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Water Resources Research

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Key Points:
- A new nationwide network of tap water isotope data across China was established
- Connection between monthly stable isotopes in tap water and precipitation is identified
- Diagnostic patterns of tap water isotopes are associated with water resource use

Supporting Information:
- Supporting Information S1
- Table S1

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Water Source Signatures in the Spatial and Seasonal Isotope Variation of Chinese Tap Waters

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Abstract Different water sources exploited for public use have different exposure to risks associated with climatic and environmental change. Isotope ratios of tap water have previously been studied as a potential tool to link public supply waters with water source characteristics at local to continental scales, providing information on the footprint of and potential risks associated with the water sources used. Work that combines intensive spatial and temporal sampling with independent water management data has been limited, however. In this study, an extensive observation network was established during 2014–2016 to provide monthly tap water sampling across China. We show that the spatial distribution of annual mean tap water isotope ratios is generally consistent with that of local precipitation across China. We identify seasonal correlation between tap water and precipitation isotope ratios in south China, where use of surface water is prevalent. In contrast, relatively invariant tap water isotope ratios elsewhere in China, which are not correlated with seasonal variation of precipitation isotope ratios, can be attributed to use of groundwater or water from river basins with longer storage times. The tap water isotope signatures identified here could be widely applied to characterize water supplies and associated sustainability challenges in different regions worldwide.

1. Introduction

Most tap water comes from local groundwater (e.g., well) or surface water sources (e.g., river, lake, or reservoir; Maupin et al., 2014). In some urban and agricultural lands, however, trans-basin water transfers also contribute to local water supplies (Liu et al., 2013). The sufficiency and safety of domestic water supplies is an increasing challenge under global climate change and population explosion (Ehleringer et al., 2016; Jasechko et al., 2017). The stable isotope composition of tap water preserves information on the source of these waters and can provide information on tap water origin and evaporation losses from water supplies, as well as vulnerabilities to environmental change, which are not obvious from traditional water management data (Bowen et al., 2007). In the past decade, the spatial pattern and seasonal variability of stable isotope ratios in tap water has aroused attention in water resource studies, including regional water use and source diagnosis (e.g., Good et al., 2014; Jameel et al., 2016; Landwehr et al., 2014; Tipple et al., 2017; West et al., 2014). In addition, information on tap water isotope ratios has also been important in other fields including ecology, food, and forensic sciences (Bowen & Good, 2015; Ehleringer et al., 2008; Ueda & Bell, 2017).

Stable isotope tracers in tap water have been reported in several case studies. For example, if the spatial distribution of isotope ratios in tap water and potential water sources is known, probable contributions of nonlocal water use within hydrologic basins can be identified, providing information on the impacts of water transfer projects (Good et al., 2014). An isotopic mass balance using tap water in an urban area demonstrated interannual and intraannual variability in evaporative water losses within a water distribution network (Jameel et al., 2016). Local water management (e.g., water source switching and conservation) responses to drought events have also been identified through the isotope composition of tap water (Tipple et al., 2017).

China is a densely populated country with complex water supply systems (Jiang, 2015). The stable isotope composition in China's tap water may be an effective source of information, complimentary to traditional records, to understand the water supply regime and resource management. Recently, the first measurements of tap water isotopes across China were reported (Zhao et al., 2017), but further studies expanding the

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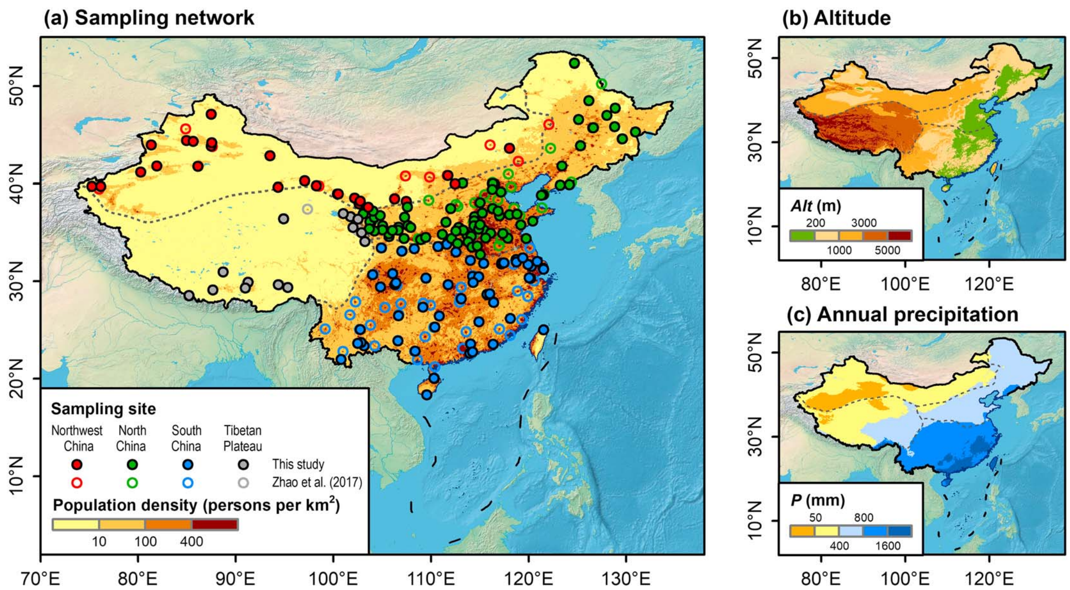

Figure 1. (a) Spatial distribution of sampling sites of tap water and population density in China. The population density in 2015 is derived from Lloyd et al. (2017). The tap water sampling locations of Zhao et al. (2017) are also shown. The four natural subregions of China (marked using dashed grey lines) are based on Zhao (1983). The satellite-derived land cover base map is from Natural Earth (http://www.naturalearthdata.com). (b) Spatial distribution of altitude in China, derived from Global 30 Arc-Second Elevation (GTOPO30; https://lta.cr.usgs.gov/GTOPO30). (c) Spatial distribution of annual precipitation amount during 1970–2000, derived from the WorldClim-Global Climate Data version 2 (Fick & Hijmans, 2017; http://worldclim.org/version2).

available data and exploring the implications of tap water isotope variability in China are needed. In this study, an enhanced observation network was established to provide monthly tap water sampling across China. The larger database, with greater spatial and temporal coverage, provides a new platform to understand the seasonal variation of stable isotopes in China's drinking water as well as the signatures of water supply and water management embedded in these data.

