the mean annual weighted value of the successive rain inputs. The data were collected from 14 prefectures (Fig. 1 and Table 1). In
10 three of these prefectures, Shiga (No. 6 in Fig. 1), Nara (8), and Tottori (9), the precipitation sampling has been continuous. In Shiga, the monitoring began in 1997 at the Kiryu Experimental Watershed (Kabeya et al., 2007; Katsuyama et al., 2010). Sampling began in 2004 at the Mt. Gomadan Experimental Forest in Nara (Katsuyama et al., 2008; Fukushima and Tokuchi, 2009), and in 2011 at the Hiruzen Experimental
15 Forest of Tottori University (Haga and Katsuyama, unpublished data). The Hiruzen Forest is located on the Okayama side of the Okayama–Tottori prefectural border and the samples were collected from the mountain peak. Therefore, we term this station “Tottori” to clearly distinguish it from Okayama (10).

20 The precipitation samples from five of these prefectures, Shiga (6), Kyoto (7), Nara (8), Tottori (9), and Kochi (12) were analyzed using the $\text{CO}_2\text{--H}_2\text{O}$ equilibrium method with a mass spectrometer (Thermo Electron MAT252) at the Center for Ecological Research, Kyoto University.

The isotope values of both stream water and of precipitation are reported as per mil (‰) units relative to the Vienna Standard Mean Ocean Water.

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2.3 Geographical and environmental parameters and criteria for contour plots

The catchment area and elevation were determined using a 250 m digital elevation model for each sampling point. Mean annual precipitation (MAP) and mean annual temperature (MAT) were extracted from Mesh climatic data for the year 2000 (Japan Meteorological Agency, 2002). Actual evapotranspiration (AET) was extracted from Ahn and Tateishi (1994), which they estimated using the Priestley–Taylor method (Priestley and Taylor, 1972).

All of the contour plots presented herein were created using the free GIS software MANDARA (<http://ktgis.net/mandara/>). The type of gridding routine used by this software is the inverse distance weighting method.

3 Results

3.1 Relationship between $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values in stream water

The measured $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values in stream water samples ranged from -13.7 to -5.9‰ (mean = -9.2‰) and from -92.2 to -35.1‰ (mean = -56.6‰), respectively (Table 2). A clear linear relationship existed between $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values (Fig. 2) as

$$\delta^2\text{H} = 6.85\delta^{18}\text{O} + 6.11 \quad (n = 1278, r^2 = 0.89, p < 0.001). \quad (1)$$

The dataset forms a flattened ellipse around the regression line. This relationship is similar to the results from previous studies in Japan. Machida and Kondo (2003) collected isotope data for 1067 rivers and shallow groundwater sources from many papers and databases as:

$$\delta^2\text{H} = 6.72\delta^{18}\text{O} + 3.94 \quad (n = 1067, r^2 = 0.91). \quad (2)$$

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Moreover, they recalculated the data for surface water and shallow groundwater presented by Mizota and Kusakabe (1994) as:

$$\delta^2\text{H} = 7.03\delta^{18}\text{O} + 7.91 \quad (n = 298, r^2 = 0.93). \quad (3)$$

Both the slope and the intercept of Eq. (1) are intermediate compared to those of Eqs. (2) and (3). The numbers of sampling point in this study exceeded those in Mizota and Kusakabe (1994) and Machida and Kondo (2003). However, the data presented in these previous studies were collected from samples in different years and seasons. On the other hand, the data we present here was systematically collected within a few months of the same year. Generally, surface waters show very limited isotopic seasonality compared to precipitation, due to leveling during infiltration and water movement processes within catchments. Therefore, the Eq. (1) will produce a more reliable general relationship between the $\delta^{18}\text{O}$ and $\delta^2\text{H}$ of stream water in Japan.

The data are grouped into 10 regions and the linear regressions applied to each region (Table 2). The regional division used followed that of the Japanese Meteorological Agency used for weather forecasts (see Fig. 3 for the locations of each region). The regressions for individual regions had a range of slopes and intercepts. For example, Kinki (F) – and Chugoku (G) regions had small slopes and intercepts, although the r^2 values were low. The data from these two regions, especially from Chugoku, had a relatively narrow range of δ values and were plotted on the upper region of the data ellipse (Fig. 2). In other regions, the slopes were relatively similar (about 6–8); however, the intercepts were varied. These results mean that the ellipse of the data in Fig. 2 is composed of many local regression lines for the individual regions.

