

STABLE ISOTOPES OF HYDROGEN AND OXYGEN IN RUNOFF WATERS IN NORTHWESTERN SICHUAN OF SOUTHWESTERN CHINA

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Abstract. In studying the hydro-ecological conditions in northwestern Sichuan of southwestern China, an area at the edge of the Qinghai-Tibet Plateau, stream waters were extensively sampled and analyzed for the stable hydrogen and oxygen isotope ratios ($\delta^2\text{H}$ and $\delta^{18}\text{O}$) which are often excellent indicators for a water body genesis and evolution. The heavy isotopes of hydrogen and oxygen were found to be, in general, gradually depleted from east to west in the region; the phenomenon is attributed mainly to continentality based on the assumption that the isotopic signatures in the precipitations were preserved in the stream waters. The $\delta^2\text{H}$ and $\delta^{18}\text{O}$ data from the Yangtze River basin were found to fall well on the global meteoric water line (GMWL), indicative of the fact that the runoff waters in this basin were primarily of meteoric origin with a regional weak evaporation prevailing during our sampling time. However, the isotopic data from a relatively small part of the area in the north belonging to the Yellow River basin are characterized by the appreciably flatter slope of the local water isotope line and lower d -excess values, suggesting that the stream waters had undergone significant evaporation due to kinetic fractionations of isotopes in a more arid climate. The stable isotope data were also applied in calculating water mixing for some selected rivers. This study represents a snapshot study of the area's hydro-ecology; more systematic studies are required in better understanding the seasonal hydrological variations.

Keywords: *meteoric water, hydro-ecology, $\delta^2\text{H}$ and $\delta^{18}\text{O}$, three-river headwaters (TRH), southwestern China*

Introduction

The hydro-ecological conditions of the Qinghai-Tibet Plateau are of critical importance to several Asian countries including a large part of China. The three-river headwaters (TRH) region, located in southern Qinghai province (31°39'–36°16'N, 89°24'–102°23'E), is known as China's water tower where China's three largest rivers (among the world's largest ones) originate. The Big Three, i.e., the Yangtze River, the Yellow River and the Lancang River, obtain, respectively, 25%, 50% and 15% of their water from the TRH region (Zhao et al., 2010). Because of the fragile primordial ecosystem and due to the increased anthropogenic activities during the past several decades, the eco-environmental problems in the region such as grassland degradation, wetland shrinkage and soil erosion started to accelerate since the 1980's and presently serious and immediate attentions are called for (Li et al., 2012; Gao et al., 2013). Li et al. (2012) found that the water loss due to evapotranspiration in the TRH region

increased 11.6 mm annually between 1980 and 2000, meanwhile exhibiting an accelerating trend. It is therefore important to understand the hydro-ecological system of the region in order to conserve and restore its environmental conditions. However, this vast area is generally not easily accessible due to high altitude, harsh climate, extreme remoteness and lack of roads.

The northwestern Sichuan area studied in this work, with gradually decreasing elevations southeasterly, forms the easternmost part of the Qinghai-Tibet Plateau. This area can be divided into two sub-regions: the Northwestern Sichuan Plateau and the Southwestern Sichuan Mountainous Terrain (*Figure 1*). The former is characterized by vast and hummocky plateaus with high elevations (3,500 to 4,000 m or higher); the latter consists of numerous high mountain ranges separated by deep valleys with elevation differentials as much as 3,000 m, known generically as the Hengduan Mountains. The river systems and wetlands in the region are intrinsically connected with those in the upper part of the TRH region. These wetlands provide many important ecological services such as lessening the greenhouse effect by taking up and storing carbon, and are invaluable environmental and ecologic resources (Avis et al., 2011). The hydro-ecological condition of this region is an important integral part of the TRH regional ecology; a thorough understanding of this region may hold the key for the entire TRH region. These wetlands have experienced significant degradation over the past 4 decades and have shrunk by some 29%, and the wetland ecosystems are increasingly fragmented at an accelerated pace (Li et al., 2012; Qi and Li, 2007; Wang et al., 2007) mainly due to the increased anthropogenic activities in the region (Li et al., 2012). In order to stabilize and possibly reverse the ecological conditions of the region, the regional hydrology including the sources of the precipitations, the general water cycle processes of the drainage basins must be understood.

The stable isotopes of oxygen (^{18}O) and hydrogen (^2H) can be accurately determined (usually as isotope ratios) with modern technologies (Richelle et al., 2004; Bowen et al., 2005a; Paul et al., 2007), and have become some of the most widely used tracers in drainage basin hydrological studies (Clark and Fritz, 1997; Kendall and McDonnell, 1998; Buttle and McDonnell, 2004). In particular, stable isotope fractionation during precipitation processes are well understood, both ^{18}O and ^2H in precipitations vary spatially and temporally depending on the moisture sources and the environmental conditions (e.g., relative humidity and temperature) during cloud formation and rainfall. The isotope ratios in precipitations, their seasonal variations at a given location, and their distribution over a region often reflect the relevant meteorological and geographical conditions such as local temperature and temperature variation, distance from the coast (continentality), latitude and altitude, amount of precipitation and duration (Clark and Fritz, 1997).

Due to their spatial and temporal variability in meteoric waters, these isotope ratios ($\delta^2\text{H}$ and $\delta^{18}\text{O}$ values) have been used to identify sources of water such as atmospheric moisture (Liu et al., 2010; Sjostrom and Welker, 2009; Vachon et al., 2010; Welker, 2000), groundwater recharge (Abbott et al., 2000; Blasch and Bryson, 2007; Adomako

et al., 2010), and plant water uptake (Brooks et al., 2010). In this work, the surface stream waters in the northwestern Sichuan region, an area covering part of both the Yangtze River and the Yellow River drainage basins (divided by the Chazhengliangzi Mountain in Hongyuan, Sichuan) were extensively sampled and analyzed for $\delta^2\text{H}$ and $\delta^{18}\text{O}$. Most rivers in the region including the Minjiang, Jialing, Fuhe and Dadu flow from north to south or from west to east to the Yangtze River (Yangtze River drainage basin), whereas the Heihe and Baihe rivers from south to north to the Yellow River (Yellow River drainage basin). The main objective of this study is to enhance our understanding of the regional hydrology of the study area through characterizing the isotopic variation patterns of the runoff waters and identifying the main influential factors including the geographical and meteorological parameters. As the TRH region is difficult to access and its hydrology study is still at a relatively early stage, the current work may help fill some of the gaps in the study of the hydrological-ecological conditions of the entire TRH region.