2. Materials and Methods

2.1. Tap Water Isotope Database

From August 2014 to March 2016, tap water samples were collected through a volunteer network across China (Figure 1). Given our study's focus on understanding dominant patterns of water use and their relation to climate, we sampled more densely in high-population regions and areas such as remote western China were undersampled relative to their land area. According to the population density map, most urban belts in western China were still well sampled. In a traditional natural zoning of China (including northwest China, north China, south China, and Tibetan Plateau), the highest number of sampling stations is located in north China and south China, which exhibit typical monsoon climate and higher precipitation amount than the other regions. Volunteers who participated in the network were instructed to collect cold tap water at a monthly basis (end of each month was recommended). The water samples were collected in 50-ml HDPE (high-density polyethylene) bottles after 10 s of allowing the tap to run, and then capped and sealed using waterproof tapes. The samples were later returned to the Stable Isotope Laboratory, College of Geography and Environmental Science, Northwest Normal University in Lanzhou, and stored in a cool environment before analysis. A total of 2,099 tap water samples at 177 sites was collected during the period (Tables S1 and S2 in the supporting information). The number of sampling stations for each month is generally stable, especially from October 2014 to September 2015, and most stations have 12 samples or slightly more (Figure S1 in the supporting information).

The tap water samples were analyzed using a DLT-100 liquid water isotope analyzer (Los Gatos Research, Inc.) at the Stable Isotope Laboratory, College of Geography and Environmental Science, Northwest Normal

University. Every isotopic standard or sample was injected sequentially 6 times using a microliter syringe, and then the arithmetic average of last four injections was accepted as the final result. The values are expressed as δ-values relative to V-SMOW (Vienna Standard Mean Ocean Water):

$$\delta_{\text{sample}} = \frac{R_{\text{sample}} - R_{\text{standard}}}{R_{\text{standard}}} \times 1000‰ \quad (1)$$

where R_{sample} is the ratio of $^2H/^1H$ (D/H) or $^{18}O/^{16}O$ in the samples and R_{standard} is the ratio of $^2H/^1H$ or $^{18}O/^{16}O$ in V-SMOW. The precision, based on replicate injections of each sample, is ±0.6‰ for δ^2H and ±0.2‰ for $\delta^{18}O$. A similar procedure was applied in our previous work (Chen et al., 2017; Wang, Zhang, Hughes, et al., 2016).

An additional data set of tap water isotopes in China, published by Zhao et al. (2017), was obtained from the Waterisotopes Database (http://wateriso.utah.edu/waterisotopes). The volunteer network of Zhao et al. (2017) ran from December 2014 to December 2015, overlapping with our sampling work, and included 780 samples at 95 sites (Figure 1 and Table S1). The monthly sampling and analysis procedures were generally similar to this study, although a different laser spectroscopy instrument was used. Combining the two databases, 2,879 samples at 272 sites were used in our analysis. A comparison of sample numbers between the two databases is shown in Figure S1.

To develop a standardized and comparable database for further analysis, the arithmetic averages of measured isotopic ratios in spring (March-April-May), summer (June-July-August), autumn (September-October-November), and winter (December-January-February) were calculated at each site for which at least 1 month in the season was sampled. Only stations covering all four seasons were used to calculate annual means, and the four seasonal values were averaged without weighting to obtain the annual value. There are 163 stations with annual mean in our sampling network, and the total counts of samples are 170 for spring, 166 for summer, 175 for autumn, and 169 for winter. Using the same procedure for data of Zhao et al. (2017), we additionally obtained annual mean values for 59 stations, and the seasonal counts are 91 for spring, 68 for summer, 65 for autumn, and 89 for winter. There is a total of 222 sampling stations with annual mean across the two networks.

2.2. Precipitation Isotope Data Products

To understand the linkage of stable isotope compositions between precipitation and tap water, we used two global data products of estimated annual and monthly δ^2H and $\delta^{18}O$ values in precipitation. (1) OIPC. Estimated monthly and annual values of stable isotopes in precipitation at the locations where tap water was sampled were obtained from the Online Isotopes in Precipitation Calculator version 3.2 (Bowen, 2018; Bowen et al., 2005; Bowen & Revenaugh, 2003). This product uses a periodically updated database of measured precipitation isotope ratios across the world and a statistical interpolation model to produce point estimates of precipitation isotope ratios and has been shown to have good performance as a predictor of long-term average values in most parts of the world (e.g., Aichner et al., 2015; Bai et al., 2017). (2) RCWIP. This was obtained from the Regionalized Cluster-based Water Isotope Prediction (RCWIP) model version 1.00 (Terzer et al., 2013; IAEA [International Atomic Energy Agency], 2018). The model groups the GNIP (Global Network of Isotopes in Precipitation; IAEA/WMO [International Atomic Energy Agency/World Meteorological Organization], 2017) stations with similar climatology using a fuzzy clustering method and uses flexible regressors for each climate cluster. This database has also been applied in many areas of the world in recent years (e.g., Christner et al., 2017; Seltzer et al., 2017). To verify the reliability of the two precipitation isotope data products, we compared their estimates to observational data from the GNIP (IAEA/WMO, 2017) and CHNIP (Chinese Network of Isotopes in Precipitation; Liu et al., 2014) across China (Figure S2 and Table S3). The long-term annual means available for each station (31 GNIP and 29 CHNIP stations) were used, and determination coefficients (r^2) between observations and estimates are 0.8561 (δ^2H) and 0.8000 ($\delta^{18}O$) for OIPC and 0.6514 (δ^2H) and 0.7308 ($\delta^{18}O$) for RCWIP, respectively.