In Japan, nationwide systematic observations of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ in precipitation have not been carried out. Under such circumstances, Tase et al. (1997) presented a local meteoric water line for Japan from the observations made at 16 stations located in the Kanto region and southwest Japan as:

$$\delta^2\text{H} = 7.3\delta^{18}\text{O} + 8.6 \quad (r^2 = 0.89). \quad (4)$$

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Comparing the regressions for stream water (Eqs. 1–3) and for precipitation (Eq. 4), the equations appear generally similar, suggesting that the isotopic compositions of precipitation are reflected in the compositions of stream water at the national scale. The smaller slopes and interceptions in the former equations reflect the effects of evaporation during infiltration processes. However, as mentioned above, the ellipse of data in Fig. 2 is composed of many local regression lines; therefore, more precipitation data are needed to interpret the meaning of slopes at regional scales to allow these results to be discussed in detail.

3.2 Spatial distribution of $\delta^{18}\text{O}$, $\delta^2\text{H}$, and d excess values in stream water

The Japanese archipelago is elongated from northeast to southwest, and mountains form the backbone. High mountains are located at the centers of Honshu and Hokkaido Islands, named the Japan Alps (near $35\text{--}37^\circ\text{N}$, $136\text{--}139^\circ\text{E}$) and Mt. Daisetsu region ($43^\circ40'\text{N}$, $142^\circ51'\text{E}$), respectively. A topographic map of the Japanese archipelago is available on the web (for example; http://en.wikipedia.org/wiki/File:Japan_topo_en.jpg).

The spatial distributions of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ are very similar (Fig. 4a and b). Generally, the values decreased from south to north. The resulting contour intervals in high mountain areas are very dense. The smallest values were observed at the Mt. Daisetsu region of Hokkaido, although the intervals in southwest Japan were very sparse.

Deuterium excess (d excess, $d = 8\ \delta^2\text{H} - \delta^{18}\text{O}$) is known, and provides information about the climate conditions of the moisture sources. The d excess values in stream water were clearly divided by the backbone mountain ranges of the Japanese archipelago; and the values in stream water were lower on the Pacific Ocean side and higher on the Sea of Japan side (Fig. 5). The d excess values in stream water samples ranged from 0.9 to 26.9‰ (mean = 16.6‰). The specifically small d excess values ($d < 8$) were only observed in Gunma Prefecture located at the northern end of the Kanto Plain, while the highest values were observed in Niigata Prefecture in the Hokuriku districts, the heaviest snowfall area in Japan. This pattern of d excess

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in some parts of Japan was reported previously in the pioneer work by Waseda and Nakai (1983). They found that the d excess of surface waters in Central and Northeast Japan tended to increase continuously from the Pacific Ocean side to the Sea of Japan side, ranging from 9.1 to 22.4.

3.3 Seasonal variation of d excess values in precipitation

Figure 6 shows typical examples of monthly d excess values observed at Tottori (Sea of Japan side, Station No. 9 in Table 1) in 2011, Shiga (Sea of Japan side, Station No. 6) in 2008, and Nara (Pacific Ocean side, Station No. 8) in 2006. The climate conditions are clearly different among these stations. In Tottori, much snow falls from December to March with low air temperatures. In Shiga, less snow occurs but much more rain falls during summer. Summer rainfall is more plentiful in Nara. However, these three stations showed similar sinusoidal d excess variations; i.e., higher during winter and lower during summer. However, we unfortunately did not have sufficient data from 2003 when stream water sampling was conducted, with the precipitation sampling having been continued for multiple years at these stations, and the annual sinusoidal patterns of d excess were repeated (e.g. Kabeya et al., 2007). Moreover, Tase et al. (1997) also reported that this seasonal pattern was commonly observed at six stations in the Kanto, Shikoku, and Kyushu regions (see also Fig. 3). Therefore, we will compare the precipitation values observed in various years with the stream water values, in the following.