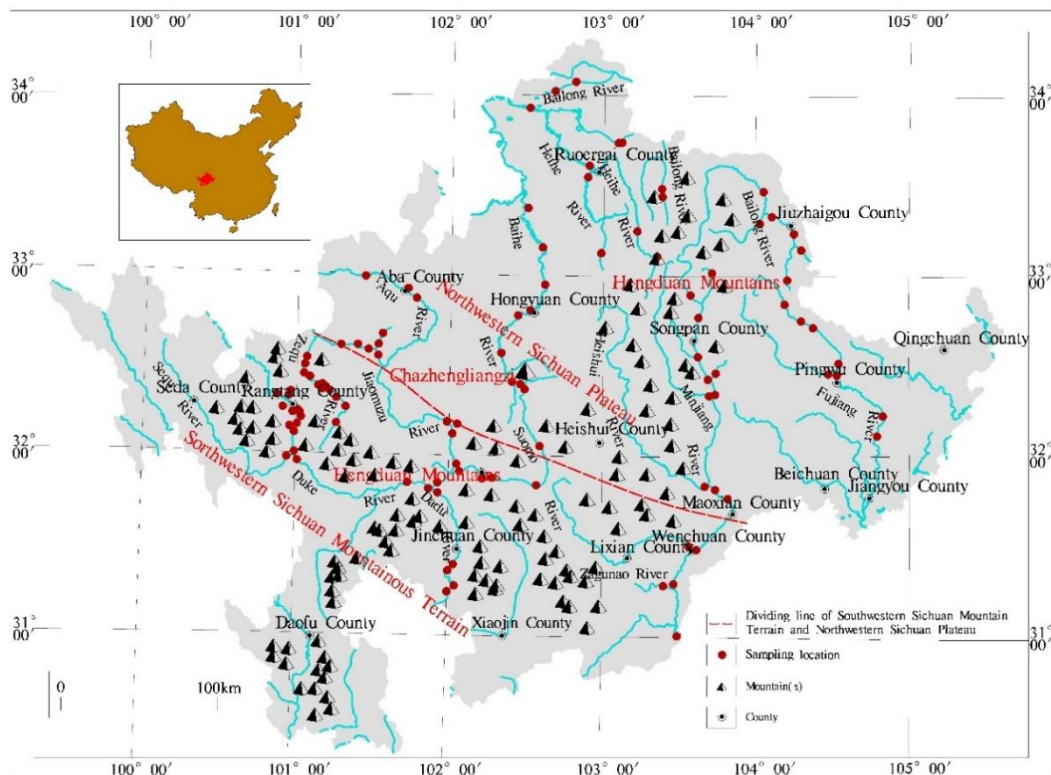


Figure 1. Map of the main rivers in the study area with sample locations

Materials and methods

Study area

The study area is at the edge of the Qinghai-Tibet Plateau and is generally known as the Northwestern Sichuan Plateau. Water samples were taken from locations in 28 counties: 13 alpine counties including Rangtang, Aba, Songpan, Ruergai and

Hongyuan in Aba Prefecture, and 18 mountainous counties including Seda in Ganzi Prefecture, Wangcang in Guangyuan City, Hanyuan in Ya'an City, and Pingwu in Mianyang City, covering a total area of 149,000 km². In total 86 stream water samples were collected for $\delta^2\text{H}$ and $\delta^{18}\text{O}$ analysis from 7 river systems (*Table 1*). Owing to logistic difficulties, the sample campaign had to be conducted in two consecutive summers of 2010 and 2011. Approximately half of the samples were taken in July of 2010 and half in July and August of 2011. The climatic conditions of the sampling area from June to August are summarized and compared in *Table 2* for 2010 and 2011 (data were downloaded from Tutiempo Network website, <http://en.tutiempo.net/>). From *Table 2*, it is clear that the climatic conditions of the area were closely similar between 2010 and 2011 during the sampling months. It is noted that August 2011 appeared to be drier than August 2010; however, this difference would not impact our discussions because the dry season essentially occurred after our sampling campaign.

Table 1. Number of river water samples taken from various river catchments

River Catchment	No. of Samples
Fujiang	8
Baishui	6
Heihe	7
Bailong	4
Minjiang	16
Duke	13
Zequ	8
Baihe	6
Suomo	5
Jiaomuzu	13
Sum	86

Table 2. Comparison of summer climatic conditions between 2010 and 2011 at selected locations in the study area

	Average temperature (°C)		Precipitation (mm)		Number of rainy days	
	2010	2011	2010	2011	2010	2011
Songpan, 32.65N 103.56E, Elevation 2852 m						
June	12.9	14.1	125.2	113.27	23	21
July	16.9	15.0	101.1	120.65	20	19
August	16.4	15.1	118.39	39.36	17	12
Seda, 32.28N 100.33E, Elevation 3896 m						
June	9.5	9.6	153.67	189.22	25	22
July	12.4	10.2	65.78	197.63	16	25
August	10.8	9.9	99	62.98	19	12
Daofu, 30.98N 101.11E, Elevation 2959 m						
June	15.4	15.8	157.21	118.12	22	21
July	17.9	15.9	111.76	121.66	17	20
August	16.9	16.2	108.2	63.25	17	9

Ganzi, 31.61N 100E, Elevation 3394 m						
June	13.5	13.7	157.99	121.91	26	23
July	16.2	14	71.65	138.69	19	23
August	15.3	13.8	89.9	71.36	21	13
Ma'erkang, 31.9N 102.23E, Elevation 2666 m						
June	15.1	15.4	155.96	169.17	27	21
July	17.7	15.5	180.6	157.22	24	22
August	16.9	16.5	145.31	42.42	24	11

Data from: <http://en.tutiempo.net/>

Methods

The sampling locations are listed in *Table 3* and shown on the map in *Figure 1*. We have exercised rigorous and systematic quality assurance measures in the field at various levels to ensure the samples were taken and labeled appropriately. We conducted the sample taking following strictly the procedures set out in the Quality Assurance Manual for Environmental Water Monitoring (Zhang, 1994). The water samples were taken at a depth of a few centimeters below the water surface to ensure that the river water was fully mixed to avoid any potential surface isotope fractionation effect due to evaporation. When sampling from a water body where two streams met, samples were taken only in the fully mixed zone, 200 m or further downstream from the meeting point. During sampling, observations of water quality, river flow rate, vegetation on both banks, rock composition and geological features were also made. More details on sampling was described elsewhere (Shi et al., 2016).

Table 3. Summary of measured sample isotopic data ($\delta^2\text{H}$ and $\delta^{18}\text{O}$ values), associated location, and other information

Catch.	Location	Elev(m)	Temp(°C)	$\delta^2\text{H}$	$\delta^{18}\text{O}$	d-excess
Fujiang River	5 km north of Xiangyanzhen	648	25.0	-65.5	-9.74	12.4
	2 km south of Pingwubazi	716	25.0	-66.8	-9.97	13.0
	3 km north of Pingwu	744	17.1	-64.7	-9.84	14.1
	1.5 km from Dapingli near Huanglong	899	17.3	-64.7	-9.83	13.9
	2 km from Liangheshui (Jiuzhaigou direction)	890	18.2	-65.3	-9.76	12.7
	500 m downstream from Hua'neng Hydro Station	1121	12.6	-70.4	-10.54	13.9
	Near Shuiniujia, Baima Township	2110	16.6	-65.8	-9.68	11.6
Baishui River	Yangdong River Divide downstream near source	2387	13.2	-67.3	-10.07	13.3
	At Wujiao, Jiuzhaigou	1598	16.4	-66.7	-10.18	14.8
	2 km from Shuanghe (Wenxian direction)	1430	17.1	-77.0	-10.96	10.7
	2.5 km south of Jiuzhaigou	1400	17.6	-75.8	-11.07	12.8

	1 km upstream Baihe Township (Heihe)	1562	14.5	-76.5	-11.16	12.8
	Under Yanlicun Bridge, Baihe Township	1789	13.8	-76.7	-11.19	12.8
	5 km upstream from Jiuzhaigou mouth	2179	11.3	-78.7	-11.49	13.2
Heihe River	10 km north of Songpan and Ruoergai border	3693	19.3	-83.9	-11.55	8.5
	25 km southeast of Rouergai	3522	20.0	-83.7	-11.32	6.8
	1 km west of Heihe divisional ranch	3429	19.9	-80.8	-10.36	2.1
	Creek, front of Axi Government Building	3493	19.5	-83.6	-11.35	7.2
	4 km northwest of Ruoergai	3437	18.0	-81.7	-10.23	0.2
	Heihe divisional ranch	3442	21.2	-80.5	-10.03	-0.3
	Maiwaxiang, Hongyuan	3555	21.6	-89.0	-12.67	12.4
Bailong River	2 km downstream Jiangza township	3019	14.8	-79.7	-11.21	10.0
	Upstream Hongxing township, near source of Bailong River	3186	14.1	-81.0	-11.15	8.2
	In Baozuo Township yard	3022	14.1	-83.0	-11.78	11.3
	500 m downstream from Baozuo Township yard	3009	20.0	-83.8	-11.85	11.0
Minjiang River	8-10 km downstream from Minjiang source	3316	13.6	-84.7	-12.16	12.5
	5 km from Chuanzhusi (Ruoergai direction)	3021	14.8	-83.3	-11.79	11.1
	8 km downstream from Chuanzhusi	2921	15.8	-86.2	-12.25	11.8
	Munigou mouth	2706	13.0	-85.6	-12.16	11.7
	Under bridge near Daxing'gou mouth	2581	14.5	-89.1	-12.65	12.1
	Minjiang mainstream	2568	17.1	-85.3	-11.99	10.6
	Minjiang mainstream	2491	17.1	-86.5	-12.33	12.2
	Mouth of Xiaohegou	2487	19.6	-81.8	-11.56	10.8
	Downstream from Heishuihe	1650	18.4	-87.1	-12.30	11.3
	Minjiang mainstream, downstream of hydro station	1653	17.6	-81.7	-11.69	11.8
	Minjiang mainstream, under Juanmenxiang Bridge	1649	20.5	-84.5	-12.11	12.4
	Minjiang mainstream, behind Zhongxing	1353	19.8	-83.4	-11.90	11.8
	Upstream of Zagu'nao River Hydro Station	1384	16.6	-90.6	-12.69	10.9
	Minjiang mainstream at Yinxing township	991	18.7	-85.0	-11.98	10.8
Shaotang River tributary	898	24.5	-83.7	-12.07	12.9	