2.3. General Circulation Model-Based Water Isotope Data

To detect the relationship between stable isotopes in precipitation and surface water across China, monthly grids of δ^2H and $\delta^{18}O$ in precipitation and runoff from 1979 to 2007, simulated using isoGSM model (Yoshimura et al., 2008), were acquired from the Stable Water Isotope Intercomparison Group Phase 2

Table 1
Regression Models of Tap Water Isotope Values Using Climatic and Spatial Parameters

No.	Variables[a]	Type	Stepwise regression
1	L^2, L, A	Spatial	No
2	L, O, A	Spatial	No
3	T, P	Climatic	No
4	T, P, R, S, V	Climatic	No
5	L, O, A, L^2, O^2, A^2	Spatial	Yes
6	T, P, R, S, V, T^2, P^2, R^2, S^2, V^2	Climatic	Yes
7	L, O, A, T, P, R, S, V, L^2, O^2, A^2, T^2, P^2, R^2, S^2, V^2	Climatic + Spatial	Yes

[a]*L*, latitude (in degree); *O*, longitude (in degree); *A*, altitude (in m); *T*, air temperature (in °C); *P*, precipitation amount (in mm); *R*, solar radiation (in kJ/[m^2/day]); *S*, wind speed (in m/s); and *V*, water vapor pressure (in kPa).

(SWING2; https://data.giss.nasa.gov/swing2). The isoGSM model incorporates isotopic fractionation processes in an atmospheric general circulation model (GCM) and uses spectral nudging toward global reanalysis dynamical fields (Yoshimura et al., 2008). The spatial resolution of this model is 1.875° (longitude) by 1.904° (latitude). The model runs used here were nudged with National Centers for Environmental Prediction (NCEP) reanalysis data (Risi et al., 2012; Sturm et al., 2010). Among several isotope-enabled GCMs that participated in SWING2, the isoGSM model showed credible performance in China, and the comparison between GNIP/CHNIP observations and GCM simulations was shown in previous studies (e.g., Che et al., 2016; Wang et al., 2015; Yang et al., 2017).

2.4. Water Resource Management Data

A yearbook of water supplies across China compiled by the China Urban Water Association (CUWA, 2016) was used to obtain basic information on approximately 600 water supply organizations (mostly on a city/prefecture scale). The proportional contribution of groundwater in water production capacity for each organization was calculated. Not all areas in China (especially in western China) are covered in the annual yearbook, but the main spatial patterns in densely populated areas are well described. To supplement this information, annual water supply assessments on a provincial basis in 2015 were referenced (NBSC [National Bureau of Statistics of China], 2016; EPB [Environmental Protection Bureau], 2016; WSD [Water Supplier Department], 2016; WRA [Water Resources Agency], 2017). According to the sources, the water supply in China is classified into three categories, that is, surface water, groundwater, and other water sources (sewage treatment/reuse, rainwater harvesting, and seawater desalinization; MWR (Ministry of Water Resources of the People's Republic of China), 2017). In the past decade, the total water supply has slightly increased, but the proportional contributions of each source have been relatively stable on a nationwide basis (NBSC (National Bureau of Statistics of China), 2017; Figure S3).

2.5. Tap Water Isoscape Prediction and Other Methods

Based on previous studies of tap water and precipitation isoscapes (e.g., Bowen & Revenaugh, 2003; Bowen et al., 2007; Lykoudis & Argiriou, 2007; Liu et al., 2008, 2014; Terzer et al., 2013; West et al., 2014; Zhao et al., 2017), several regression models using climatic (air temperature, precipitation amount, solar radiation, wind speed, and water vapor pressure) and spatial parameters (latitude, longitude, and altitude) were explored to develop an isoscape of tap water across China. All the candidate methods used in this study were listed in Table 1. For example, the method 1 is a multiple regression $\delta = aL^2 + bL + cA + d$, where L and A are latitude and altitude, respectively. The Global Topographic 30 Arc-Second Digital Elevation Model (GTOPO30) was used in the tap water isoscape model. The long-term climatic parameters were derived from the WorldClim-Global Climate Data version 2 (Fick & Hijmans, 2017). We used Akaike's Information Criterion (West et al., 2014), adjusted determination coefficient (r_{adj}^2), mean absolute error, and root-mean-square error (Table S4) to select the method 7 in Table 1 as the optimal one in this article. The final isoscape is a combination of regressed estimates and interpolated residual error using Kriging, as recommended in Bowen et al. (2007) for U.S. tap water.

In calculation of correlation coefficient in this study, Pearson's correlation (r) and two-tailed t test were applied.

3. Results and Discussion

3.1. Basic Pattern

Based on data from 222 stations sampled across four seasons (Figure S4), the annual mean value of δ^2H ranges between −149.7‰ and −31.1‰ (median = −58.9‰), and the δ^{18}O value in tap water ranges between −18.5‰ and −3.4‰ (median = −8.4‰). D-excess ($d = \delta^2\text{H} - 8\delta^{18}\text{O}$), a metric for the evaporation of waters, varies from −19.8‰ to 20.0‰, and the median value is 9.4‰. If all 2,879 samples are considered (Figure S4), these ranges are slightly wider (from −153.9‰ to −22.6‰ for δ^2H, from −19.2‰ to 1.2‰ for δ^{18}O, and from −46.7‰ to 24.7‰ for D-excess). We estimate a *tap water line*, expressing the correlation

