

Evaporative enrichment of stable isotopes ($\delta^{18}\text{O}$ and δD) in lake water and the relation to lake-level change of Lake Qinghai, Northeast Tibetan Plateau of China

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Abstract: Stable isotopic compositions ($\delta^{18}\text{O}$ and δD) have been utilized as a useful indicator for evaluating the current and historical climatic and environmental changes. Therefore, it is vital to understand the relationship between the stable isotopic contents in lake water and the variations of lake level, particularly in Lake Qinghai, China. In this study, we analyzed the variations of isotope compositions ($\delta^{18}\text{O}$, δD and d-excess) in lake water and precipitation by using the samples that were collected from Lake Qinghai region during the period from 2009 to 2012. The results showed that the average isotopic contents of $\delta^{18}\text{O}$ and δD in lake water were higher than those in precipitation, which were contrary to the variations of d-excess. The linear regression correlations between $\delta^{18}\text{O}$ and δD in lake water and precipitation showed that the local evaporative line (LEL) in lake water ($\delta\text{D}=5.88\delta^{18}\text{O}-2.41$) deviated significantly from the local meteoric water line (LMWL) in precipitation ($\delta\text{D}=8.26\delta^{18}\text{O}+16.91$), indicating that evaporative enrichment had a significant impact on isotopic contents in lake water. Moreover, we also quantified the E/I ratio (evaporation-to-input ratio) in Lake Qinghai based on the lake water isotopic enrichment model derived from the Rayleigh equation. The changes of E/I ratios (ranging from 0.29 to 0.36 between 2009 and 2012) clearly revealed the shifts of lake levels in Lake Qinghai in recent years. The average E/I ratio of 0.40 reflected that water budget in Lake Qinghai was positive, and consistent with the rising lake levels and the increasing lake areas in many lakes of the Tibetan Plateau. These findings provide some evidences for studying the hydrological balance or water budget by using $\delta^{18}\text{O}$ values of lake sedimentary materials and contribute to the reconstruction of paleo-lake water level and paleoclimate from an isotopic enrichment model in Lake Qinghai.

Keywords: stable isotopes; d-excess; lake level; E/I ratio; isotopic enrichment model; paleoclimate

Citation: WU Huawu, LI Xiaoyan, LI Jing, JIANG Zhiyun, LI Guangyong, LIU Lei. 2015. Evaporative enrichment of stable isotopes ($\delta^{18}\text{O}$ and δD) in lake water and the relation to lake-level change of Lake Qinghai, Northeast Tibetan Plateau of China. *Journal of Arid Land*, 7(5): 623–635. doi: 10.1007/s40333-015-0048-6

Stable isotopic concentrations ($\delta^{18}\text{O}$ and δD) of natural waters can provide additional information for understanding the current and historical climatic and environmental changes because these isotopic signals preserved in the environmental archives (i.e. authigenic calcite, ostracodes, gastropods and diatom silica) are widely used to reconstruct past climate changes (Zhang et al., 1989; Hammarlund et al., 2002; Leng and Mar-

shall, 2004; Holmes et al., 2007; Liu et al., 2009; Li et al., 2012). Reconstruction of past and modern climate changes can be deduced from the stable isotopes in lake water (i.e. Qinghai Lake (Lister et al., 1991), Sub-Arctic Lake (Jonsson et al., 2009) and Tibetan Plateau lakes (Yuan et al., 2011)) because they sensitively recorded some information of isotopic compositions of meteoric water, enhancing the understanding

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Received 2014-10-28; revised 2015-03-02; accepted 2015-03-17

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of externally forced climatic change. This sensitivity is closely related to the shifts of isotopic contents in precipitation associated with hydrological cycles (i.e. moisture sources, trajectories and recycled water vapor) (Friedman et al., 2002; Vachon et al., 2010; Kong et al., 2013). Additionally, shifts in stable isotopic compositions in lake water are often attributed to many factors, for example, routine meteorological parameters (i.e. temperature, relative humidity and wind speed), input water and E/I ratio (evaporation-to-input ratio) of a lake with a hydrologically closed basin setting (Lister et al., 1991; Ricketts and Anderson, 1998; Leng and Marshall, 2004; Li et al., 2012; Xiao et al., 2013).

Lake Qinghai, situated in the arid and semi-arid northeastern Tibetan Plateau, is one of the most sensitive regions to global climate change. Due to its unique geographical location with a 'climatic triple junction' influenced by the Indian monsoon, the East Asian monsoon and a westerly circulation, Lake Qinghai has recently become an ideal place for paleoclimate research. The lake has attracted more attention from the international Quaternary communities for the following reasons: (1) both of the lake water level and lake area have experienced fluctuated changes and sensitively responded to climatic change during the Geologic age (Qin and Huang, 1998; Li et al., 2007; Hao, 2008); (2) reconstructing the paleoenvironment and paleoclimate of this region can be carried out by applying the stable oxygen-18 from lacustrine carbonate and geochemical sediments (Lanzhou Institute of Geology, CAS, 1979; Lister et al., 1991; Henderson et al., 2003; Shen et al., 2005; Liu et al., 2007); and (3) the closed-basin lake is strongly influenced by anthropogenic disturbances (Qin and Huang, 1998; Hao, 2008). A number of stable isotope records from lake sediments have been generated across the Lake Qinghai (e.g. Lister et al., 1991; Henderson et al., 2003; Leng and Marshall, 2004). These previous results provide important implications for paleoclimate reconstruction in this region. But there is a lack of studies about the relationship between isotope compositions in modern lake water and recent lake level (Lister et al., 1991). Lister et al. (1991) demonstrated that isotopic compositions in lake water were closely linked with lake-level changes subjected to a long-term hydrologic and climatic regime. The $\delta^{18}\text{O}$

of ostracodes and fine-grained carbonate was widely used to evaluate the changes of water level and precipitation in Lake Qinghai. It can reflect the isotopic compositions in input water (i.e. precipitation and runoff) and evaporative enrichment (Henderson et al., 2003). More importantly, this approach has been optimally applied in remote basins or mountainous regions (e.g. Tibetan Plateau) with limited instrumental records (Lister et al., 1991; Henderson et al., 2003). However, little information is available regarding the variations of isotopic compositions in modern lake water for discussions of their relationships with lake-level fluctuations in Lake Qinghai.