	Minjiang mainstream	711	21.8	-83.1	-11.96	12.6
Duke River	Gangmudarewo Village	3298	13.4	-98.3	-14.02	13.9
	Rangkegare Village	3253	15.3	-98.7	-14.01	13.4
	Gangmudake Village	3205	12.9	-97.7	-13.76	12.4
	Rangtangdamuda Village	3277	13.0	-98.8	-13.80	11.6
	Sequ River at Heiqiao	3008	15.5	-107.5	-14.87	11.4
	Heiqiao, Rangtang	3007	14.0	-99.1	-13.97	12.6
	Ergewu Village, Heiqiao	3009	14.8	-101.2	-14.22	12.6
	Guanyinqiao, Jinchuan	2078	14.0	-103.3	-14.46	12.4
	Baiwan, Ma'erkang	2300	17.7	-101.0	-14.41	14.3
	Gangmudarewo Village	3382	9.1	-99.8	-13.85	11.0
	Gangmudarewo Village	3341	12.3	-100.4	-14.11	12.6
	Rangkegare Village	3264	9.6	-107.4	-14.71	10.3
	Gangmuda'angke Village	3193	12.0	-106.3	-14.86	12.6
	Zequ River	Xumuda Village, Shangrangtang township	3480	13.0	-100.7	-13.96
Upper Rangtang		2869	13.1	-100.9	-14.09	11.8
Middle Rangtang		3489	13.7	-101.1	-14.10	11.8
Yigenmenduo Village, Middle Rangtang		3412	13.4	-101.8	-14.21	12.0
Gaduo, Rangtang		3480	14.9	-100.1	-14.23	13.8
South Muda, Rangtang		3362	15.9	-100.3	-13.98	11.6
Rongmuda Village		3292	18.5	-100.5	-14.32	14.1
Yigenmenduo Village		3411	13.2	-101.5	-13.44	6.0
Baihe River	Xigou, Hongyuan	3635	11.0	-94.1	-13.29	12.2
	Anqu, Hongyuan	3519	13.5	-92.5	-13.46	15.2
	Hongyuan	3491	15.3	-96.8	-13.55	11.6
	Baihe at Tangke	3463	17.3	-87.6	-11.81	6.9
	5 km upstream Waqie	3462	16.4	-88.8	-12.16	8.5
	Baihe, upstream Amu Township	3477	17.8	-87.7	-11.90	7.4
	Rangkou, Hongyuan	3574	11.5	-96.3	-13.39	10.9
	Laomaokang Village	3225	10.8	-95.9	-13.21	9.8
	Suomo, Ma'erkang	2530	10.8	-100.1	-13.71	9.6
	Ma'erkang	2645	11.4	-96.5	-13.98	15.3
	Songgang, Ma'erkang	2495	13.3	-96.1	-13.83	14.5
Jiaomuzi River	Jia'erduo (Aqu River)	3430	17.5	-84.1	-11.93	11.4
	Anjiang (Aqu River)	3248	17.8	-87.1	-12.38	11.9
	Kehe Village (Aqu River)	3070	15.4	-91.3	-12.73	10.5
	Kuasha, Aba	2985	15.4	-92.9	-12.91	10.4
	Kuasha (Aqe River)	2985	13.5	-85.7	-12.20	12.0
	Kuasha (Ma'erqu River)	2985	15.0	-86.2	-12.38	12.9
	Gabo, Ma'erkang	2536	15.4	-92.1	-13.01	12.0

Gabo	2496	15.3	-96.6	-13.84	14.1
Jiaomuzu, Ma'erkang	2500	15.8	-92.0	-13.09	12.7
Baiwan, Ma'erkan	2344	16.1	-92.0	-13.11	12.9
Shuangjiangkou, Ma'erkang	2318	17.2	-99.5	-13.82	11.1
Kehe Village	3064	20.0	-100.1	-13.83	10.6
Gabo	2469	13.1	-96.7	-13.95	15.0

Notes : Catch. - Catchment; Elev - Elevation; Temp - Temperature

Hydrogen and oxygen isotopes ($\delta^2\text{H}$ and $\delta^{18}\text{O}$ values) were determined by the Sichuan Provincial Key Isotope Laboratory at the Chengdu University of Technology, using high temperature conversion elemental analysis – isotopic ratio mass spectrometry (TC/EA-IRMS) with the instruments supplied by Thermo Fisher (TM Flash 2000 coupled with a mass spectrometer, MAT-253). The accuracy of the isotopic ratio analysis relative to the Vienna-Standard Mean Ocean Water (VSMOW) was determined to be $\pm 0.2\text{‰}$ and $\pm 2.0\text{‰}$ for $\delta^{18}\text{O}$ and $\delta^2\text{H}$, respectively. Water samples were analyzed without pre-treatment. For more details about the analytical procedure used, the reader is referred to Burgoyne and Hayes (1998), Hoefs (2009), and Richelle et al. (2004).

Results and discussion

$\delta^{18}\text{O}$, $\delta^2\text{H}$, *d*-excess and local meteoric water line (LMWL)

The measured stable hydrogen and oxygen isotope δ -values are presented in *Table 3* together with the associated information of sample locations and the relevant river catchments/drainage basins. Because altitude and temperature are two of the most important parameters governing isotopic fractionations in precipitation (see discussions below), these two parameters were therefore also measured and are included in *Table 3*. Since the climatic conditions in 2010 and 2011 were very similar during the sampling times (*Table 2*), the measured isotopic data were analyzed and discussed without distinction of the sampling year. The parameter *d*-excess can convey additional information about the water bodies studied, therefore the derived *d*-values are also tabulated in *Table 3*.

The measured $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values range from, respectively, -14.87‰ to -9.68‰ and -107.5‰ to -64.7‰ for the water samples taken from the various river systems in the study area. When the data are plotted in a $\delta^2\text{H}$ – $\delta^{18}\text{O}$ diagram as shown in *Figure 2*, the data points align well with the GMWL as defined by Craig (1961), suggesting that the area's stream water was likely of meteoric origin primarily. Recently, Liu et al. (2014) studied and reported the isotopic compositions of precipitations in China based on the data collected from 29 stations including two within the present study area: 1. Maoxian (MX), elevation 1,826 m, located near the southeast border of the study area, and 2. Gonggashan (GG), elevation 2,950 m, located at the southwest border of the study area.