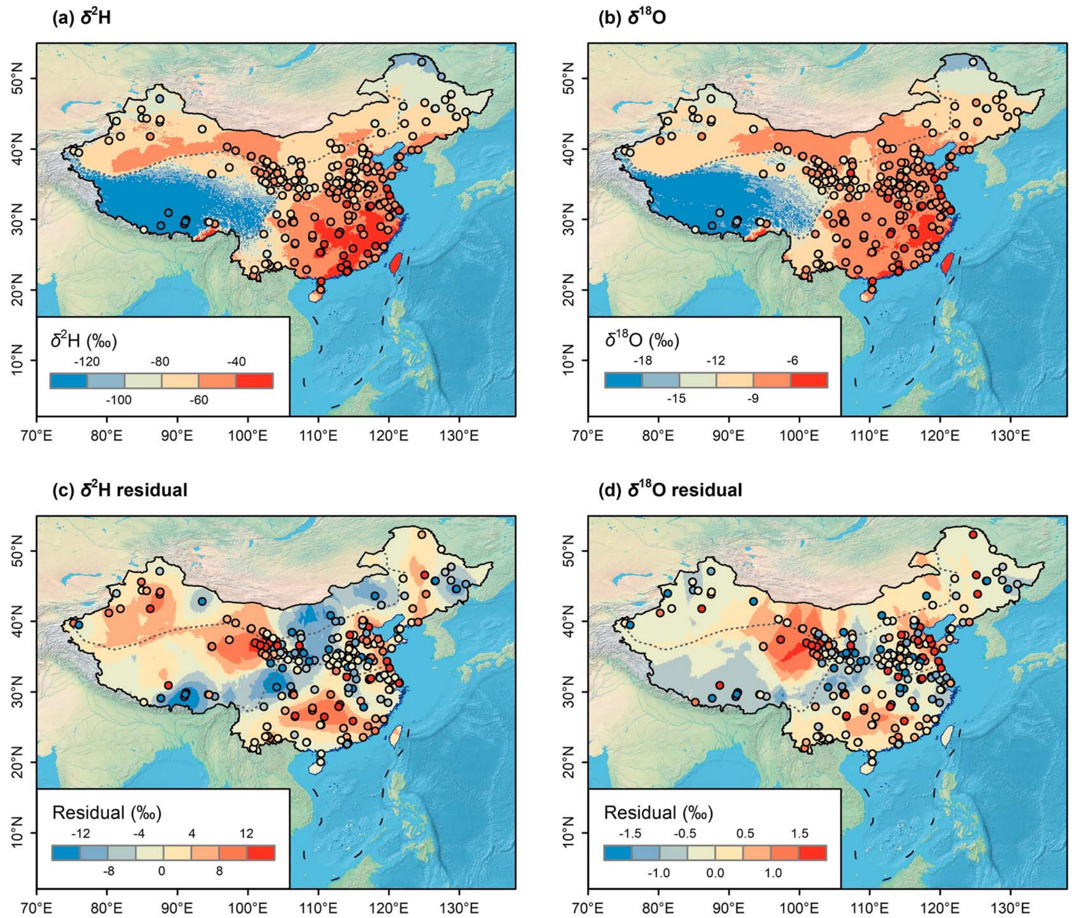

Figure 2. Spatial distribution of annual mean tap water δ^2H and δ^{18}O values and residuals (observation minus estimate) across China. Subregion boundaries and base map as in Figure 1.

between H and O isotope ratios for Chinese tap water, using ordinary least squares regression, as $\delta^2H = 7.66\delta^{18}O + 5.81$ ($r^2 = 0.94$, $p < 0.01$, $n = 222$) using station means, or $\delta^2H = 7.57\delta^{18}O + 5.14$ ($r^2 = 0.93$, $p < 0.01$, $n = 2879$) using raw monthly data. The slope and intercept of the tap water line are less than those of global meteoric water line ($\delta^2H = 8\delta^{18}O + 10$; Craig, 1961), but close to those in China's precipitation ($\delta^2H = 7.48\delta^{18}O + 1.01$, based on 928 monthly samples during 2005–2010, Liu et al., 2014), and a previously published tap water line in China ($\delta^2H = 7.72\ \delta^{18}O + 6.57$ using 780 tap water samples; Zhao et al., 2017).

A stepwise regression model using spatial and climatic parameters (L, L^2, A, A^2, T, T^2, P^2, S^2, V, and V^2 for δ^2H and L, L^2, A, A^2, P^2, S, and V^2 for δ^{18}O; Table S4) was applied to predict δ^2H and δ^{18}O isoscapes of tap water in China (Figure 2). Generally, low values are seen in the western and middle portions of the Tibetan Plateau as well as the northwestern/northeastern margins of China. The coastal region in eastern China usually is characterized by relatively high isotope ratios in tap water (Figures 2a and 2b). Compared with simple interpolation without spatial and climatic parameters, this method incorporating auxiliary variables may provide an isoscape with greater spatial detail, and values for regions with sparse measurements can be better estimated. For example, the Zhao et al. (2017) tap water isoscape features concentric *bulls-eye* artifacts over poorly sampled regions of the Tibetan Plateau and northeastern China, reflecting the limited sampling and the interpolation method selected (Inversed Distance Weighting). In this study, the isotope maps over these regions express predictions based on variation in ancillary variables that are strongly correlated with physical isotope effects (especially elevation and latitude). Across China (Figures 2c and 2d), the optimal regression model exhibits residuals within ±10‰ for δ^2H and ±1‰ for δ^{18}O, respectively, in most areas, which is better than for previous precipitation isoscapes in China (e.g., Liu et al., 2008).

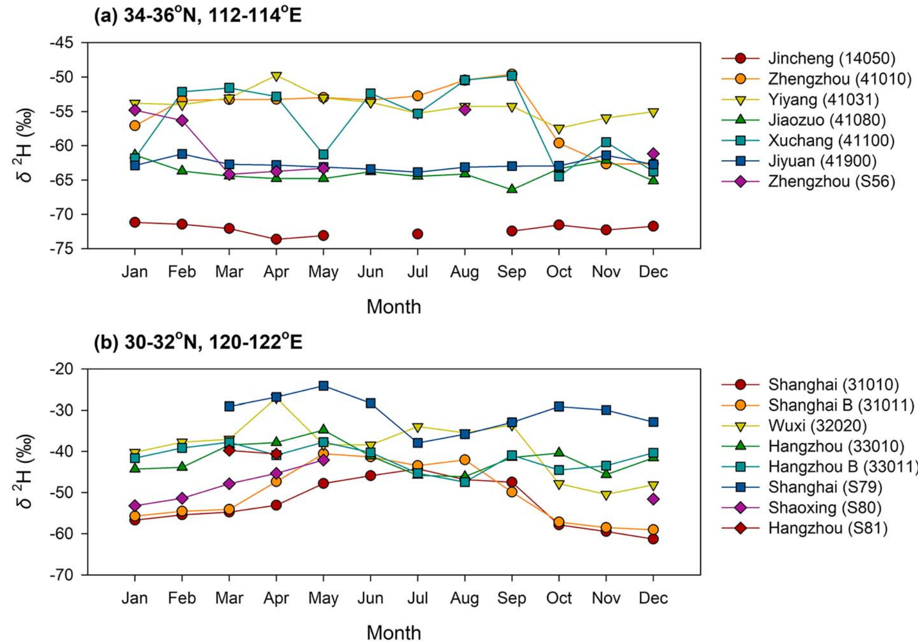

Figure 3. Monthly variation in tap water δ^2H at two typical areas: (a) 34–36°N, 112–114°E, and (b) 30–32°N, 120–122°E. The stations with IDs of five digitals (e.g., 14,050) are collected in this study, and the IDs of letter "S" and two digitals (e.g., S56) are acquired from Zhao et al. (2017). The geographic coordinates of the selected stations are shown in Table S1 in the supporting information.