4 Discussion

4.1 Correlations between $\delta^{18}\text{O}$ and environmental or geographical parameters

Generally, the $\delta^{18}\text{O}$ of precipitation is affected by various environmental and geographical variables. Here, we will discuss the effect on the $\delta^{18}\text{O}$ of stream water of these parameters as a proxy of precipitation. Stream water $\delta^{18}\text{O}$ had strong positive

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correlations with MAT (Fig. 7a) and AET (Fig. 7c), as well as a negative correlation with latitude (LAT) (Fig. 7d) and elevation (ELV) (Fig. 7e), variables that are commonly used to describe the temperature, latitude, and elevation effects in precipitation. These relationships are similarly found in rivers across the United States (Kendall and Coplen, 2001), and the trends with MAT and LAT in groundwater isotopic compositions have also been observed in Finland (Kortelainen and Karhu, 2004).

The correlation between $\delta^{18}\text{O}$ and MAT is particularly strong (Fig. 7a). The large variation in MAT reflects the geographical features of the Japanese archipelago extending north and south. The AET was calculated by Priestley and Taylor's (1972) method, which increases in proportion to the net radiation. Thus, AET generally decreases at higher latitudes. Indeed, the correlation coefficient between latitude and AET was very high ($r = -0.88$). Therefore, the positive correlation between $\delta^{18}\text{O}$ and AET (Fig. 7c) covers both the temperature effect (Fig. 7a) and the latitude effect (Fig. 7d). The scatter around latitude 35–37° N in Fig. 7d reflects the elevation effect around the Japan Alps area; and the relationship between $\delta^{18}\text{O}$ and ELV around this area (35–37° N, 136–139° E) is $\delta^{18}\text{O} = -0.0027 \text{ ELV} - 8.4$ ($r^2 = 0.60$, $p < 0.01$; figure not shown). The slope of the regression line for the elevation effect (Fig. 7e), that is, the isotopic lapse rate of stream water was $-0.28\text{‰}/100\text{ m}$, which is the same as the global isotopic lapse rate of precipitation (Poage and Chamberlain, 2001). This result suggests that stream waters retain the properties of precipitation, and thus, the spatial patterns of stream water samples may be a suitable proxy for precipitation. However, the amount effect, which is commonly observed to be negatively correlated with precipitation, was not as clear (Fig. 7b). Moreover, no clear relationship was found between $\delta^{18}\text{O}$ and catchment area (ARA) (Fig. 7f).

Based on these relationships, a multiple regression model was developed to identify the controls of environmental (MAT, MAP, and AET) and geographical (LAT, ELV, and ARA) parameters on stream water $\delta^{18}\text{O}$. As noted above, however, AET was highly controlled by latitude and therefore was not appropriate as a predictor variable. The

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and Fritz, 1997; Dutton et al., 2005). Table 1 shows the comparison of d excess values between precipitation and stream water. On the Pacific Ocean side, the d excess values of precipitation and stream water were nearly equal: the difference was less than 1 ‰. On the Kanto Plain (stations 2, 3, and 4), the values were very close. On the Sea of Japan side, however, the values of stream water were clearly larger than those of precipitation, with a difference of more than 3 ‰, except in Fukuoka Prefecture. These results mean that seasonal biases exist to recharge on the Sea of Japan side. Compared to the Pacific Ocean side, the Sea of Japan side experiences a great deal of snowfall, and the d excess of meteoric input was larger in winter (Fig. 7). Thus, the water recharge from late snowmelt may have affected summer stream water, even though our stream water sampling was conducted during the summer (from July to October). Fukuoka Prefecture, an exception, is located on Kyushu Island and has a warm climate with less snowfall. Thus, the d excess of the precipitation and of the stream water was similar. Therefore, in particular on the Sea of Japan side, we must take into account the winter snowpack and spring snowmelt when considering the recharge processes of the stream. The largest d excess of precipitation was observed in Tottori Prefecture facing the Sea of Japan. The precipitation samples were collected at the border of Tottori and Okayama prefectures (Haga and Katsuyama, unpublished data), and snowmelt recharges the stream water in both prefectures. Therefore, as Yamamoto et al. (1993) pointed out, the d excess of the stream water in Okayama Prefecture was relatively large (Table 1).