In this study, we evaluated the evaporative enrichment of isotopic concentrations in lake water and its relations to recent lake-level changes in Lake Qinghai, which can help us to establish information regarding the relationships between instrumental isotopic values of modern lake water and isotopic records of carbonate sediments from Lake Qinghai and to reconstruct the paleoclimate and paleolevel of Lake Qinghai. The objectives of this study were as follows: (1) to investigate the characteristics of isotopic compositions in lake water and precipitation in Lake Qinghai region; (2) to deduce the E/I ratio of Lake Qinghai from a modified isotopic enrichment model based on the isotopic compositions in lake water and precipitation; and (3) to reveal the potential relationships of E/I ratio and isotopic concentrations in lake water with lake level because recent lake level and surface area fluctuations have been increasing since 2004 (Hao, 2008).

1 Study area and methods

1.1 Study area

Lake Qinghai, the largest inland brackish lake in China, is an intermontane and a hydrologically closed basin in the northeastern Tibetan Plateau (99°36'–100°47'E, 36°32'–37°15'N; 3,200 m; Fig. 1). The lake is situated in a cold and high-altitude climate zone dominated by the East Asian monsoon, with the moisture derived mainly from the low-altitude oceans, the cold, dry polar airflow from the Siberian high-pressure system and the westerly jet stream. The annual mean temperature of the drainage basin is 0.1°C, and the mean annual precipitation is

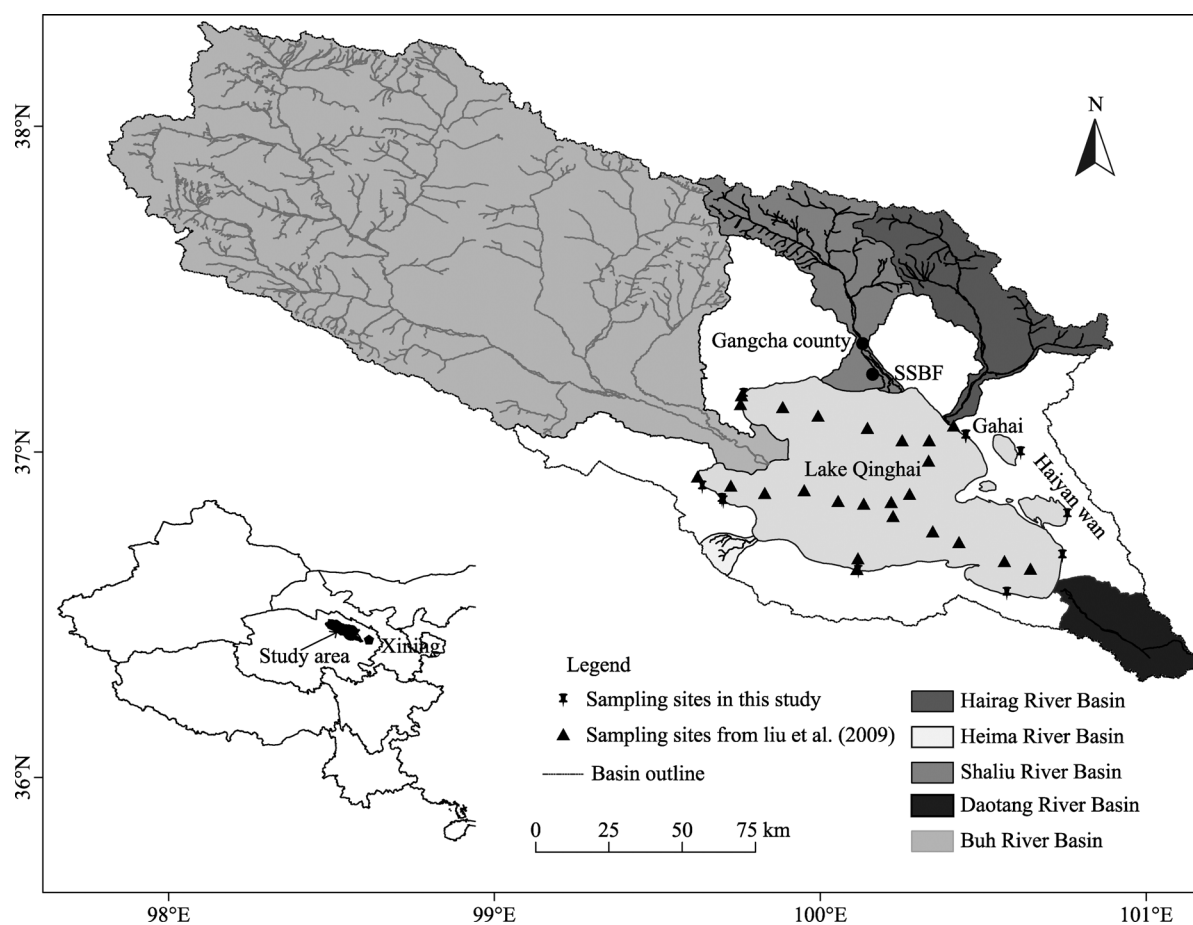


Fig. 1 The geographic location of the study area and sampling sites. SSBF, Sanjiaocheng Sheep Breeding Farm.

approximately 400 mm with more than 65% of the precipitation falling between June and September (Liu et al., 2009; An et al., 2012). The mean annual evaporation is approximately 1,300 mm, with over 65% of it occurring in the warmer summer season. During November and March next year, the lake surface is often frozen with a maximum ice thickness of 0.8 m. The lake output has no visible surface flow primarily via evaporation loss, which has resulted in $\text{Na}^+ - \text{Mg}^{2+} - \text{SO}_4^{2-} - \text{Cl}^-$ type water with a mean salinity of 14.1 g/L and alkaline water with a pH range of 9.1–9.4 (Lister et al., 1991; Henderson et al., 2003; Henderson and Holmes, 2009).

Lake Qinghai covers approximately 4,264 km² of surface area (in 2005), with the basin area of 29,699 km² and an average water depth of 21 m. The whole basin can be divided into six sub-basins (Fig. 1). Approximately 40 rivers are seasonally recharged into this closed lake, with the surface runoff accounting for

46% of the total lake input. Buh River, the longest and largest river in the whole basin, has a basin area of 14,337 km² and takes up approximately half of the discharge volume of the total runoff (Li et al., 2007; Henderson and Holmes, 2009). Because of no overflow in Lake Qinghai, the lake water residence time is approximately 33.4 years and the lake water temperature is significantly stratified from the surface (about 16°C) to bottom (about 6°C). However, this temperature-stratified phenomenon is disturbed by autumn overturn with high wind speeds (Lanzhou Institute of Geology, CAS, 1979; Lister et al., 1991).