Seasonal variation in tap water isotope ratios differs across the four major subregions of China (Figure S5). For most areas except south China, the isotope composition in tap water for each month is close to the annual mean value. Recently, Allen et al. (2018) suggested using sine functions to describe precipitation isotope seasonality in the estimation of isoscapes with seasonal cycles. Because of the limited seasonal variation in tap water isotopes for most parts of China, the sinusoid method does not work well for the data analyzed here. In south China, tap water samples are higher in spring and summer and lower in autumn and winter months.

To present monthly details of some well-sampled locations, we divided the sampling network into 2° (latitude) by 2° (longitude) grid cells and found two cells with ≥5 stations recording data for all 12 months. These are situated in north China (34–36°N, 112–114°E; Figure 3a) and south China (30–32°N, 120–122°E; Figure 3b), respectively. Most sampling stations in the north China group (Figure 3a) present a relatively stable trend, and those which do show changing values exhibit abrupt, stepwise shifts more consistent with a change in water source than with seasonal variation in isotope ratios of a single source (Jameel et al., 2016). In contrast, sites in south China exhibit larger, progressive fluctuations of tap water isotope composition (Figure 3b). For almost all the stations, higher values occur in spring or summer months. Generally, the focal sites in Figure 3 are consistent with and support the results of the regional summary (Figure S5).

3.2. Relationship Between Isotopes in Tap Water and Precipitation

We compare the tap water data directly with precipitation values estimated from the Global Network of Isotopes in Precipitation (IAEA/WMO, 2017) and other sources, using the Online Isotopes in Precipitation Calculator version 3.2 (Bowen, 2018) and the Regionalized Cluster-based Water Isotope Prediction model version 1.00 (IAEA, 2018; Figures 4 and S6). The tap water values reported here show spatial patterns that are similar to those of China's precipitation (Figure S7). The stable isotope ratios of tap water across the entire country correlate significantly with those of precipitation at the 99% significance level (Figures 4a and 4b). On an annual basis, determination coefficients (r^2) are 0.6156 (OIPC) and 0.6022 (RCWIP) for δ^2H and 0.4794 (OIPC) and 0.5213 (RCWIP) for δ^{18}O, respectively. Except for values from the Tibetan Plateau, with wide ranges of isotope composition observed and simulated in many studies (e.g., Gao et al., 2015; Tian et al., 2003; Yao et al., 2013), data from most subregions cluster in a relatively small region of H and O isotope space.

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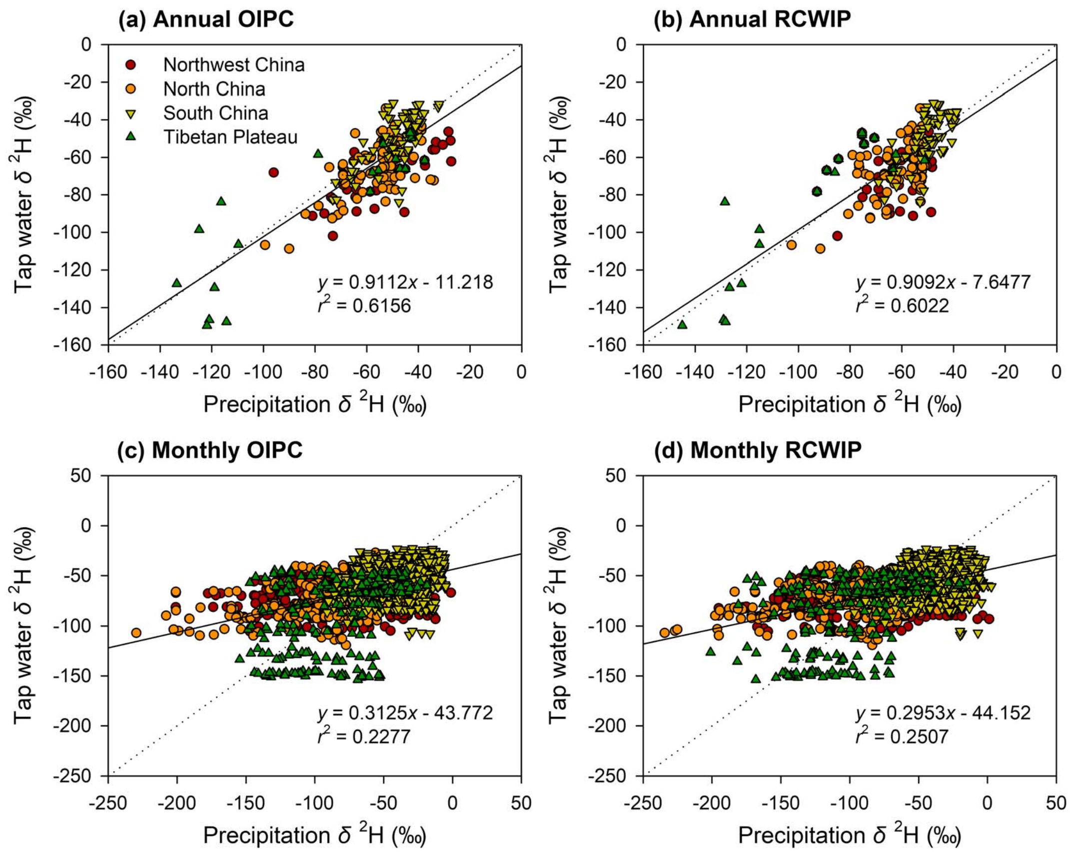

Figure 4. Correlations between δ^2H values of tap water and precipitation across China using annual and monthly (a and c) OIPC and (b and d) RCWIP data products. The dotted line is the line of equality ($y = x$), and the solid line is the linear least squares fit. The plot showing δ^{18}O is provided as Figure S6 in the supporting information.

Differences between annual average tap water and local precipitation isotope ratios show little regionalization, though there is a slight tendency for tap water in most regions except southern China to have lower values than local precipitation (Figures 5a and 5b). This is in contrast to results for U.S. tap waters, which showed strong regional differences relative to local precipitation (Bowen et al., 2007).