The comparison of d excess values in precipitation with stream water demonstrates that although meteoric water has relatively spatially homogeneous isotopic compositions (Tase et al., 1997), the recharged stream water does not necessarily reflect the pattern of the meteoric water. The delayed contribution of snowmelt controlled the isotopic compositions of stream water in snowy regions. As there is no clear relationship between the catchment areas or elevation with d excess values (figures not shown), the difference in the elevation of the actual stream water recharge area cannot be the case of differences in d excess signatures. Moreover, the mean

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residence time of stream water is not controlled by catchment size (e.g., Tetzlaff et al., 2009; Katsuyama et al., 2010); therefore, the delayed contribution of snowmelt is not controlled by geographical parameters. To discuss this more directly, we need a nationwide systematic data set of isotopic compositions in precipitation. Tase et al. (1997) reported the only example of such observation. However, unfortunately, their stations were mainly located on the Pacific Ocean side, and no stations existed in the northern part of Japan, i.e., the snowy regions of Tohoku and Hokkaido. In other words, although comprising only one episode of sampling, our nationwide systematic data set of stream water reveals not only spatially integrated but also temporally integrated isotope signals of precipitation, and the stream water d excess values reflect the dampening of precipitation input; i.e., the differences in rainfall–runoff dynamics among the catchments.

4.3 Vulnerability of water resources to global warming

As discussed above, winter snowfall recharges summer stream water at the Sea of Japan side, which experiences a great deal of snowfall. This result means that summer water supply in this region is highly dependent on winter snowfall. Brooks et al. (2012) clarified that the water sources during summer depend on winter snow accumulated in the mountains in western Oregon, and have pointed out the vulnerability of the system to the influences of a warming climate, as snowpack volume is predicted to decline in the future. A decrease in snowfall caused by global warming, and the consequent vulnerability of water resources, are also predicted to occur in Japan (Kazama et al., 2008). Moreover, mean air temperature is an important environmental parameter in determining the $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values of stream water, as shown in Eqs. (6) and (8). Global warming can also change vegetation cover and evapotranspiration rates in the watershed, and the resulting amount of annual runoff (Gedney et al., 2006). This implies that the importance of evapotranspiration rates as a control parameter will change over the long-term. Needless to say, transit times differ among stream waters. Therefore, the isotopic signature of stream water reflects the history of precipitation in

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the catchment. In other words, the isoscapes of stream water represent an integrated reflection of the contribution of these environmental factors, and of the past and present climatic conditions. Therefore, this kind of research should be conducted continuously every few decades because the effects of climate change will be reflected in these isoscapes over time, and can provide information regarding changes in regional hydrological- and water-resource conditions.

5 Conclusions

The $\delta^{18}\text{O}$ and $\delta^2\text{H}$ isoscapes of stream water in Japan showed clear spatial distributions, which were effectively explained by three parameters: latitude, elevation, and mean annual temperature. These parameters are commonly known as control factors of the isotopes of precipitation. Therefore, our data set is applicable, to some extent, as a proxy for the isotopic composition of precipitation in Japan. This result reflects the advantage and importance of isotope contents in stream water at a national scale, because stream water is more easily collectable than is precipitation. However, the comparison of d excess proved that the stream water d excess values were biased toward the values of winter precipitation in snowy regions, although our sampling campaign was conducted during the summer. These results do not merely signify the importance of continuously observing precipitation in snowy regions, but also warn us to discreetly use the temporal variations in isotopic signals when estimating, for example, rainfall–runoff processes and/or the mean residence time of stream water. Isoscapes of stream water reflect the recharge processes from source water and the distribution of water resources. Therefore, this technique will provide a valuable method for hydrological and ecological research, as well as in predicting the impacts of climate change and estimating the vulnerability of water resources at the regional scale. In particular, these results from Japan, a country with a wide range of climatic and geographical conditions across a small land area, represent a case study that will facilitate similar studies in other regions.

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Table 1. Comparison of d excess values between precipitation and streamwater.