1.2 Sampling and isotopic analysis

To well reveal how the isotopic contents ($\delta^{18}\text{O}$ and δD) in lake water respond to the changes in lake water level, two parts of isotopic data were analyzed. One part of $\delta^{18}\text{O}$ and δD data in lake water, precipitation and river water was collected from previous studies (Zhang et al., 1989; Henderson et al., 2003; Liu et al.,

2009). The other part of isotopic data was calculated from our field samplings between 2009 and 2012, with different frequencies of lake water and precipitation samples. Firstly, the lake water dataset consisted of 62 samples, which included 59 lake samples that were collected during 2009 and 2010 in different months and 3 lake samples in July and August 2012. All the lake water samples were collected from the littoral zone at a depth of 10–15 cm, where the lake water was well mixed and non-stagnant. Secondly, 36 event-based precipitation samples were collected from July to September in 2012 at the Sanjiaocheng Sheep Breeding Farm (SSBF), which is approximately 10 km away from the Gangcha county (Fig. 1), and 121-month precipitation isotopic data were obtained from the whole basin between May and October during 2009 and 2010. Finally, all the lake water and precipitation samples were transferred into clean and sealed polyethylene bottles (30 mL) for storage.

Stable isotope compositions ($\delta^{18}\text{O}$ and δD) of all samples were analyzed at the State Key Laboratory of Earth Surface Processes and Resource Ecology, Beijing Normal University, using an LGR DLT-100 (model: 908-0008) Laser Adsorption Spectroscopy (Los Gatos Research, Inc., Mountain View, CA, USA). The isotope composition data were expressed conventionally as δ values (‰), which represented the deviations in per mil (‰) from V-SMOW (Vienna Standard Mean Ocean Water) standards for oxygen-18 and deuterium, such that $\delta_{\text{sample}} = 1000 \times [(R_{\text{sample}}/R_{\text{VSMOW}}) - 1]$. Where, R is the $^{18}\text{O}/^{16}\text{O}$ or $^2\text{H}/^1\text{H}$ and denotes the ratio of elements in the sample and in the standard. The measuring precision was $\pm 1.2\text{‰}$ for δD and $\pm 0.3\text{‰}$ for $\delta^{18}\text{O}$.

1.3 Description of isotopic model

The model of lake water isotopic enrichment was derived from the classical Rayleigh equation (Dansgaard, 1964; Yuan et al., 2006).

$$\delta_L \times 10^{-3} = (\delta_L^0 \times 10^{-3} - 1) F^{(\alpha-1)} + 1. \quad (1)$$

Where, δ_L and δ_L^0 represent the isotopic compositions of the instantaneous lake water (from our collected samples) and the initial bulk lake water (assumed to the intersection of the local meteoric water line (LMWL) and local evaporative line (LEL)), respectively; F is the fraction of water that remains in the

lake and equals to $1 - E/I$ (E represents the evaporation overlay of the lake, and I represents the input water volume, including direct precipitation and inflow water (river water, groundwater and glacial water)); and α is the isotopic fractionation factor between the vapor-liquid phases and is dominated by equilibrium and kinetic processes during the evaporation. Based on the evaporation theory concerning the isotopic fractionation factor in the open surface water from Craig and Gordon (1965), we calculated the isotopic fractionation factor using the following equation (Yuan et al., 2006):

$$\alpha = \alpha_k \left[\frac{\alpha_{eq} - hf_a(\delta_v - 1) \times 10^{-3} / (\delta_L - 1) \times 10^{-3}}{1 - h + \alpha_k h(1 - f_a)} \right]. \quad (2)$$

Where, α_k and α_{eq} are the kinetic and equilibrium fractionation factors, respectively; h, δ_v and δ_L denote the relative humidity, the isotopic compositions of the atmospheric water vapor and the instantaneous lake water, respectively; f_a is the fraction of advected water vapor in the boundary layer over the lake; α_{eq} is a temperature-dependent factor controlled by the water surface temperature. The equilibrium fractionation factors for ^{18}O and ^2H were given as follows according to the study of Majoube (1971).

$$1000 \ln(\alpha^{18}\text{O}) = (1.137 \times 10^6)/T^2 - (0.415 \times 10^3)/T - 2.0667, \quad (3)$$

$$1000 \ln(\alpha^2\text{H}) = (24.844 \times 10^6)/T^2 - (76.248 \times 10^3)/T + 52.612. \quad (4)$$

Where, T is the air-water interface temperature of surface lake water.

The kinetic fractionation factor is largely determined by different molecular diffusivity ratios of water isotopomers, which can be approximately calculated following the study of Stewart (1975):

$$\alpha_k = (D_H/D_L)^n. \quad (5)$$

Where, D_H and D_L are the molecular diffusivities for heavy (D or ^{18}O) and light (H or ^{16}O) isotopomers, respectively; and n is the molecular diffusion factor, which refers to the stage of molecular diffusivity ranging from 0 (completely turbulent diffusion) to 1 (molecular diffusion). The ratios of D_H/D_L and $^{18}\text{O}_H/^{16}\text{O}_L$ in Eq. 5 refer to 0.984 and 0.969, respectively (Cappa et al., 2003).

1.4 Modeling the E/I ratio

Modeling the E/I ratio is dependent on many factors, such as the isotopic compositions in input water (δ_i), atmospheric vapor (δ_v) and lake water (δ_l), the surface lake water temperature, relative humidity and fraction of advected atmospheric vapor (f_a). The isotope compositions in input water for Lake Qinghai can be deduced from the intersection of LEL with LMWL, which roughly represent the weight mean isotope compositions of lake water inflow (Gibson et al., 1993; Mayr et al., 2007). The calculated values of δ_i from 2009 to 2012, which were slightly higher than the contents of isotopic compositions in river water and groundwater (Zhang et al., 1989; Cui, 2011), are shown in Table 1. Previous studies have demonstrated that isotopic contents in atmospheric vapor were substantially lower than those in precipitation, and that the condensation process between atmospheric vapor and precipitation could be regarded as an equilibrium fractionation process (Jacob and Sonntag, 1991; Wen et al., 2010). Yin et al. (2008) determined that the contents of $\delta^{18}\text{O}$ in 255 atmospheric vapor samples that were collected from July 2005 to March 2006 at the Delingha station ranged from -39.5‰ to 1.5‰ , with an average of -21.2‰ , which was assumed to be approximately equal to that in Lake Qinghai. In this study, we assumed that the isotopic compositions in atmospheric vapor were in equilibrium with those in precipitation, thus δD of atmospheric vapor in Lake Qinghai was deduced from the global meteoric water line (GMWL; $\delta\text{D} = -159.6\text{‰}$).

