Spatial distribution and controlling factors of stable isotopes in meteoric waters on the Tibetan Plateau: implications for paleoelevation reconstruction

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Abstract

Debates persist about the interpretations of stable isotope based proxies for the surface uplift of the central–northern Tibetan Plateau. These disputes arise from the uncertain relationship between elevation and the $\delta^{18}\text{O}$ values of meteoric waters, based on modern patterns of isotopes in precipitation and surface waters. We present a large river water data set (1,340 samples) covering most parts of the Tibetan Plateau to characterize the spatial variability and controlling factors of their isotopic compositions. Compared with the amount-weighted mean annual oxygen isotopic values of precipitation, we conclude that river water is a good substitute for isotopic studies of precipitation in the high flat (e.g., elevation > 3,300 m) interior of the Tibetan Plateau in the mean annual timescale. We construct, for the first time based on field data, contour maps of isotopic variations of meteoric waters ($\delta^{18}\text{O}$, δD and $d\text{-excess}$) on the Tibetan Plateau. In the marginal mountainous regions of the Plateau,

25 especially the southern through eastern margins, the $\delta^{18}\text{O}$ and δD values of river
26 waters decrease with increasing mean catchment elevation, which can be modeled as
27 a Rayleigh distillation process. However, in the interior of the Plateau (north of
28 30°N /Trans-Himalaya range), northward increasing trends in $\delta^{18}\text{O}$ and δD values are
29 pronounced and present robust linear relations; d-excess values are lower than the
30 marginal regions and exhibit distinct contrasts between the eastern (8‰–12‰) and
31 western (<8‰) Plateau. We suggest that these isotopic features of river waters in the
32 interior of the Tibetan Plateau result from the combined effects of: 1) mixing of
33 different moisture sources transported by the South Asian monsoon and Westerly
34 winds; 2) contribution of moisture from recycled surface water; and 3) sub-cloud
35 evaporation. We further provide a sub-cloud evaporation modified Rayleigh
36 distillation and mixing model to simulate the isotopic variations in the western Plateau.
37 Results of this work suggest that stable isotope-based paleoaltimetry studies are
38 reliable in the southern through eastern Plateau margins; towards the central–northern
39 Plateau, this method cannot be applied without additional constraints and/or large
40 uncertainties.

41
42 **Key words:** Tibetan Plateau, river water, stable isotope, paleoaltimetry, sub-cloud
43 evaporation, back trajectory analysis

1. Introduction

Observations that the isotopic values ($\delta^{18}\text{O}$ and δD) of meteoric waters (e.g., precipitation and river water) decrease with increasing elevation (Dansgaard, 1964; Garzione et al., 2000b; Gonfiantini et al., 2001; Poage and Chamberlain, 2001; Rozanski et al., 1993) and that authigenic carbonates can preserve the isotopic composition ($\delta^{18}\text{O}$) of paleo-meteoric waters (Cerling and Quade, 1993; Hoke et al., 2009; Talbot, 1990), led to the application of stable isotope-based paleoaltimetry to reconstruct the surface uplift history of orogenic belts and plateaus (Chamberlain and Poage, 2000; Garzione et al., 2000a; Rowley et al., 2001). Applications of this method in the southern Tibetan Plateau yield consistent results between different researchers (e.g., Currie et al., 2005; Ding et al., 2014; Garzione et al., 2000a; Rowley et al., 2001; Saylor et al., 2009), that also agree well with other paleoelevation proxies, such as foliar physiognomy and clumped isotope studies (Huntington et al., 2014; Spicer et al., 2003). However, using the same method to interpret topographic growth in the central–northern Tibetan Plateau is debated. For example, paleoelevation estimations for the Eocene and Miocene east Hoh Xil basin could differ by more than 3,500 m and 4,000 m, respectively (Bershaw et al., 2012; Cyr et al., 2005; Polissar et al., 2009; Quade et al., 2011). To a large extent, these disputes arise from the uncertain relationship between elevations and $\delta^{18}\text{O}$ values of meteoric waters associated with competing fractionation processes in this deep continental setting (Bershaw et al., 2012; Hren et al., 2009; Tian et al., 2007; Yao et al., 2013), which result in large deviations from the isotopic values predicted from a simple Rayleigh distillation process on which stable isotope-based paleoaltimetry is based (Rowley and Garzione, 2007; Rowley et al., 2001).

This study addresses the spatial distribution and controlling factors of meteoric water isotopes on the Tibetan Plateau. As a direct condensate from the atmosphere, precipitation is the most commonly used source for studying meteoric waters. However, due to the limited and uneven distribution of monitoring stations on the Tibetan Plateau (Fig. 1), precipitation is a less ideal object to study isotopic variations on a plateau scale. In situations like this, surface waters, especially river water, may provide a reasonable substitute for precipitation. River water is mainly supplied by precipitation that infiltrates the groundwater table or flows overland. River water can capture the isotopic compositions of local precipitation (Kendall and Coplen, 2001; Rowley and Garzione, 2007). In addition to the fact that river waters are much easier to sample, their isotopic compositions represent the amount-weighted average value of precipitation in the upstream catchment area. If sampled outside of the main rainy season, the isotopic compositions of stream water reflect those of groundwater input. Although infiltration to groundwater might be biased to times when precipitation is greater than potential evaporation, it has been argued that the isotopic compositions of river water can provide an amount-weighted integration of annual precipitation, information that is useful in constructing the elevation- $\delta^{18}\text{O}$ relation for stable isotope-based paleoaltimetry studies (Rowley and Garzione, 2007).