The reported $\delta^2\text{H}$ values are from -83.3 to 8.5 with a mean of -49.5 for MX, from -147.8 to 1.4 with a mean of -76.5 for GG; and $\delta^{18}\text{O}$ from -11.98 to 0.31 with a mean of -7.77 for MX, from -19.50 to -2.35 with a mean of -10.82 for GG. These data compare very well with our data of samples from the same general areas (see *Figure 2* and *Table 2* for data from Fujiang River and Duke River, respectively) considering that the data by Liu et al. (2014) are monthly precipitation-weighted averages while our data are one-time values. Therefore, it is assumed in this work that the samples taken from the streams represent the local precipitations in terms of isotopic compositions. Our data span relatively narrow ranges when compared to the China national data of $\delta^{18}\text{O}$ (-29.47 to -2.75 ‰) and $\delta^2\text{H}$ (-229.6 to 45.4 ‰) (Liu et al., 2014). This is, perhaps, expected due to the fact that the area is located in the Asian monsoon region and our sampling times coincided with the monsoon prime times (Liu et al., 2014). The area was receiving rainfalls almost daily during our sampling times with the total precipitations of 100-200 mm/month. As the sampling was conducted during the warmest time of a year, the melting of alpine snow and ice was also expected at the peak time. There is usually no isotopic fractionation during snowfalls as the solid phase does not exchange isotopes with the atmospheric moisture; and snowpacks tend to conserve the isotopic composition of the cloud moisture with an elevated d-excess value (Gat et al., 2001). The value of d-excess varies locally and was shown to correlate with the physical conditions such as relative humidity, air temperature and sea surface temperature of the oceanic source area of the precipitation moisture (Merlivat and Jouzel, 1979). Thus the waters would be expected to be more enriched with heavy isotopes and with an elevated d-excess if the streams were heavily recharged by meltwaters, and this is not the case in this work with Aqu River as possibly an exception (*Table 3*). Because of the heavy and frequent rainfalls, the ground water contribution to the streams is also considered to be insignificant. The “catchment isotope effect” (Rozanski et al., 2001) in the region during the sampling times was such that the isotopes of the stream waters may be considered having essentially the same composition as those in the precipitation waters.

As noted earlier that the isotopic ratios ($\delta^2\text{H}$ and $\delta^{18}\text{O}$ values) are distributed quite evenly along the GMWL in *Figure 2* (when the data from the Yellow River basin are excluded, see below for details). This may indicate that, as the stream waters were primarily of meteoric origin, the rain moisture may originate primarily from a single remote location and rainfalls formed along the way with different distances of moisture transport. Recently, Wang et al. (2016) used the entropy analysis method of stable isotopes in precipitation to study the monsoon systems in China, found that in southern China, the water vapor is mainly supplied by the Indian monsoon. “one stream of the Indian monsoon (II) passes eastward from the southwestern border of China and then turns to the northeast of the south central China before reaching eastern China, where its intensity weakens after producing heavy rainfalls.” However, the authors also recognized the impeding effect of the Himalayas that impedes the movement of the Indian monsoon from the south to north and the movement of the winter monsoon from the north to south (Wang et al., 2016).

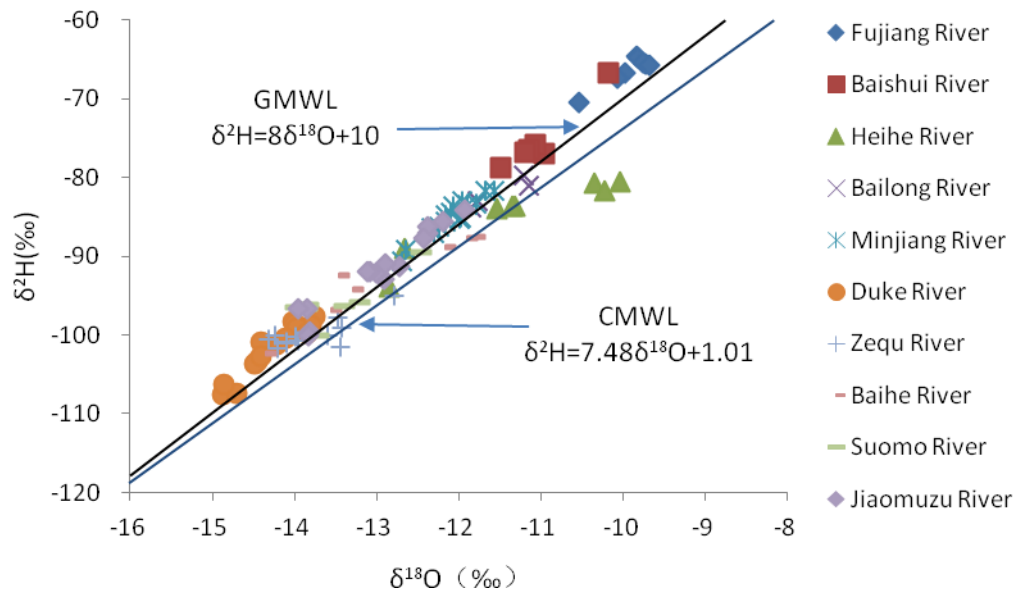


Figure 2. Diagram of hydrogen and oxygen isotopic ratios ($\delta^2\text{H}$ vs. $\delta^{18}\text{O}$) for all water samples analyzed in this study. Note that the upper line represents the global meteoric water line (GMWL), and the lower line represents the China meteoric water line (CMWL)

These observations are also supported by the d-excess data. In the study area the d-values span a relatively large range from -0.25 to 15.33 with a mean of 11.30 (Table 3), which is close to the global average (Rozanski et al., 1993). In examining the d-values, no apparent anomalies nor any reasonable patterns (e.g. decreasing values when further away from alpine glaciers) could be found, suggesting a predominantly well-mixed, relatively homogeneous source for the river waters, and contributions from the melting of alpine snow and ice were likely insignificant. Lower d-values relative to the surrounding areas were found in the Heishui County region (Figure 3); this may be attributed to high evaporation in the region where abundant sunshine is available throughout the year and the landscape is characterized by interweaved high mountain ranges and deep valleys (elevation differential as much as 1,000–2,000 m). This unique combination of climatic and geographic conditions gave result to widely uneven precipitations and large temperature swings between day and night in the Heishui region. However, the rather low d-values may not be fully counted for by the evaporation effect; it is suspected that some water component previously affected by evaporation and stored in the riparian zone might have been pushed into the streams by the newly fallen precipitation through infiltration and hyporheic flow (National Research Council, 2002). The lower d-values found in the Heihe River and Beihe River area will be further discussed in a later section.