If the monthly data are considered instead of annual data (station mean), the correlation coefficients between stable isotope ratios in precipitation and tap water are much lower, although the positive correlation remains (Figures 4c and 4d). Comparisons between tap water and precipitation isotope ratios at a monthly basis may provide useful information on water source characteristics (Figures 5c and 5d). In south China, characterized by humid climate, as well as southern portions of north China, significant positive correlations in monthly basis data are widespread. This is not the case in northwest China, where the climate is arid and semiarid (Figure 1) and few significant correlations were observed.

On a monthly basis, the differences in stable isotope compositions between tap water and precipitation are not a constant for any of the subregions, reflecting the damping of monthly variability in tap water relative to precipitation (Figures 6 and S8). Seasonal difference offsets between tap water and local precipitation isotopes are the largest in northwest China (based on the range of monthly median values; Figure 6a), where tap water values are much lower than those of precipitation during summer and much higher than those of precipitation in winter. The seasonal characteristic in north China (Figure 6b) is similar to that in northwest China, but the tap water isotopes are closer to those in precipitation. In contrast, south China presents a different seasonal pattern of lower variability in tap-precipitation monthly offsets and slightly enriched heavy isotopes in summer tap water relative to precipitation. In the Tibetan Plateau (Figure 6d), a wide range of offsets reflects the complexity of topographic effects on isoscapes and diversity of water sources across this high elevation region (Yao et al., 2013). If the reference is changed from the

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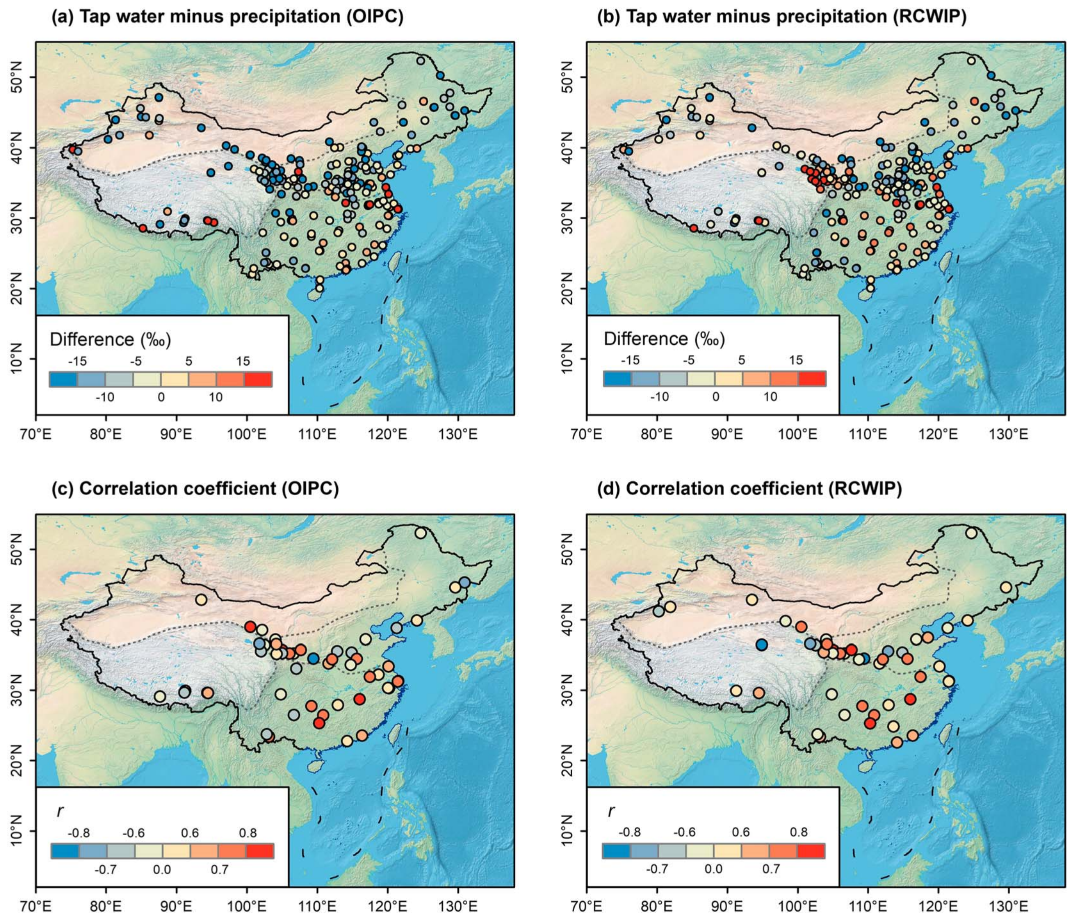

Figure 5. (a and b) Annual average tap water minus precipitation isotopes, and (c and d) correlation coefficient between monthly tap water and precipitation isotopes for δ^2H across China using OIPC and RCWIP data products. For correlation coefficients, only stations with statistically significant correlations (0.05 level) are shown. Subregion boundaries and base map as in Figure 1.

precipitation isotopes for each month to annual mean precipitation isotopes (Figure S9), the difference between the two types of water is quite stable all the year round, although a slight seasonal cycle can be seen for south China.

3.3. Water Source Signatures in Tap Water Isotopes

Water use records suggest that most of the sampled tap water originated from either groundwater or surface water sources. The seasonal variability observed here for tap water isotope compositions at some sites may therefore correlate with that in these water sources. If this is the case, tap water isotope monitoring provides a basis for linking public supply water with the isotopic properties of its source, providing, for example, the potential to identify changes in sources used or in source-water properties from measurements of samples collected at the point of use (e.g., Jameel et al., 2016).

Water resources management data provide context for the interpretation of isotope signatures in Chinese tap water samples (Figure S3). In China, surface water plays a dominant role in tap water supplies (80.8% in 2014, 81.4% in 2015, and 81.3% in 2016, respectively), while the contribution of groundwater is 18.3% in 2014 and 17.5% in 2015 and 2016; other water sources (e.g., sewage treatment and rainwater harvesting) contribute a very limited proportion (0.9% in 2014, 1.1% in 2015, and 1.2% in 2016; MWR, 2015, 2016, 2017). Interbasin transfer of water may influence the interpretation of isotope data in terms of water sources. In an isotope assessment in western United States (Good et al., 2014), the relationship between tap water and other water (rain and surface water) was applied to investigate possible sources of tap water. In China's surface water supplies, water transfer between larger drainage basins is relatively limited (3.9% in 2014 and 3.5% in 2015; MWR, 2015, 2016).