Station No.	Prefecture	Sampling Location of Precipitation ^a		MAT	Latitude N	Longitude E	d excess (%) ^b		Data source of Precipitation d excess	Reference of Site description	Remarks
		Side	Site				Precipitation	Streamwater			
1	Akita	J	Akita Univ.	11.7	39.7	140.1	17.0	20.3	Kawaraya et al. (2005)		c,d
2	Ibaraki	P	Mt. Tsukuba	9.7	36.2	140.1	12.0	13.6	Yabusaki et al. (2008)		
3	Saitama	P	Rissho Univ.	15.0	36.1	139.4	13.6	12.9	Yabusaki (2010)		c
4	Kanagawa	P	Oobora Watersheds, Tanzawa Mountains	15.1	35.5	139.2	13.3	14.0	Oda (unpub.)	Oda et al. (2013)	
5	Toyama	J	Toyama City	14.1	36.7	137.2	15.4	19.6	Satake et al. (1984)		c,d
6	Shiga	J	Kiryu Exp. Watershed	13.5	34.9	136.0	13.8	20.3	Katsuyama (unpub.)	Katsuyama et al. (2010)	
7	Kyoto	J	Kyoto Univ.	15.1	35.0	135.8	12.7	18.5	Katsuyama (unpub.)		
8	Nara	P	Mt. Gormadan Exp. Forest	8.9	34.1	135.6	13.9	13.7	Katsuyama (unpub.)	Fukushima and Tokuchi (2009)	
9	Tottori	J	Hiruzen Exp. Forest, Tottori Univ.	10.5	35.3	133.6	18.7	21.7	Haga and Katsuyama (unpub.)	Sano et al. (2014)	
10	Okayama	J	Okayama Univ.	16.2	34.7	133.9	11.8	17.6	Yamamoto et al. (1993)		c
11	Kagawa	P	Takamatsu City	16.3	34.3	134.0	11.3	12.3	Tase et al. (1997)		c
12	Kochi	P	Mt. Takatori	13.1	33.3	133.0	13.7	13.8	Shinomiya and Sakai (unpub.)	Shinomiya and Yoshinaga (2008)	
13	Fukuoka	J	Ochozu Experimental Watershed	16.2	33.6	130.5	12.9	13.7	Asano and Chiwa (unpub.)	Ide et al. (2009)	
14	Kagoshima	P	Takakuma Exp. Forest, Kagoshima Univ.	14.6	31.5	130.8	14.6	14.5	Asano and Tateno (unpub.)	Jitousono et al. (2010)	

^a “J” and “P” mean Sea of Japan side and Pacific Ocean side, respectively, and “MAT” means mean annual temperature.

^b The d excess values for precipitation are annual weighted means with the amount of precipitation, and the values for streamwater are the arithmetic means of all data in each prefecture.

^c Meteorological data are from nearest observation station by the Japan Meteorological Agency (AMeDAS).

^d The d excess of precipitation are arbitrarily read by us from the original papers.

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Table 2. Range of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values in streamwater samples and liner regressions for each region.

Region	<i>n</i>	$\delta^{18}\text{O}$		$\delta^2\text{H}$		slope	intercept	r^2
		max.	min.	max.	min.			
A Hokkaido	94	−9.0	−13.7	−54.7	−92.2	6.94	6.79	0.87
B Tohoku	167	−7.6	−12.7	−47.9	−79.9	6.27	1.73	0.84
C Kanto–Koshin	226	−7.1	−13.1	−46.1	−89.2	7.07	4.50	0.93
D Hokuriku	124	−7.8	−13.2	−43.7	−87.9	7.76	19.38	0.90
E Toukai	105	−6.8	−12.4	−38.6	−80.1	6.57	3.20	0.96
F Kinki	175	−6.3	−9.9	−37.7	−62.0	5.15	−6.96	0.62
G Chugoku	117	−7.5	−10.1	−44.6	−56.1	2.96	−25.59	0.40
H Shikoku	111	−6.1	−9.8	−35.1	−63.2	8.02	13.91	0.93
I Northern Kyusyu	119	−5.9	−9.0	−38.0	−56.7	6.27	0.31	0.87
J Southern Kyusyu	40	−6.4	−8.6	−38.3	−56.3	6.80	4.84	0.73
National	1278	−5.9	−13.7	−35.1	−92.2	6.85	6.11	0.89

^a Regional division is shown in Fig. 3.