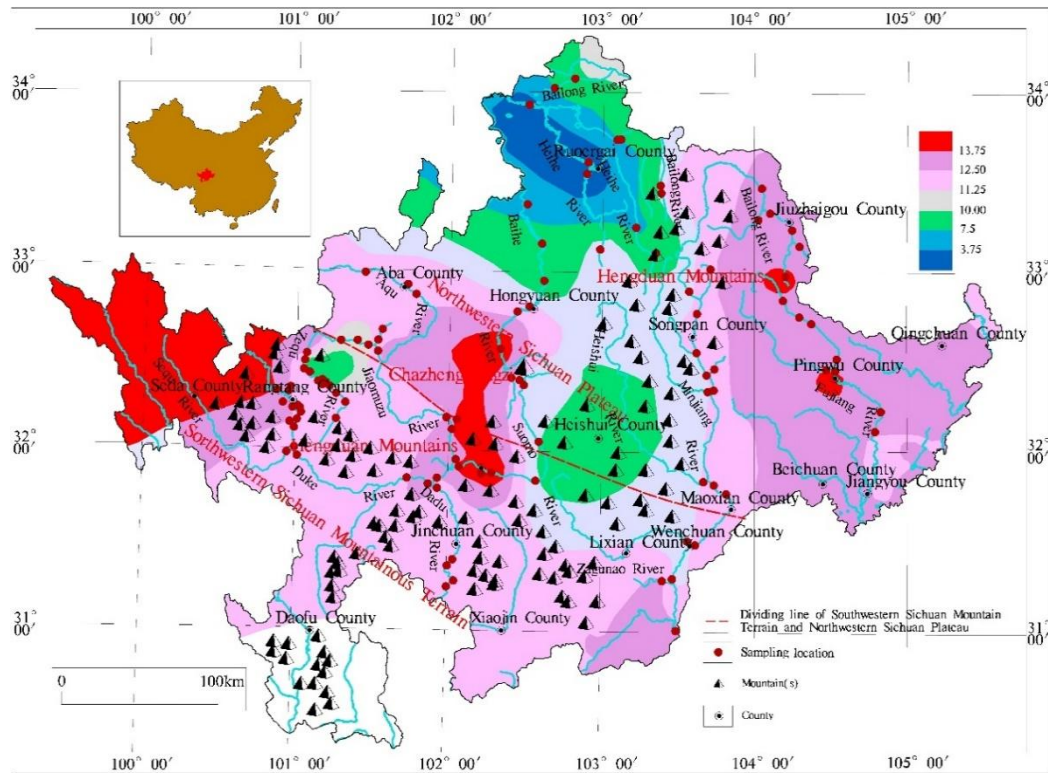


Figure 3. Spatial distribution of *d*-excess in the study area

Effects of temperature, altitude and continentality

The stable isotope ratios $\delta^2\text{H}$ and $\delta^{18}\text{O}$ are shown in *Figures 4* and *5* as mapped contours through interpolation. In examining the contours in both figures, it is noted that there is a general trend: both ^2H and ^{18}O become progressively more depleted from east to west across the region. The study area is located inland at the west margin of the south China (SC) region and near the Tibet (TP) and north China (NC) regions according to Liu et al. (2014). The precipitation moisture of this area has a complex origin and is composed of components from the South China Sea, the Pacific and Indian Oceans, and local evaporation and circulation. This complex situation may be visually appreciated by the back-trajectories of the air masses on June 30 and July 4, 2010 and 2011 from two selected locations near the center of the studied area depicted in *Figure 6*, produced using the HYSPLIT transport and dispersion model (Stein et al., 2015; Rolph, 2016). In this region, the air mass has a general trend of undergoing progressive rainout of heavy isotopes during the pole-ward (latitude effect) and westward (continental effect) atmospheric transport (Liu et al., 2014). However, the southwesterly monsoon from the Indian Ocean may be effectively obstructed from reaching here by the Qinghai-Tibet Plateau, this is supported by studies such as Liu et al. (2007, 2014). It is postulated that the moisture for precipitation in this region originated primarily from the Pacific Ocean and the South China Sea during a period shortly prior to the sample campaigns. That is, the moisture air masses traveled mainly from east to west, isotopically enriched rain

formed and fell from diminishing moisture mass and the residual moisture became progressively depleted in heavy isotopes with respect to earlier rains from the same cloud. This westerly continental effect explains well the observed isotope ratio contours seen in *Figures 4* and *5*, where no apparent pole-ward isotopic fractionation pattern is observed which is perhaps due to the ineffectiveness of the southwesterly monsoon in penetrating the Qinghai-Tibet Plateau (Liu et al., 2007, 2012).

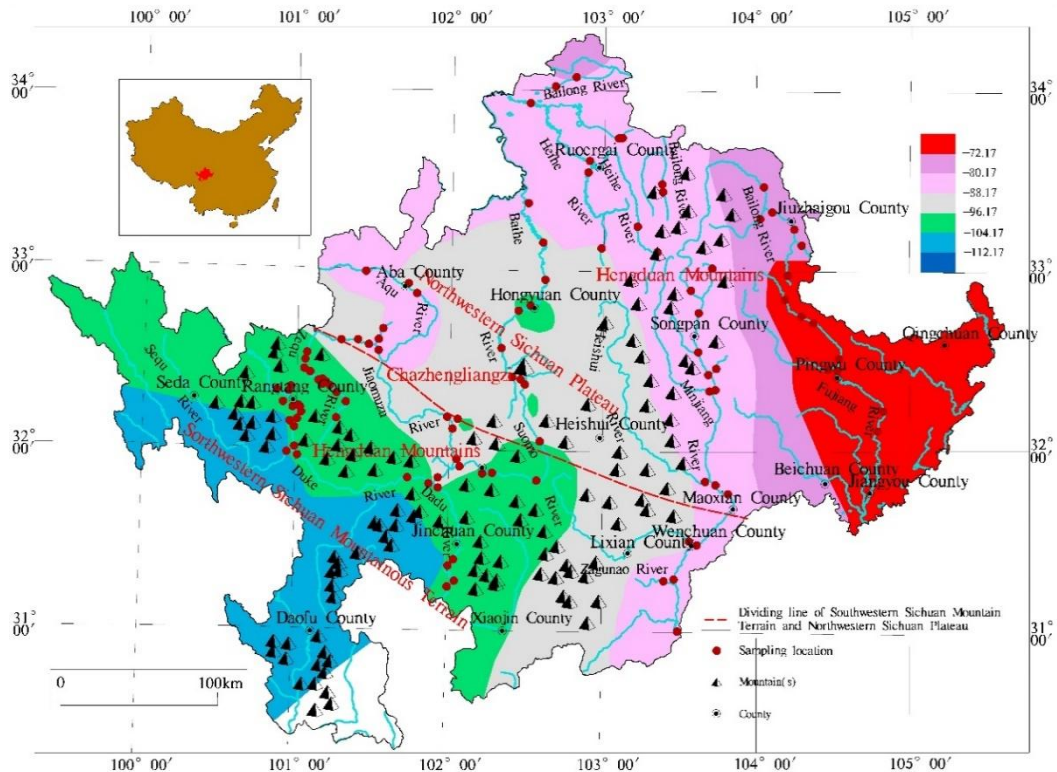


Figure 4. Spatial zonation of $\delta^2\text{H}$ in the study area

This noted general trend of isotopic composition change from east to west due to continental effect is fundamentally governed by temperature change. With decreasing temperature, rainfalls becomes more depleted in the heavier isotopes, ^2H and ^{18}O (^2H and ^{18}O are linearly correlated). Similar effects have been shown with increasing elevation and increased distance from the equator, both of which correspond to lower temperatures (Bowen et al., 2005b; Terzer et al., 2013). Our statistical analysis shows that both $\delta^2\text{H}$ and $\delta^{18}\text{O}$ are positively correlated with temperature, the Pearson product-moment correlation coefficients 0.492 and 0.517 for $\delta^2\text{H}$ -temperature and $\delta^{18}\text{O}$ -temperature, respectively, were obtained. Some early work (e.g., Zhang and Wu, 2007) suggests that the correlation between isotopic fractionation and temperature tends to be stronger when the temperature is lower.

Though the general zonation of $\delta^2\text{H}$ and $\delta^{18}\text{O}$ in the region (*Figures 4* and *5*) is believed to be governed by continentality, with increasing altitude the elevation and

other effects (e.g. amount effect) are superimposed on the continental effect and might become predominant resulting in a local enrichment of the heavier isotopes. This may be observed in *Figure 5* where both ^2H and ^{18}O are noticeably more depleted near Hongyuan presumably due to high altitude. Notwithstanding the altitude effect, the other factors including the amount effect (Dansgaard, 1964) and anthropogenic effect may also have played a role that should be fully investigated. For example, the east part of the study area is more densely populated and economically more developed; the increased anthropogenic activities including large amount of agricultural consumption of surface waters might have cut down the water runoff volumes and augmented natural evaporation and transpiration in the area, resulting in a more arid-like climate which in turn resulted in heavier isotopes relatively enriched (*Figures 4 and 5*).