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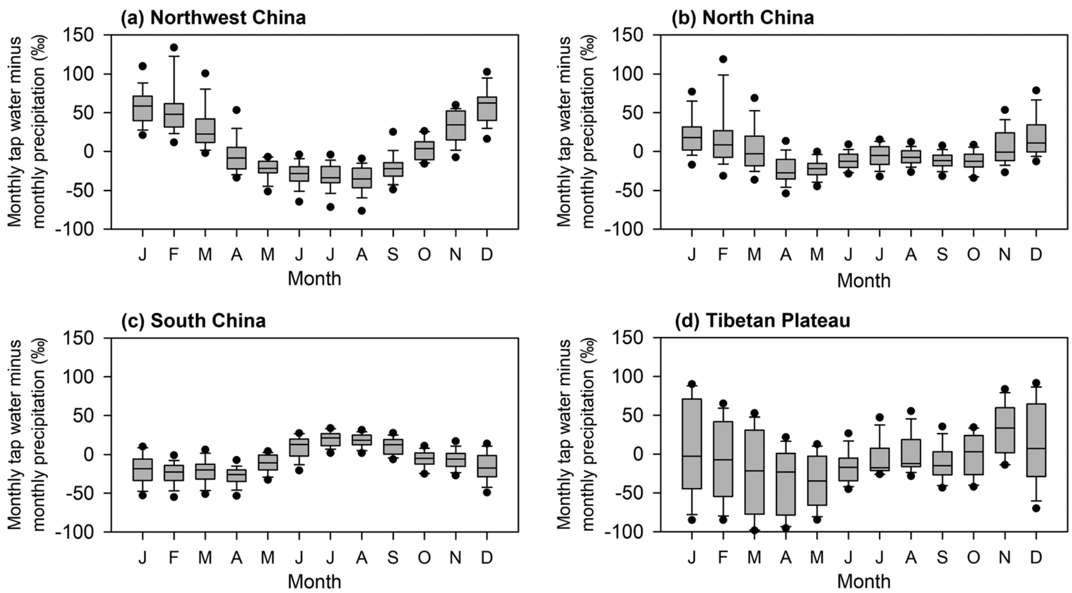

Figure 6. Isotopic variation of monthly tap water minus local monthly precipitation using OIPC-based δ^2H values in each subregion across China. Only stations covering all 12 months are used. The boxes represent 25th–75th percentiles, and the line through the box represents the median; the whiskers indicate the 90th and 10th percentiles; the points above and below the whiskers indicate the 95th and 5th percentiles. Plot showing RCWIP-based δ^2H is provided in Figure S8 in the supporting information.

In the absence of comprehensive databases of surface and groundwater isotope ratios it is difficult to make direct comparisons of source water and tap water values on a national scale. Given the ultimate derivation of surface and groundwater from regional precipitation (Peng et al., 2011; Wang, Zhang, Che, et al., 2016), however, precipitation isotope data represent a reasonable reference point for comparison of tap water data. Precipitation isotope ratios may be modified by a range of processes before water is sampled from taps. Natural flows within surface water systems may modify the spatial distribution of isotope ratios (Bowen et al., 2011; Dutton et al., 2005), and mixings within surface and groundwater systems may damp seasonal variation in precipitation isotope inputs (Sakakibara et al., 2017). Where ancient groundwater, recharged under different climate conditions, is used, the isotope ratios of extracted water may be quite different from

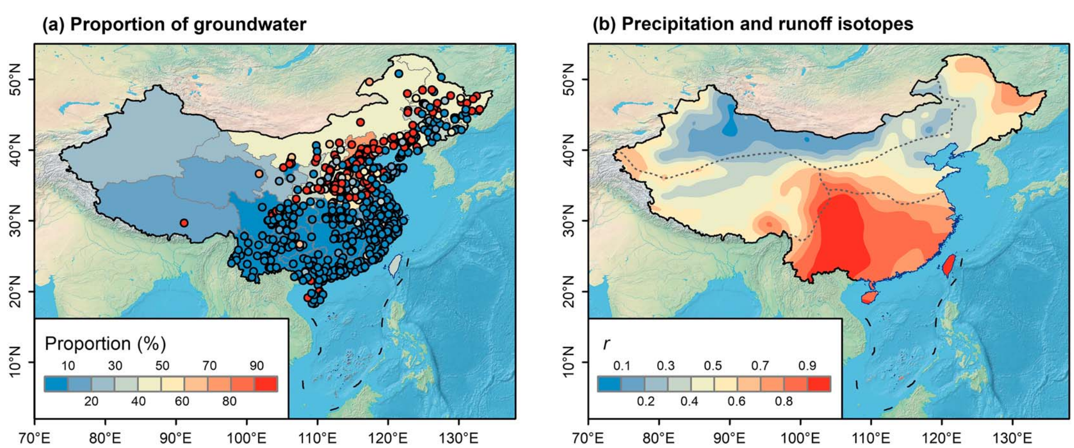

Figure 7. (a) Spatial distribution of the contribution of groundwater to local water supplies, from water management data across China. Shades denote the proportions on a provincial basis in 2015. The circles denote proportions for each water supplier (mostly on a city/prefecture scale) in 2015, where available. The grey curves are the provincial boundaries in China. Base map as in Figure 1. (b) Spatial distribution of correlation coefficient between monthly δ^2H in precipitation and runoff across China simulated using isoGSM during 1979–2007. Subregion boundaries and base map as in Figure 1. The map showing δ^{18}O is given in Figure S10 in the supporting information.