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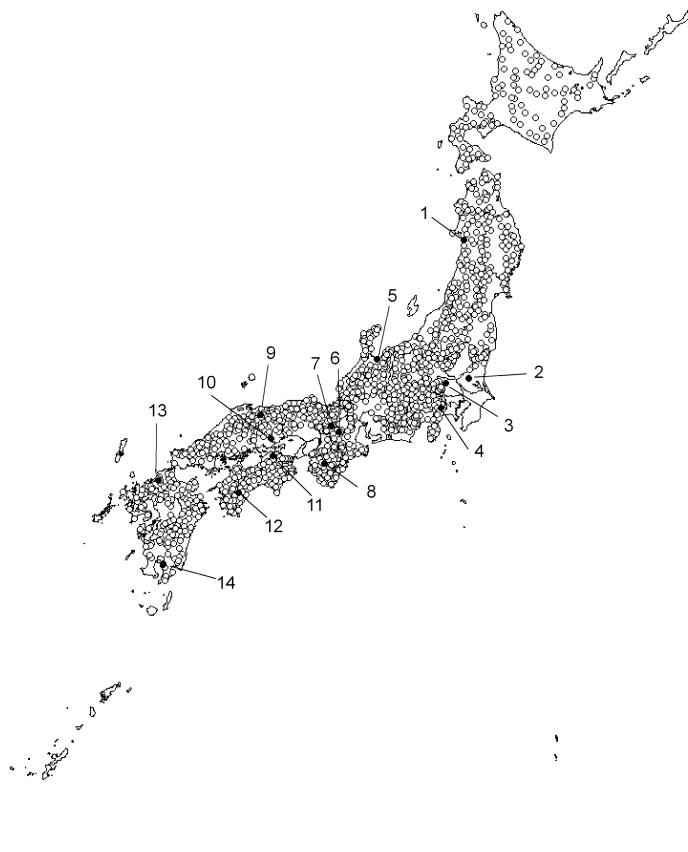
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Figure 1. Index map showing locations of the 1278 streamwater sampling points and referred 14 sampling stations of precipitation. Note: open circles: streamwater sampling points. Filled circles: precipitation sampling station. The station numbers correspond to those in Table 1.

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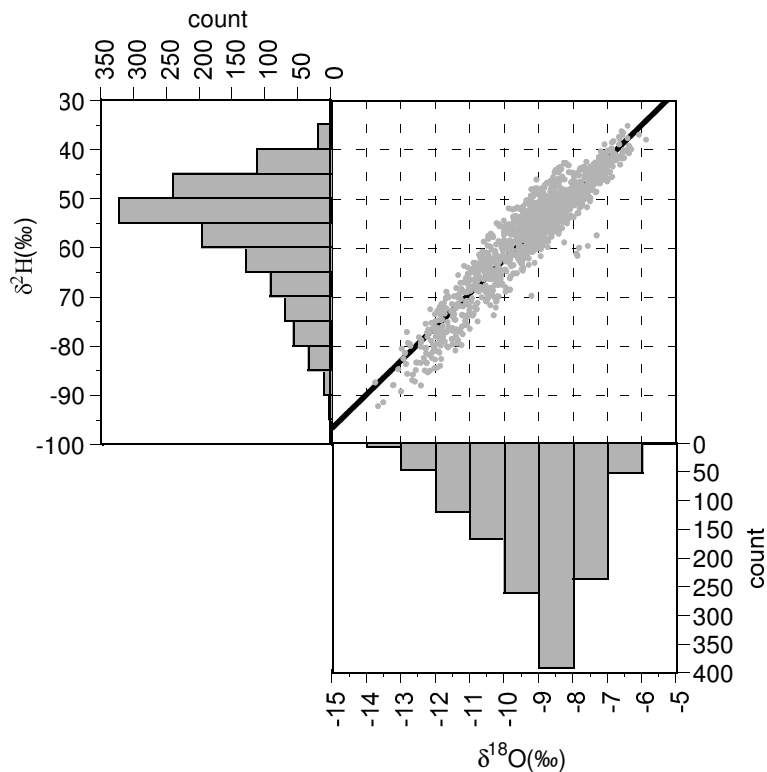


Figure 2. Relationship between $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values and their frequency distributions in streamwater.