According to the meteorological observation station at the Gangcha county, the annual mean air temperature is approximately 0.1°C , the maximum daily mean temperature is 20.7°C and the mean surface water temperature during the free-ice period is 16°C (Lanzhou Institute of Geology, CAS, 1979). The variations of surface water temperature were assumed to range from 0 to 28.8°C . The annual mean relative humidity (h) is approximately 0.54 based on the meteorological observation station at the Gangcha county and the values of h ranged from 0.32 to 0.80 in the modeling of the E/I ratio. The fraction of advected atmospheric vapor (f_a) theoretically changed from 0 to 1. However, previous studies about Pyramid Lake of western United States showed that most of the water vapor

above the water surface was derived from the lake itself, for example, f_a was less than 0.25 (Benson and White, 1994; Benson and Paillet, 2002).

To obtain the optimal E/I ratio, we calculated the optimal values of four parameters (i.e. surface water temperature (T), relative humidity (h), the fraction of advected atmospheric vapor (f_a) and molecular diffusion factor (n)) by using the minimization of root mean square error (RMSE) between $(\text{E}/\text{I})_{\delta^{18}\text{O}}$ and $(\text{E}/\text{I})_{\delta\text{D}}$ (Eq. 6; Fig. 2).

$$\text{RMSE} = \sqrt{\frac{\sum_{i=1}^n (F - \bar{F})_{\delta^{18}\text{O}}^2 + (F - \bar{F})_{\delta\text{D}}^2}{n}}. \quad (6)$$

Where, F is the same to that in Eq. 1. Finally, the optimal values of the four parameters were deduced from the minimization of RMSE, i.e. $T=14.4^\circ\text{C}$, $h=0.515$, $f_a=0.207$ and $n=0.336$ (Fig. 2). The optimal value of surface water temperature ($T=14.4^\circ\text{C}$) in Lake Qinghai is similar to that in the southern Tibetan Plateau during the ice-free season (Yuan et al., 2011). The optimal value of relative humidity ($h=0.515$) falls in the range of daily relative humidity (0.500–0.600) and is lower than the annual average relative humidity ($h=0.540$). The optimal value of f_a in Lake Qinghai is higher than that reported by Benson and Paillet (2002) and Yuan et al. (2011), which is related with windy climate conditions during the ice-free season. The average values of E/I ratio from 1985 to 2002 are shown in Table 1. These results are consistent with the variations of E/I ratios in 27 lakes of the southern Tibetan Plateau (Yuan et al., 2011) and similar to the previous study of Mayr et al. (2007).

2 Results

2.1 Variations of stable isotopic compositions (δD and $\delta^{18}\text{O}$) in precipitation and lake water

Temporal variations of δD and $\delta^{18}\text{O}$ in precipitation from 2009 to 2012 are shown in Fig. 3, with greater fluctuations in event-based isotope compositions in 2012 than those in monthly-based isotope compositions from 2009 to 2010. In 2012, the event-based $\delta^{18}\text{O}$ isotope compositions in precipitation varied from -14.79‰ to 0.15‰ , with a mean value of -6.89‰ ($\pm 0.64\text{‰}$, $n=36$); while δD values varied from -107.80‰ to 20.60‰ , with a mean of -42.40‰

Table 1 Variations of E/I ratio and isotopic contents ($\delta^{18}\text{O}$ and δD) in lake water and input water, and slope (a) and intercept (b) of linear regression correlations between $\delta^{18}\text{O}$ and δD

Year	$(E/I)_{\delta^{18}\text{O}}$	$(E/I)_{\delta\text{D}}$	$(E/I)_{\text{Ave.}}$	LEL		δ_{L}							
				a	b	δD			$\delta^{18}\text{O}$			δ_{i}	
						Max.	Min.	Mean	Max.	Min.	Mean	δD	$\delta^{18}\text{O}$
1985	0.60	0.58	0.59	NaN	NaN	34.50	3.60	10.80	4.20	1.04	2.14	-55.00	-10.00
2001	NaN	NaN	NaN	NaN	NaN	NaN	NaN	NaN	2.64	2.85	NaN	NaN	NaN
2009 ^a	NaN	NaN	NaN	NaN	NaN	NaN	NaN	NaN	3.71	-0.57	1.97	NaN	NaN
2009 ^b	0.36	0.37	0.36	5.76	-1.88	17.06	-9.57	6.19	3.70	-0.89	1.40	-46.60	-7.76
2010	0.33	0.37	0.35	5.66	-2.27	12.35	-18.61	3.42	2.87	-2.75	1.01	-45.80	-7.69
2012	0.27	0.31	0.29	6.41	-5.87	-3.76	-8.79	-6.46	2.89	-0.31	-0.09	-60.90	-10.30

Note: δ_{L} represents the mean values of $\delta^{18}\text{O}$ and δD in lake water. δ_{i} is deduced from the intersection of the local evaporative line (LEL) with local meteoric water line (LMWL) for $\delta^{18}\text{O}$ and δD in input water. a and b denote the slope and intercept of the LEL, respectively. $(E/I)_{\delta^{18}\text{O}}$ and $(E/I)_{\delta\text{D}}$ are calculated from $\delta^{18}\text{O}$ and δD , respectively. The data of 1985, 2001 and 2009^a are derived from the studies of Zhang et al. (1989), Henderson et al. (2003) and Liu et al. (2009), respectively. The data of 2009^b, 2010 and 2012 are calculated from our field samplings. NaN denotes no data. Max.: maximum; Min.: minimum.