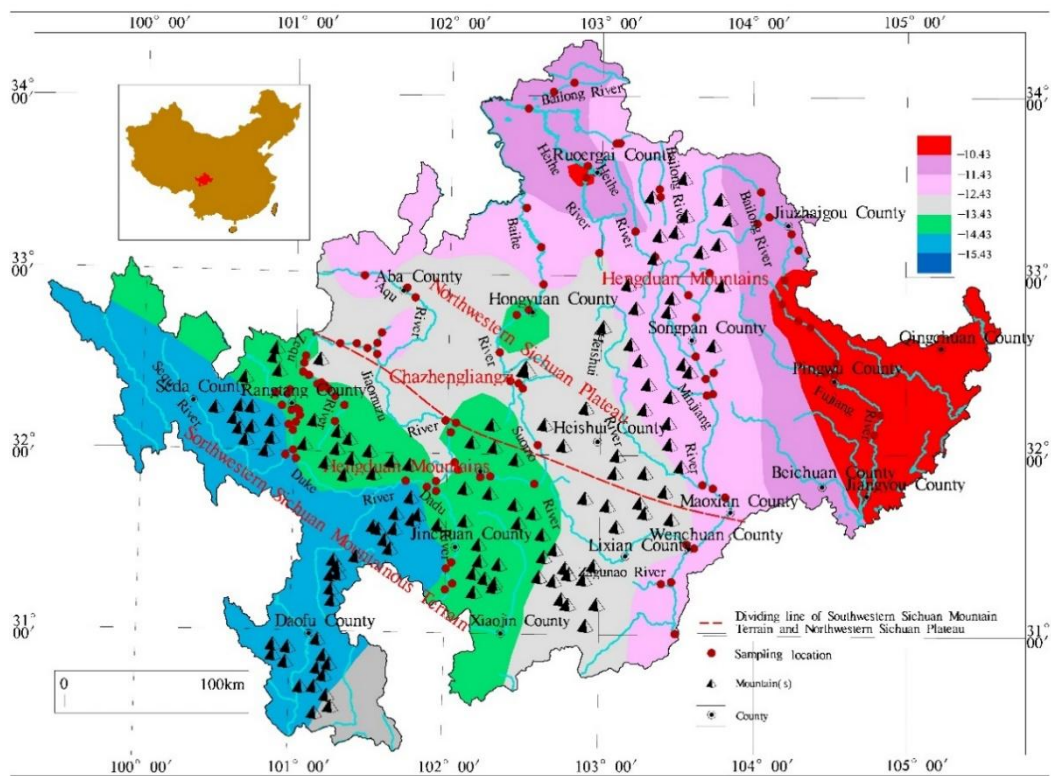


Figure 5. Spatial zonation of $\delta^{18}\text{O}$ in the study area

Long-term data sets are necessary to fully understand the processes of water cycles because seasonal and climate changes can be significant (Stumpp et al., 2014). For this reason, the results reported in this work may be regarded as a snapshot of the area's water cycle processes at the wettest time of a year and much of the discussion should be taken in this context. As seasonal variations are expected to be greater for rivers that are mainly recharged by surface runoff from recent precipitations (Rozanski et al., 2001) such as the rivers in this study, more systematic sampling and investigations are necessary to fully understand the area's runoff isotope hydrology.

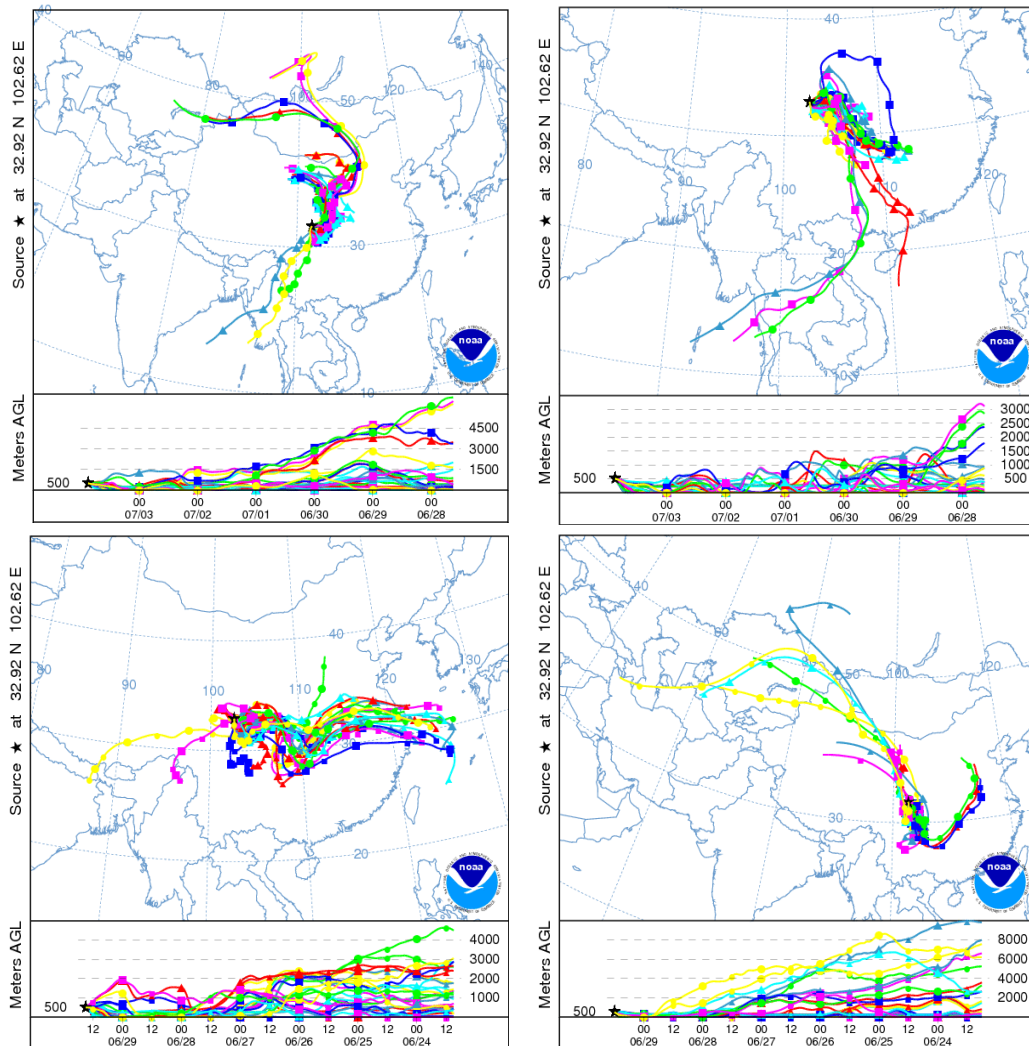


Figure 6. Week-long back trajectories of air masses on June 30 (bottom) and July 4 (top), 2010 (left) and 2011 (right) at 2 selected locations near the center of the study area

Isotope distinction between the drainage basins of Yangtze River and Yellow River

The meteoric water line specific to a certain location is controlled by local climatic factors including the oceanic moisture source and secondary processes affecting the isotopic composition of water vapor such as evaporation and re-circulation of moisture from large water bodies (Clark and Fritz, 1997). These factors together define a particular local meteoric water line (LMWL). A closer examination of *Figure 2* reveals that some data points deviate appreciably from the GMWL which otherwise represents the stable hydrogen and oxygen isotope ratio data quite well. However, when only the data from the Yangtze River basin are plotted in *Figure 7*, a single LMWL is obtained: $\delta^2\text{H}=8.06\delta^{18}\text{O}+12.69$ with $R^2 = 0.98$, which is almost identical to the GMWL as revised by Rozanski et al. (1993): $\delta^2\text{H}=8.17\delta^{18}\text{O}+10.35$, or by Gat et al. (2001): $\delta^2\text{H}=7.8973^{18}\text{O}+9.0133$. This is indicative of the fact that heavy and frequent rainfalls prior to our sampling in the region had rendered the stream waters resemble the