Several studies focusing on river waters have been carried out on the Tibetan Plateau (Bershaw et al., 2012; Ding et al., 2009; Hren et al., 2009; Liu, 2014; Quade et al., 2011; Xu et al., 2014). These studies confirm the competing influence of the South Asian monsoon and Westerly moisture sources (Hren et al., 2009), as well as the important role of local surface water recycling (Bershaw et al., 2012). However, large gaps in river water sampling from the western, central and eastern Plateau (Fig.

1) leave challenges in interpreting the controlling factors for isotopic variations across the whole Tibetan Plateau region.

In this study, we carried out wide-spread sampling of river waters in parts of the Tibetan Plateau that still lack data (Fig. 1, red dots). In combination with previous published river water data (Bershaw et al., 2012; Ding et al., 2009; Hoke et al., 2014; Hren et al., 2009; Liu, 2014; Liu et al., 2014; Quade et al., 2011; Xu et al., 2014), this large data set improves our knowledge of the spatial variability of stable isotopic compositions of river waters on the Tibetan Plateau, and also allows us to address the following major questions: 1) Is river water a good substitute for precipitation to study the spatial isotopic variations on the Tibetan Plateau on the mean annual timescale? and 2) What are the controlling factors for isotopic variations of meteoric waters (river water/precipitation) on the Tibetan Plateau? We address the first question through a comparison of river water data with available precipitation data, and the second question by looking at the spatial variations of $\delta^{18}\text{O}$, δD and d-excess values. Finally, with this new understanding, we discuss the implications for stable isotope-based paleoaltimetry studies in the Tibetan Plateau.

Fig. 1. Topographic relief map of the Tibetan Plateau, showing river water sampling locations: Red dots, from this study; Blue, green and cyan squares, from literature 1–9. Big thick arrows

indicate major external air masses of the Tibetan Plateau (brown: South Asian monsoon; pink: East Asian monsoon; yellow: Westerlies; Araguás-Araguás et al., 1998). Large white letters are major mountain chains. Two east–west trending dashed lines are boundaries between three isotopic domains (northern, central and southern; Yao et al., 2013). Large black dots are precipitation monitoring stations. References: 1, Bershaw et al., 2012; 2, Hoke et al., 2014; 3, Hren et al., 2009; 4, Liu, 2014; 5, Quade et al., 2011; 6, Xu et al., 2014; 7 Ding et al., 2009; 8, Garzzone et al., 2004; 9, Bai et al., 2012.

2. Geography and moisture transport

2.1 Geography

The Tibetan Plateau is the highest and largest topographic feature on Earth (Fig. 1). Except the northeast and southeast, all the other margins are bounded by very high elevation (>6,000 m) and steeply sloped (gradients up to 6%) mountain chains; such as the Himalayan Mountains, Longmen Shan, and Kunlun Shan (“Shan” means mountain in Chinese) (Fig. 1). The eastern Plateau (mainly east of 93°E) is characterized by rugged terranes, with high-discharge, south–southeastward flowing rivers cutting deeply (2,000–3,000 m) into the landscape. On the contrary, the western Plateau is relatively flat, with most regions between 4,500–5,000 m. Interestingly, this east–west topographic difference is also accompanied by difference in annual precipitation amount: high in the east (>400 mm/year) and low in the west (<400 mm/year) (Maussion et al., 2014).

The northern Plateau, represented by the Qaidam basin and Qilian Shan, is relatively low elevation regions (2,500–4,500 m), and receives little precipitation. The western-most Plateau, locally known as the Pamir Plateau, is also characterized by rugged landscape (Fig. 1). Different from the other parts of the Tibetan Plateau, which are dominated by summer precipitation (>80–90% of annual precipitation amount), the Pamir Plateau receives most of its precipitation during the winter season (Fig. 2).

Fig. 2. Contour map of summer precipitation index, defined as the percentage of summer (May to October) precipitation amount over annual precipitation amount (1980-2010). 0, all winter precipitation; 1, all summer precipitation. Thick red outlines are the 2,000 m altitude contour of the Tibetan Plateau and surrounding regions. Note that all the river water samples in this study are located to the east of 79°E (Fig. 1), which are dominated by summer precipitation (>80%). Original precipitation data is from the NOAA Earth System Research Laboratory's GPCC full data reanalysis V6 combined.