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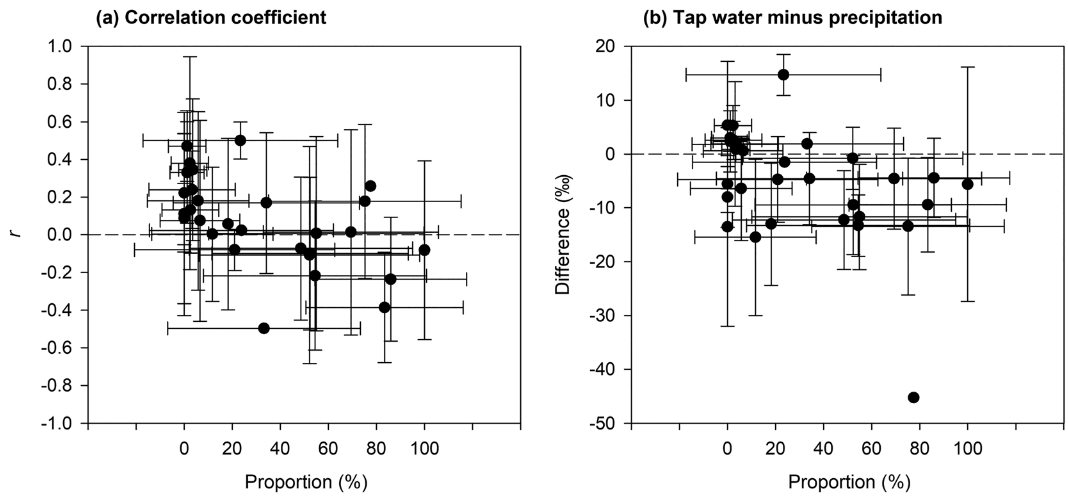

Figure 8. (a) Relationship of the correlation coefficient between monthly δ^2H in tap water and precipitation (*r*) with the proportion of groundwater in total water supply capacity on a provincial basis in China. (b) Difference between annual δ^2H values for tap water and precipitation (tap water minus precipitation) plotted against the proportion of groundwater in total water supply capacity on a provincial basis in China. The solid circles and error bars denote the arithmetic average and standard deviation for each province, respectively. Figures for δ^{18}O is shown in Figure S12 in the.

(usually lower than) modern precipitation values (Jiráková et al., 2011). This latter effect is likely to be of limited importance in the current study, since most groundwater sources in China's water supply are derived from shallow subsurface layers (85.8% in 2014 and 91.1% in 2015, respectively), instead of deep confined water (MWR, 2015, 2016).

Yearbook data across China (CUWA, 2016) suggest that water supplies in south China are almost totally supplied by local surface water, and those in the remote western provinces are also dominated by surface water. In contrast, the contribution of groundwater in some provinces of north China is generally high, with many areas entirely relying on groundwater as primary water source (Figure 7a). Comparing this pattern with those in Figure 5, it is clear that strong seasonal correlations between local precipitation and tap water isotope ratios occur primary in areas where the local water supplies are dominated by surface water.

The degree to which surface water isotope variability mimics that of precipitation depends on a range of factors, including basin geometry, water residence time, and surface water and groundwater interaction (Sakakibara et al., 2017). The correlation between isotope ratios in tap water and precipitation in south China (Figure 5) suggests that seasonal isotope variation in precipitation may be transferred through surface water systems to tap water in this region, but to test the plausibility of this conclusion, we evaluated output from an isotope-enabled GCM (Yoshimura et al., 2008; Figures 7b, S10, and S11). The simulated stable isotope composition in runoff exhibits a similar spatial distribution to that of precipitation across the study region (Figure S11). On a seasonal basis, correlations between precipitation isotope ratios and those of runoff are strong across most of south China (Figure 7b). In other regions the relationship is much weaker. This suggests that in south China, where climate is more humid (Figure 1) and the residence time of water within drainage basins may be lower, transfer of seasonal precipitation isotope variation through surface water systems to tap water is a plausible mechanism to explain tap water isotope variability. In northwest China, with arid climate (Figure 1), isotopes in surface water are not expected to be similar to those of local precipitation and strong seasonal tap water/precipitation isotope correlations would not be expected, nor are they observed.

The relationship between water source and correlation coefficient (or difference) between isotopes in tap water and precipitation can also be presented on a provincial basis (Figures 8 and S12). In provinces with a very low contribution of groundwater in local water supplies (mostly distributed in south China), the stable isotope ratios of tap water are quite similar to those of precipitation. However, for areas dominated by groundwater use, tap water values are uncorrelated with and usually isotopically depleted relative to precipitation.

Considering the geographical pattern of precipitation isotope ratios across arid northwestern China (Liu et al., 2008; Wang, Zhang, Hughes, et al., 2016), runoff (through surface or subsurface flow) of isotopically light, high elevation precipitation, and its use in adjacent basins can explain the observed pattern. Many of the groundwater-dominated sites occur in northwest China, where population is mostly distributed at low-lying oases and available water resources originate primarily from precipitation in nearby mountains. Winter precipitation in this region is very limited, and amount-weighted precipitation isotope ratios are higher than the tap water values on an annual basis (Figures 5a and 5b). Tap water in this region is also characterized by limited seasonal isotopic variability (Figure S5a), despite large variations in precipitation isotope ratios (Wang et al., 2017; Zhang & Wang, 2016, 2018), leading to the largest observed seasonal offsets between tap water and precipitation (Figure 6a). This argues for a relatively weak interaction between local precipitation and surface water in the systems supplying the water, consistent with the dominant use of groundwater or water from river basins with a relatively long residence time.

4. Conclusions

The spatial pattern of tap water isotope ratios is jointly influenced by natural process and human activities, meaning that tap water isoscapes can be useful for the study of hydrology, water management, ecology, food science, forensic science, and other domains. Stable isotope compositions of tap water can reflect water origin, but large-scale relationships between water source types and tap water isotope ratios have not adequately been described. Data from a newly established nationwide network documenting stable hydrogen and oxygen isotope ratios of tap water across China are consistent with other studies in showing that on an annual basis, and across the large geographic study area, the isotope ratios of tap water positively correlate with those of local precipitation. Seasonal and monthly variation in tap water at individual sites and averaged within provinces, however, shows contrasting patterns across China that appear to be related to the types of water sources exploited. Widespread use of surface water from systems with relatively fast interaction between precipitation and surface water is reflected in more variable isotope ratios of south China tap water, which are correlated with seasonal precipitation isotope ratio shifts. The observed pattern suggests that existing water infrastructure in this region relies on hydrological systems with limited storage capacity, potentially enhancing its susceptibility to short-term fluctuations in precipitation water inputs. In contrast, relatively invariant tap water isotope ratios elsewhere in China, which are not correlated with seasonal precipitation water isotope variation, can be attributed to use of groundwater or water from river basins with longer storage times. In these locations long-term limitations on precipitation recharge is likely the primary risk to water supply sustainability. We suggest that the general tap water isotope signatures identified here could be widely applied to characterize water supplies and associated sustainability challenges in different regions worldwide.

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