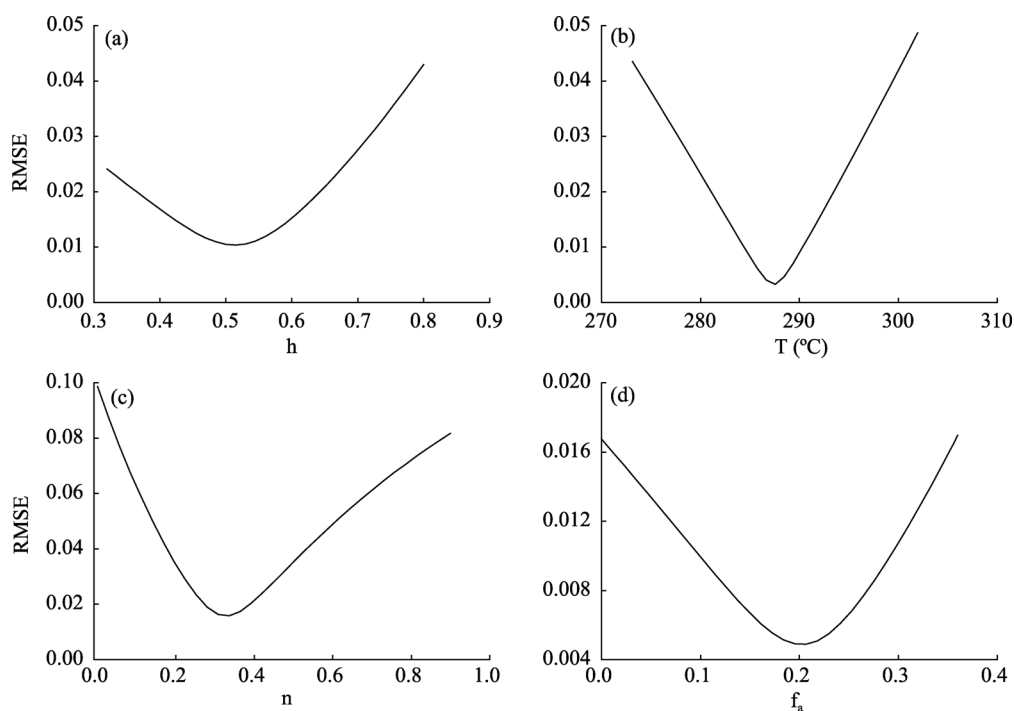


Fig. 2 Optimaization of four isotopic enrichment model parameters for (a) relative humidity (h), (b) surface water temperature (T), (c) molecular diffusion factor (n) and (d) fraction of advected water vapor (f_a). The root mean square error (RMSE) is derived from the relative error between $(E/I)_{\delta^{18}\text{O}}$ and $(E/I)_{\delta\text{D}}$.

($\pm 5.65\%$, $n=36$). By contrast, the monthly-based $\delta^{18}\text{O}$ and δD isotopic compositions in precipitation ranged from -16.69% to -2.80% (mean value of $-7.81\% \pm 0.34\%$, $n=81$) and -123.28% to -9.14% (mean value of $-46.90\% \pm 2.81\%$, $n=81$) in 2009, respectively. Moreover, the values of $\delta^{18}\text{O}$ ranged from -11.53% to -4.32% (mean value of $-7.82\% \pm 0.33\%$, $n=40$), and δD ranged from -75.12% to -13.74%

(mean value of $-46.80\% \pm 2.71\%$, $n=40$) in 2010. The average values of δD (-45.9%) and $\delta^{18}\text{O}$ (-7.59%) in precipitation in this study were higher than those (-92.10% for δD and -15.12% for $\delta^{18}\text{O}$) in a study of Zhang et al. (1989).

The values of $\delta^{18}\text{O}$ and δD in lake water also showed great variations (Figs. 3a and b), ranging from -2.75% to 3.70% for $\delta^{18}\text{O}$ and -18.61% to 17.06%

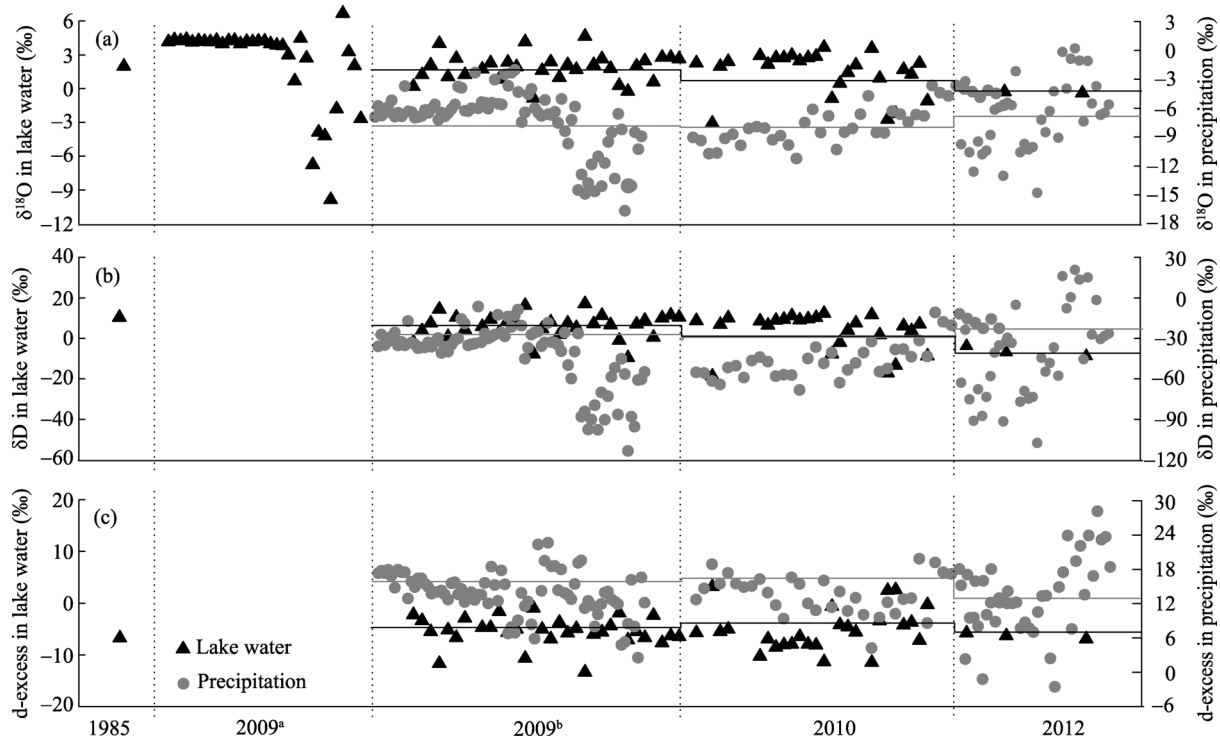


Fig. 3 Temporal variations of isotopic compositions ($\delta^{18}\text{O}$, δD and d-excess) in lake water and precipitation. Gray and black solid lines represent the average values of isotope compositions. Isotopic data in 1985 and 2009^a are derived from the studies of Zhang et al. (1989) and Liu et al. (2009), respectively; and the data of 2009^b, 2010 and 2012 are calculated from our field samplings.