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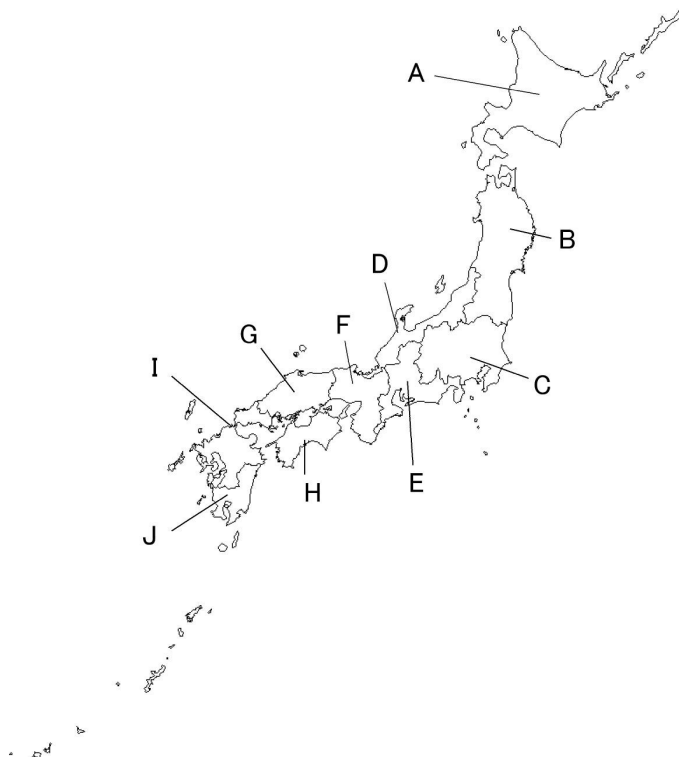
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Figure 3. Index map of regional deviation. Note: regional division is shown in Table 2.

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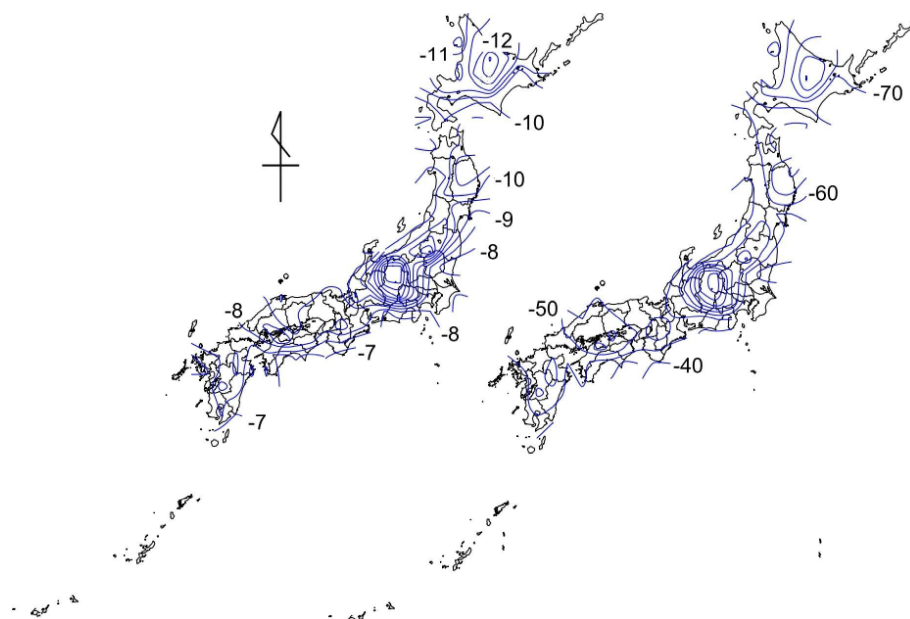
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Figure 4. Spatial distribution of **(a)** $\delta^{18}\text{O}$ (left panel) and **(b)** $\delta^2\text{H}$ (right panel) values in streamwater.