rainwaters so closely, and the streams in the Yangtze River drainage basin were recharged and maintained mainly by the rainfalls and the surface runoff. The lower the relative humidity is, the faster the evaporation rate would be and the greater the kinetic fractionation becomes. Humidity affects hydrogen and oxygen fractionations differently such that the slope of the evaporation line would vary due to changes in relative humidity. At very low relative humidity (< 25%), the slope of the evaporation line would be close to 4; for moderate relative humidity (25% to 75%) the slope would be between 4 and 5; only for relative humidity above 95% would the slope approach 8, i.e., the slope of the meteoric water line (Clark and Fritz, 1997). Therefore, from *Figure 7* it is inferred that humid conditions likely prevailed with relative humidity above 95% much of the time in the region. For a comparison, Liu et al. (2014), based on 928 groups of precipitation data, reported a Chinese Meteoric Water Line (CMWL) as $\delta^2\text{H}=7.48\delta^{18}\text{O}+1.01$, as also shown in *Figure 7*. It appears that there is a fair amount of difference between our local water line (i.e., LMWL) and the CMWL; however, considering our data range is relatively very narrow when compared to the data used for deriving the CMWL (Liu et al., 2014), the difference is not that significant. Nevertheless, it is noted that our data are systematically located above the CMWL, which is reflected by the relatively big difference in the d-values (12.69 vs. 1.01) indicating that the source of vapor for the area's precipitation was likely unique, i.e., sufficiently different from the bulk from that the CMWL was derived.

As noted earlier, the rivers belonging to the Yangtze River basin have rather different characteristics of stable hydrogen and oxygen isotopes from west to east across the area (*Table 3*). In the west of the Northwestern Sichuan Plateau the rivers including the Duke River, the Zequ River, the Suomo River, and the Jiaomuzu River are relatively enriched in the light isotopes; an exception is the Aqu River, a tributary of the Jiaomuzu River, which is relatively enriched in heavy isotopes and its d-values are lower than the other rivers. In comparison, the rivers in the east of the Northwestern Sichuan Plateau including the Fujiang River, the Minjiang River, and the Baishui River are relatively enriched in heavy isotopes. As discussed above, the isotope ratios change from east to west likely due to the continental effect, whereas the Aqu River was probably also appreciably recharged by alpine lake waters which were collection of glacier meltwater. However, the hydrogen and oxygen isotope ratios of the Fujiang River and the Minjiang River also have a trend of increasing from north to south possibly due to the altitude effect as the elevations increase appreciably (by as much as 2,000 m) from north to south along the two rivers. Besides, these two rivers have been more significantly affected by anthropogenic activities that may have added additional impact to the hydrogen and oxygen isotope ratios. For example, the water samples taken from the lower reach of the Minjiang River are slightly enriched in heavy isotopes with $\delta^2\text{H}/\delta^{18}\text{O}$ as -70.4/-10.54 and -81.7/-11.69 respectively from two locations (500 m downstream of the Yilixiang Hua'neng Hydro Station, and downstream after the Heishui River enters the Minjiang River), compared to the values of -90.6/-12.69 in its upper reach (upstream of the Zagu'nao River Hydro Station).

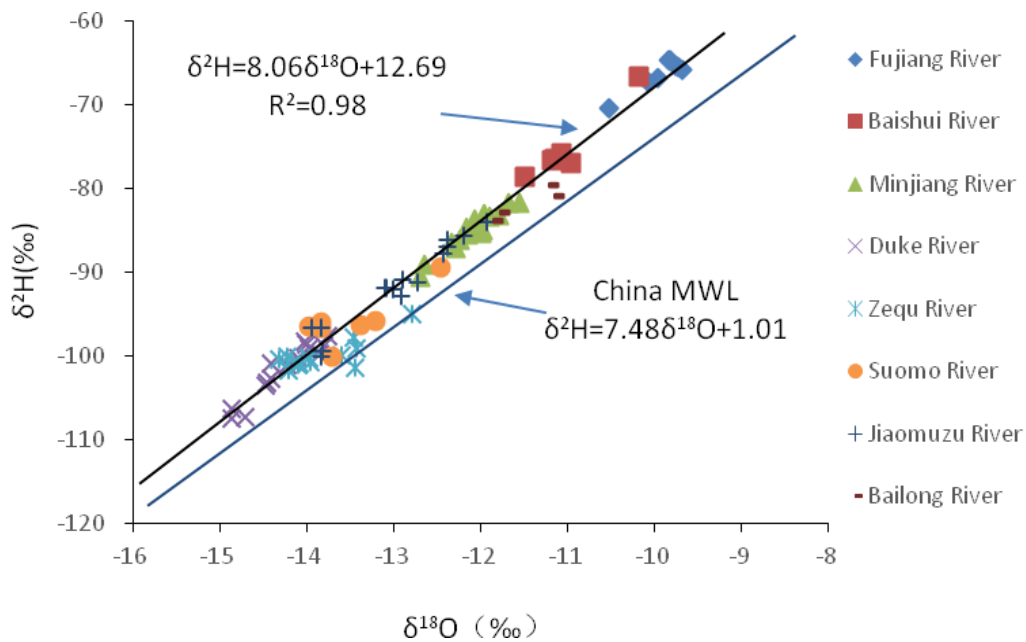


Figure 7. $\delta^2\text{H}$ vs. $\delta^{18}\text{O}$ diagram for stream water samples collected from the Yangtze River basin

The relatively small area in the northern part of the study area (*Figure 1*) including the Heihe River and the Baihe River catchments belongs to the Yellow River drainage basin. When the stable hydrogen and oxygen isotope ratios from this area only (i.e., the data points that deviate from the GMWL in *Figure 2*) were plotted as shown in *Figure 8*, a local evaporation line (LEL) was obtained: $\delta^2\text{H}=4.77\delta^{18}\text{O}-30.99$ with $R^2=0.93$, which differs appreciably from the GMWL (Rozanski et al., 1993; Hoefs, 2009), the CMWL (Liu et al., 2014), and the LMWL of the Yangtze River basin (*Figure 7*). The much smaller slope (4.77) is usually an indication of a more arid climate (i.e., lower relative humidity) (Gat, 1996).

The $\delta^2\text{H}$ and $\delta^{18}\text{O}$ values of water samples from the Yellow River basin range from -100.1 to -79.7 and from -13.98 to -10.03, respectively, varying over wider ranges (particularly the Heihe River) in comparison with those from the Yangtze River basin. In particular, the Heihe River and the Baihe River extend hundreds of kilometers from south to north before eventually flowing into the Yellow River, and their hydrogen and oxygen isotope compositions exhibit a general trend of increasingly more depleted in heavy isotopes. The d-excess data are presented as contoured lines in *Figure 3*; the d-values of water samples from this area are noticeably lower with a minimum of -0.25, also exhibiting a trend of decreasing from the upper reach to the lower reach of the rivers (i.e. from south to north). According to Gat (1996), in more arid areas evaporation from the falling rain droplets beneath the cloud base or from surface waters can result in the enrichment of the heavy isotopes in the remnant raindrops along the evaporation line, the resulting precipitation would show a smaller d-value

than at the cloud base. Therefore, even though both this small area belonging to the Yellow River basin and the east part of the study area display some isotopic features in common (e.g., relatively less depleted in heavy isotopes and smaller d-values), the underlying governing factors are different. This area is at or near the north China (NC) region where $\delta^{18}\text{O}$ was shown to be closely related to the wind directions, relative humidity and vapor pressure; this is distinctively different from the case of the SC region where $\delta^{18}\text{O}$ was shown to depend on the remaining fraction of the vapor phase in an isothermal condensation process (Liu et al., 2014).