for δD from 2009 to 2012. The average values of δD and $\delta^{18}\text{O}$ in lake water in this study were 4.44‰ ($\pm 0.34\%$, $n=70$) and 1.17‰ ($\pm 2.06\%$, $n=70$), respectively, which were all lower than those in the studies of Zhang et al. (1989) and Liu et al. (2009). The values of d-excess (d-excess= $\delta\text{D}-8\delta^{18}\text{O}$; Dansgaard, 1964) in precipitation changed from -2.63% to 28.10‰, with an average of 14.90‰ from 2009 to 2012 (Fig. 3c), which was substantially higher than the global average d-excess value (10‰; Dansgaard, 1964). The d-excess values of lake water ranged from -12.54% to 3.39‰, with an average value of -4.91% (Fig. 3c).

In this study, a significant positive correlation between δD and $\delta^{18}\text{O}$ in lake water and precipitation was examined (Fig. 4). The regression correlation between δD and $\delta^{18}\text{O}$ in precipitation was defined as LMWL ($\delta\text{D}=(8.26\pm 0.12)\delta^{18}\text{O}+(16.91\pm 1.02)$), and the LMWL in this study deviated from the GMWL ($\delta\text{D}=8\delta^{18}\text{O}+10$; Craig, 1961). Moreover, the regression correlation between δD and $\delta^{18}\text{O}$ in lake water (LEL; $\delta\text{D}=(5.88\pm 0.08)\delta^{18}\text{O}-(2.41\pm 0.24)$) also deviated significantly from the GMWL.

2.2 Changes in lake-water level, air temperature and precipitation

There was a large fluctuation in lake-water level during the period between 1959 and 2011 in Lake Qinghai, with a decrease from 3,196.55 (in 1959) to 3,192.97 m (in 2004), but an increase in recent years (2005–2011; Fig. 5). In contrast, the air temperature and annual precipitation showed an increasing trend from 1958 to 2012, with the rates of $0.33^\circ\text{C}/10\text{a}$ ($R^2=0.637$) and $5.24\text{ mm}/10\text{a}$ ($R^2=0.026$), respectively. Moreover, the annual precipitation was typically exhibited an increasing trend from 2004 to 2011, with the values higher than the average annual precipitation (381.4 mm).

3 Discussion

3.1 Variations in oxygen-18 and deuterium compositions in lake water and precipitation

Stable isotopic compositions (δD and $\delta^{18}\text{O}$) in precipitation are generally regarded as the integrated tracers of atmospheric processes worldwide. These compositions are not only influenced by temperature

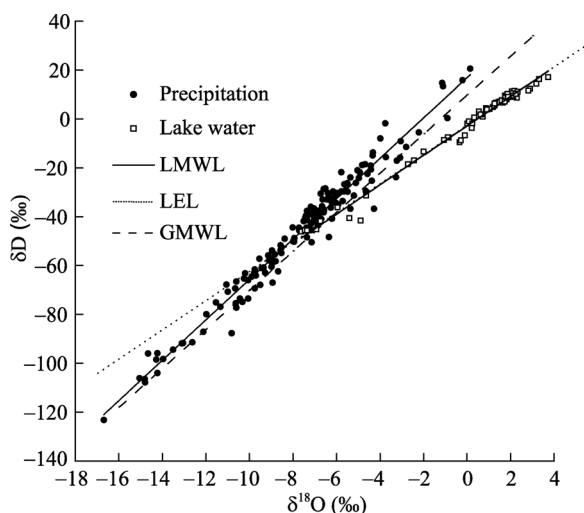


Fig. 4 Linear regression correlations between δD and $\delta^{18}O$ in lake water and precipitation in Lake Qinghai. LMWL, local meteoric water line; LEL, local evaporative line; GMWL, global meteoric water line.

during the condensation of raindrops in cloud and evaporation process during their descent (Araguás-Araguás et al., 1998; Peng et al., 2007; Pang et al., 2011), but also influenced by air trajectories from various moisture sources (Vuille et al., 2005; Xie et al., 2011; Wu et al., 2015) and local recycled vapor (Kurita and Yamada, 2008; Kong et al., 2013). During the sampling period in this study, the average value of $\delta^{18}O$ (-7.59‰) in precipitation was nearly identical to the weighted $\delta^{18}O$ in precipitation (-7.74‰) at the Delingha station (Tian et al., 2003; Yao et al., 2013). Compared with the southern stations (i.e. Lhasa and Nyalam station) in Tibetan Plateau, higher concentration of $\delta^{18}O$ in precipitation was observed in Lake Qinghai Basin. These results were primarily attributable to the distinctive differences of moisture sources (Tian et al., 2001a, 2003; Yao et al., 2013). Moisture sources were mainly dominated by continental air

masses in the northern Tibetan Plateau, i.e. Delingha and Lake Qinghai regions; whereas they were dominated by monsoons from the southern Indian Ocean and the Bay of Bengal (BoB), with progressive distillation depletion for isotopic compositions of air masses during orographic ascent, cooling and rainout effect in the southern Tibetan Plateau (Tian et al., 2001a, b). In addition, high concentrations of isotope compositions in precipitation from the Lake Qinghai Basin were possibly influenced by the local recycled moisture with heavier isotopic compositions from the Lake Qinghai itself and from the plants and soils, which may be a main contributor to the local precipitation (Froehlich et al., 2008; Kurita and Yamada, 2008).