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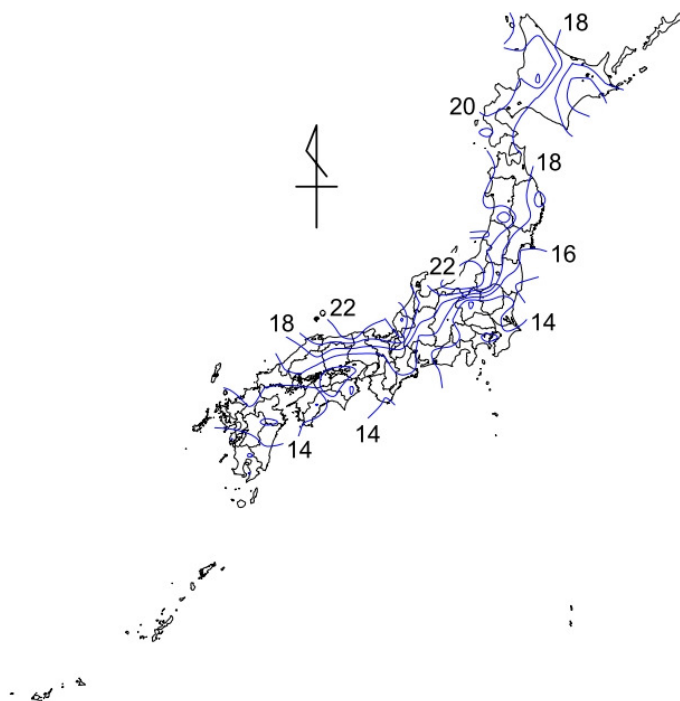
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Figure 5. Spatial distribution of d excess values in streamwater.

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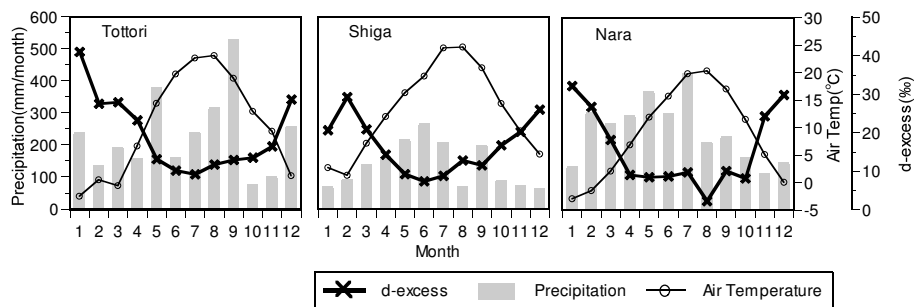


Figure 6. Seasonal variations of d excess values in precipitation.

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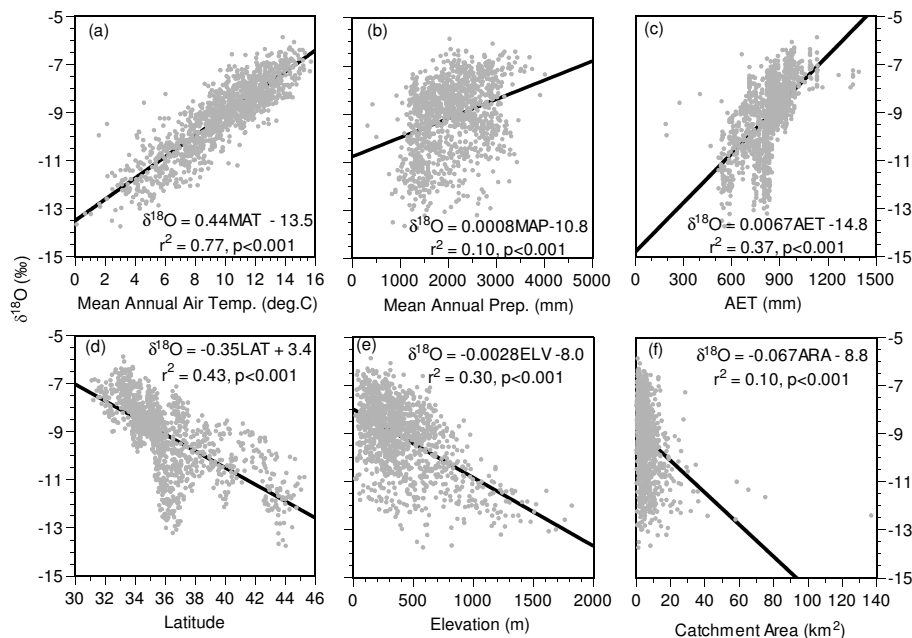


Figure 7. Correlations of $\delta^{18}\text{O}$ and environmental or geographical parameters.

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