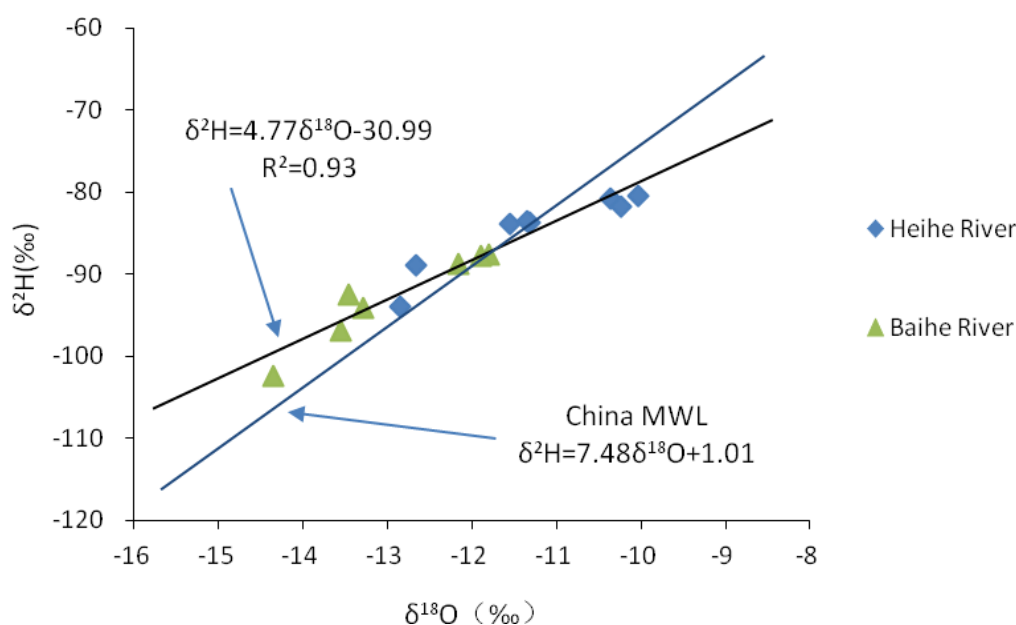


Figure 8. $\delta^2\text{H}$ vs. $\delta^{18}\text{O}$ diagram for water samples collected from the Yellow River basin

Mixing of some major rivers

When two or more tributaries merge to form a mainstream, the mixing ratio from each can be quantitatively calculated based on the isotopic mass balance principle. The fundamental equation (Eq. 1) can be written as:

$$r_T m_T = r_1 m_1 + r_2 m_2 + \dots + r_n m_n \quad (\text{Eq. 1})$$

where r is the fractional abundance of a stable isotope, m the molar quantity of the stable isotope, and the subscripts T and n denote, respectively, the merged mainstream and a tributary. When 2 tributaries merge to form a mainstream, the mass balance equation (Eq. 2) for ^{18}O can be written as:

$$r_{18\text{O}} m_{18\text{O}} = r_{18\text{O}_1} m_{18\text{O}_1} + r_{18\text{O}_2} m_{18\text{O}_2} \quad (\text{Eq. 2})$$

where (Eq. 3):

$$r_{18O} = \frac{m_{18O}}{m_{18O} + m_{16O}} \quad (\text{Eq. 3})$$

For all practical purposes, r_{18O} may be substituted by $\delta^{18}O$ and Eq. 2 becomes (Eq. 4):

$$\delta^{18}O m_{18O} = \delta^{18}O_1 m_{18O_1} + \delta^{18}O_2 m_{18O_2} \quad (\text{Eq. 4})$$

Eq. 4 is not exact because of the approximation of the δ values for r values. The errors due to this approximation have been carefully evaluated and proven negligible for all practical applications (Fry, 2003). Eq. 4 can be rearranged (note that $m_{18O} = m_{18O_1} + m_{18O_2}$) to read as (Eq. 5):

$$\frac{m_{18O_1}}{m_{18O}} = \frac{\delta^{18}O - \delta^{18}O_2}{\delta^{18}O_1 - \delta^{18}O_2} \quad (\text{Eq. 5})$$

where $\frac{m_{18O_1}}{m_{18O}}$ is the fraction contribution of tributary 1 to the mainstream.

The Dadu River is the most important tributary of the Minjiang River that in turn is a major tributary of the Yangtze River. The Dadu River has three sources: the east source Suomo River, the west source Duke River, and the main source Zumuzu River. At the confluence where the Sequ River merges into the Duke River, samples were taken and analyzed, and the following $\delta^{18}O$ values were obtained: -14.9‰, -14.0‰ and -14.2‰, respectively, from Sequ River, upstream of Duke River, and downstream of Duke River. Using Eq. 5 it is calculated that $\frac{m_{18O_1}}{m_{18O}}$ equals 0.28, i.e. the component from the Sequ River to the Duke River was 28% at the time of sampling.

Further upstream of the Sequ River, the Angkegou Stream in Rangtang is another tributary of the Duke River, but is much smaller than the Sequ River. The following $\delta^{18}O$ values were obtained around the point where the Angkegou Stream enters the Duke River: -14.9‰, -13.8‰ and -13.8‰, from the Angkegou Stream, upstream and downstream of the Duke River, respectively. Apparently, given the precision of our isotope analysis, it is not feasible to calculate the contribution of the Angkegou Stream to the Duke River using the oxygen isotope ratios with any confidence.

The Suomo River and the Duke River merge at Baiwan and then flow into the Dadu River. Near the joint, the $\delta^{18}O$ values were obtained as -13.1‰, -14.4‰ and -13.8‰, respectively, for the water samples from the Suomo River, Duke River and the Dadu River. The contribution of the Duke River to the Dadu River was calculated to be 55% and Suomo River to the Dadu River to be 45%.

The calculations based on mass balance are valid only when the tributary waters are fully mixed in the mainstream. Using the same method, the contribution of Ake River to

Ma'erqu (a secondary tributary of the Yangtze River, upper reach of the Jiaomuzhu River) was calculated to be 75%; but it was noted during field work that the Ake River flow was too small and its contribution to the Ma'erqu River could not possibly be that high. This apparent error was likely due to the fact that the sample taken from downstream Ma'erqu River with $\delta^{18}\text{O} = -12.4$ was most likely not representative of the well-mixed water body due to physical difficulty in sampling: a steep cliff prevented the sample from being taken near the center of the Ma'erqu River. Instead, the sample was taken near the cliff where the water was suspected to be predominantly from the Ake River ($\delta^{18}\text{O} = -12.2$), and a thorough mixing with the Ma'erqu River water had not yet achieved. Note that these river mixing calculations are approximate, as the uncertainties in the δ -values ($\pm 0.2\text{‰}$ and $\pm 2.0\text{‰}$ for $\delta^{18}\text{O}$ and $\delta^2\text{H}$, respectively) are not evaluated.

Conclusions

In general, the stable oxygen and hydrogen isotope ratios are correlated with the distance to the east coast of China, in agreement with a continentality model. That is, the stream waters were gradually depleted in heavy isotopes from east to west in the area studied. This phenomenon is attributed to the successive rainfalls from the same moisture cloud and the preservation of isotopic signature in the runoff from the precipitations. Local deviations from this general trend may be explained by the elevation effect and some anthropogenic factors.

The $\delta^2\text{H}$ and $\delta^{18}\text{O}$ data of the samples from the Yangtze River basin, which covers the majority of the study area, align well with the CMWL and particularly with the GMWL, suggesting that the stream waters in this area were primarily of meteoric origin (including ice and snow melting) with a regional weak evaporation effect prevailing. However, the data from a relatively small area belonging to the Yellow River basin are characterized by an appreciably flatter slope of the isotope water line, suggesting that the waters had undergone significant evaporation due to kinetic fractionations of isotopes in a more arid climate. These observations are also supported by the d-excess values.

Since the samples were taken during summer time, this study provides a snapshot of the region's river hydrology during the rainy season. As the stable isotope ratios can vary greatly both over time and space, for a more complete understanding of the local river hydrology more systematic sampling and analysis over time will be necessary. Precipitation samples should be collected and analyzed together with the stream water samples so that a direct correlation between the runoff waters and precipitations could be more firmly established. Nevertheless, the stream isotope data presented in this work should be valuable to those who are concerned with the TRH eco-system or, more broadly, the eco-environment of the Qinghai-Tibet Plateau.

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