The fluctuations of isotopic compositions (δD and $\delta^{18}O$) in lake water, particularly in a perennial closed-basin lake (i.e. Lake Qinghai), are closely associated with the input water (including inflow and direct precipitation) and output water (evaporation). The isotopic values of lake water in Lake Qinghai (4.44‰ for δD and 1.16‰ for $\delta^{18}O$) have become increasingly more depleted recently, compared with the result (10.8‰ for δD and 2.14‰ for $\delta^{18}O$) of Zhang et al. (1989) and the finding (1.97‰ for $\delta^{18}O$) of Liu et al. (2009). Meteorological factors, i.e. precipitation, air temperature and evaporation loss, directly affect the lake water isotope values. Generally, the effect of air temperature on evaporative loss controls the degree of enrichment in δD and $\delta^{18}O$ for terminal lakes; while indirect precipitation and inflow (groundwater, glacial and river water), which are primarily dependent on the amount of precipitation, have great influence on the depletion of δD and $\delta^{18}O$ for terminal lake water. Therefore, Lake Qinghai may experience a relatively weakened evaporation as

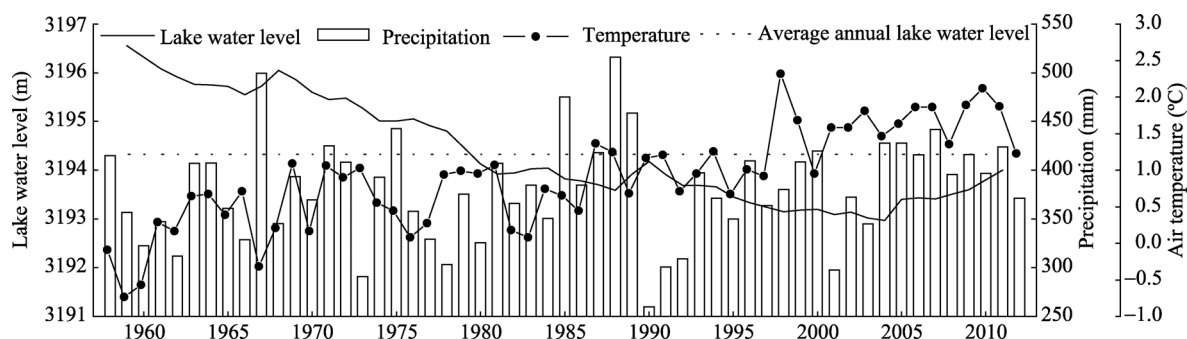


Fig. 5 Variations in lake water level from 1959 to 2011 with both the precipitation and air temperature from 1958 to 2012 in Lake Qinghai

deduced from the variations in isotopic composition in lake water.

3.2 Characteristics of d-excess

The d-excess value is a useful indicator of detecting the moisture sources and evaporative process because it is relatively invariant during transport (Merlivat and Jouzel, 1979; Pfahl and Wernli, 2008) and can be used to infer the effects of evaporative fractionation on isotope compositions in precipitation and lake water (Peng et al., 2007; Vallet-Coulomb et al., 2008; Pang et al., 2011). Meanwhile, d-excess values can significantly vary from different regions. For example, precipitation usually has larger d-excess values ($>10\%$) due to the interaction of dry air masses with the continental recycled water vapor complement in the Great Lakes or in the eastern Mediterranean region (Gat and Carmi, 1970; Gat et al., 1994; Machavaram and Krishnamurthy, 1995). However, in arid and semi-arid regions, some precipitation samples have lower d-excess values ($<10\%$) or even negative values (Peng et al., 2004; Pang et al., 2011). The average d-excess value (14.9%) in precipitation in Lake Qinghai was much higher than the global average (10%). Due to non-equilibrium fractionation that occurs during evaporation from the land surface, the d-excess value of evaporated moisture is higher than that of land surface water (Tian et al., 2003; Henderson-Sellers et al., 2004). Thus, the higher d-excess value in precipitation may be influenced by the contribution of evaporative moisture from land surface water in Lake Qinghai Basin.

Additionally, the magnitude of d-excess value for precipitation is closely related to the meteorological conditions at the moisture source region, such as the relative humidity over the evaporative surface, climatic conditions of sea surface and wind speed at the source region of the atmospheric moisture (Merlivat and Jouzel, 1979). The average d-excess value of lake water (-4.91%) in Lake Qinghai was substantially lower than the global average value (10%), indicating that the isotopic compositions in lake water have experienced a strong evaporation process (Dansgaard, 1964; Gat and Matsui, 1991; Peng et al., 2007). The meteorological features of Tibetan Plateau are typically characterized by strong convective

activity, radiation and wind (Lanzhou Institute of Geology, CAS, 1979; Kurita and Yamada, 2008). These characteristics primarily contribute to the strong evaporation in the Lake Qinghai region. Due to the slower diffusion of H_2^{18}O relative to H_2^{16}O during evaporation that occurs at the air-water interface, lower d-excess values of lake water can result from an evaporative effect.

3.3 Regression correlations between δD and $\delta^{18}\text{O}$

The isotopic compositions (δD and $\delta^{18}\text{O}$) in meteoric water vary with different geographical parameters. However, most rainfall and snowfall samples fall close to the GMWL (Craig, 1961; Rozanski et al., 1993). In fact, the regression correlation between δD and $\delta^{18}\text{O}$ in precipitation (LMWL) varied from different regions. The slope and intercept of LMWL diversely deviated from the GMWL which was controlled by numerous local geographic and meteorological factors, i.e. moisture sources, air mass trajectory, evapotranspiration effects at local and synoptic scales, local climatic conditions, precipitation formation processes and secondary evaporation during precipitation descent. The LMWL ($\delta\text{D}=8.26\delta^{18}\text{O}+16.91$) of Lake Qinghai was similar with that of Delingha station ($\delta\text{D}=8.5\delta^{18}\text{O}+15.2$; Tian et al., 2001b), which deviated from the GMWL. Some climatic information could be deduced by comparisons between the slopes and intercepts of GMWL and LMWL in previous studies (Araguás-Araguás et al., 1998; Peng et al., 2004). For example, Araguás-Araguás et al. (1998) stated that the slope value of >8 in LMWL was primarily found in the South Pacific Domain, where air masses were mainly characterized by high contents of isotopic compositions from complex monsoon systems that experienced stronger distillation effects from the ocean to inland (Indian Monsoon and East Asian Monsoon). Thus, we can infer that the isotopic contents in precipitation in Lake Qinghai Basin are influenced by heavier summer monsoons from the Indian Ocean, which bring more moisture to the Tibetan Plateau because of moisture source from Indian Ocean, forming depleted isotope compositions (Tian et al., 2001a; Hao, 2008).

In general, the concentrations of δD and $\delta^{18}\text{O}$ in diverse water bodies (river, groundwater and lake wa-

ter) experience various degrees of fractionation processes during the water cycle due to the evaporation effect. The slope and intercept of LEL ($\delta D=5.88\delta^{18}O-2.41$) of Lake Qinghai evidently deviated from the GMWL and LMWL, and differed from those of the LEL ($\delta D=5.2\delta^{18}O-38.9$) of 27 lakes in the southern Tibetan Plateau (Yuan et al., 2011) and the evaporative line ($\delta D=3.3\delta^{18}O-43$) of Dabusun Lake in the Qaidam Basin (Yang et al., 1995). The deviations between LMWL and LEL of Lake Qinghai can also be used to examine the evaporative enrichment of isotope fractionation in lake water.

3.4 Relationships between the E/I ratio, isotopic compositions and lake water level

Lake water level, particularly for Lake Qinghai, responds to changes in precipitation and evaporation that are integrated over the entire lake and basin. Moreover, the water-level record of Lake Qinghai is an important indicator for understanding the current climate change and evaluating the impact of climate change on environments and regional economy (Qin and Huang, 1998; Bianduo et al., 2006; Li et al., 2007). The rising water level is closely linked with the changes in precipitation and air temperature. Precipitation directly affects the input water of the lake, while the changes in air temperature affect the output water of the lake by evaporation and result in permafrost layer degradation and glacial melting. Simultaneously, air temperature, which may accompany the change of relative humidity, affects the isotopic compositions ($\delta^{18}O$ and δD) for both precipitation and evaporation. Meanwhile, the contents of isotopic compositions in lake water and the value of E/I closely respond to the water-level change in Lake Qinghai.

Shifts in perennial closed-basin lake levels are also reflected by the ratio of evaporation (E) to input water volume (I). Evaporation from lake results in isotopic enrichment (^{18}O and D), causing a positive shift in isotopic composition of lake water. The modeling E/I ratios showed a decreasing trend from 1985 to 2012, with a rising water level and an evidently increasing trend of precipitation (Table 1; Fig. 5). Thus, the decline in evaporation and the increase in input water volume of Lake Qinghai can be inferred from the changes of E/I ratios (0.29 to 0.36), which possibly

well reflected the recent rising lake level (2004–2011). Lister et al. (1991) reported that the great influence of environmental factors on lake level and $\delta^{18}O$ occurred under a given climatic regime. The overall trend of $\delta^{18}O$ in lake water with negative values can be interrupted by a negative shift (I>E) for a relatively higher lake level, whereas a positive shift (E>I) with a lower lake level has a positive $\delta^{18}O$ value. The isotopic compositions in lake water exhibit negative values (ranging from -0.09‰ to 2.14‰ for $\delta^{18}O$ and -6.46‰ to 10.80‰ for δD ; Table 1). Simultaneously, the lake level was lower during our study periods and was below the average water level (3,194.36 m). Therefore, understanding the variations in the E/I ratios and isotopic compositions ($\delta^{18}O$ and δD) in lake water are important for studying the climatic and hydrological regime, particularly in remote areas, such as Tibetan Plateau with limited instrumental observation data.

Although the modeling E/I ratio appeared well matched with lake-level change under an ideal scenario via Rayleigh equation, some unclear factors still remained. Firstly, we assumed that the lake hydrologic balance keeps in an equilibrium process during different sampling periods, namely, input water does not evidently alter the lake water storage. Secondly, the dynamically hydrologic processes of lake may impact the application of Rayleigh equation into the calculation of E/I ratio because the isotopic contents of lake water are subject to evaporation and dilution. Thus, we should further test our modeling E/I ratio via comparison with the methods of Gibson and Edwards (2002) in our next works to well reveal the E/I ratio of a closed-basin lake and to evaluate the dynamic processes of remote lake.

Additionally, the hydrochemistry of lake water also sensitively responds to the changes of lake level and the E/I ratio, i.e. the total dissolved solids (TDS), total inorganic carbon (TIC), Ca^{2+} and HCO_3^- (Yuan et al., 2006, 2011; Liu et al., 2009). Liu et al. (2009) reported that the oxygen-18 composition in lake water showed a positive correlation with salinity due to the decreases in input water and residual water ratios, which resulted from the evaporative loss. Evaporation can result in an enrichment of isotopic contents until the isotopic compositions in lake water reaches an isotopic steady-state. Due to the lack of recent salinity

data, we can only speculate that water salinity in Lake Qinghai is currently lower than that in previous studies (Lister et al., 1991; Liu et al., 2009) according to the rising water level and increasing precipitation (Fig. 5). Meanwhile, the lake volume and lake area also increased during recent years (Qin and Huang, 1998; Hao, 2008).

4 Conclusions

This study presented the variations of isotopic compositions ($\delta^{18}\text{O}$ and δD) in lake water and precipitation, as well as quantified the E/I ratio based on the isotopic evaporative enrichment of lake water in Lake Qinghai Basin. The results indicated that precipitation had a wider range of isotopic compositions and higher depleted isotope values than lake water. The effect of evaporative isotopic enrichment on lake water was also revealed being a lower average d-excess value (-4.91‰) and lower values of slope and intercept (5.88 and -2.41 , respectively), compared with those values in precipitation. The signatures of isotopic compositions in precipitation were forced by different local hydrological processes and moisture sources from continental air masses.

The average E/I ratio that was calculated by $\delta^{18}\text{O}$ and δD was 0.40. The changes of E/I ratios (from 0.29 to 0.36) clearly revealed the shifts in lake levels of Lake Qinghai in recent years (2004–2011). This study can provide some implications for studying the paleoclimate and paleolake level in this region. The isotopic compositions in lake water and E/I ratios showed various degrees of isotopic fractionation that accompanied different lake levels. These results can provide some important evidences, that is, isotopic compositions in lake water and sedimentary materials are indicators for evaluating the lake hydrological balance and water budget.

Acknowledgments

This study was financially supported by the National Natural Science Foundation of China (41130640, 91425301, 41321001, 41401057) and the projects from the State Key Laboratory of Earth Surface Processes and Resource Ecology, Beijing Normal University. We appreciate Dr. CUI Buli for providing some stable isotopic data of lake water and precipitation.

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