2.2 Moisture origin and transport

There are three main external sources providing moisture for precipitation on the Tibetan Plateau, including the South Asian monsoon, mid-latitude Westerlies and East Asian monsoon (Fig. 1) (Araguás-Araguás et al., 1998; Tian et al., 2007). The first two provide the majority of moisture (Chen et al., 2012; Feng and Zhou, 2012), whereas the latter one appears to be significant only at the eastern-most margin and exerts little influence on the remaining Plateau (Curio et al., 2015).

Along the transport path of these air masses into the interior of the Tibetan Plateau, high mountain chains on the margins (Fig. 1) block moisture from entering the Plateau. A large amount of moisture is precipitated when forced to ascend over high relief topography. River valleys that both parallel or cut through the mountain

chains serve as the main channels for external moisture to penetrate the interior of the Plateau, such as the Yarlung Tsangpo valley and Thakkhola graben in the Himalaya Mountain and the Min and Dadu river valleys in the eastern Plateau.

In the interior of the Tibetan Plateau, the relative significance of external moisture, mainly the northward extension of the South Asian monsoon, is not well constrained. Stable isotopic studies of precipitation (Tian et al., 2007; Yao et al., 2013) suggest that regions south of 30°N are dominated by the South Asian monsoon (south domain, Fig. 1); regions north of 35°N are dominated by Westerlies (north domain); while regions in between are transitional between these two different moisture sources (central domain).

3. Methods

3.1 River water sampling

The primary goal for sampling in this study is to cover parts of the Tibetan Plateau that still lack river water data (Fig. 1). Ideally, river water samples should be collected outside the main rainy season and the spring snowmelt season when rivers show base-level flow that is representative of groundwater, the isotopic compositions of which might reflect the amount-weighted mean annual precipitation composition. Samples that lie to the east of 96°E were collected during the late spring and early summer (May–early June) of 2012 and 2013, which is before the main rainy season (middle June–early September). Samples were collected mostly from small (drainage areas <100 km²) and medium-sized rivers (100–1,000 km²; auxiliary Table S1). River water samples in the west–central Plateau were collected in the late autumn (October–middle November) of 2012 and 2013, which is the dry season. In this region, about 60% samples are collected from small rivers or springs (auxiliary Table S1).

3.2 Isotope measurement

All water samples were measured in the SIREAL stable isotope laboratory at the University of Rochester, using a liquid water isotope analyzer (model LWIA-24d) with a GC-PAL auto-sampler. For all water samples, at least two separate analyses were conducted for replication of results. Each analysis consists of 14 water injections, with a minimum of 7 injections, excluding the first 3, required to calculate the isotopic composition. Analytical error is determined from the mean of multiple injections. Two in-house standards, both calibrated to Vienna Standard Mean Ocean Water (VSMOW) and Vienna Standard Light Antarctic Precipitation (VSLAP), were analyzed after every three samples for standardization. Isotopic results for both oxygen and hydrogen are reported using standard delta (δ) notation (‰, per mil) with respect to VSMOW, and analytical errors are better than $\pm 0.1\text{‰}$ (1σ) and $\pm 1.2\text{‰}$ (1σ), respectively.

3.3 Data compilation

The main criterion for use of river water data in this study is that they are representative of the isotopic composition of local meteoric water. We therefore exclude river waters that were sampled from very large trunk rivers because they capture rainfall isotopic signal over a very large drainage area ($>5,000 \text{ km}^2$; 11 samples, auxiliary Table S1). In addition, we exclude river waters that exhibit either anomalous $\delta^{18}\text{O}$ values ($>5\text{‰}$, 2 samples) or anomalous d-excess values ($>5\text{‰}$, 41 samples), compared with the average isotopic values of adjacent river waters within a 50 km radius (auxiliary Table S1).

In total, there are 1,340 river water samples used, including 458 newly collected and 882 compiled samples (Bai et al., 2012; Bershaw et al., 2012; Ding et al., 2009; Garzzone et al., 2004; Hoke et al., 2014; Hren et al., 2009; Liu, 2014; Quade et al.,

2011; Xu et al., 2014). In this data set, there are 1056 samples with both $\delta^{18}\text{O}$ and δD values, 256 samples with only $\delta^{18}\text{O}$ values, and 28 samples with only δD values.

3.4 Contouring of stable isotopic values

Based on the individual river water isotopic values, we construct the contour maps of $\delta^{18}\text{O}$, δD and d-excess values using Kriging gridding with linear variogram. In this method, each grid node value is based on the known data points neighboring the node; and each data point is weighted by its distance away from the node. This gridding method can compensate for clustered data by giving less weight to the cluster in the overall prediction.

We note that although Kriging gridding and contouring interpolated the isotopic values in sparsely sampled regions, such as the Qaidam basin and the northwestern Plateau (Fig. 1), they may be unreliable in these regions. This is also the case in most of the Himalaya region, where samples are lacking and spatial isotopic changes are very large associated with steep topographic gradients (Garzzone et al., 2000b).

3.5 HYSPLIT back trajectory analysis

We use the HYbrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT) (Draxler and Rolph, 2013) to track air parcel trajectories in four locations in the western Plateau (29.5°N, 88.0°E; 32.5°N, 86.0°E; 32.5°N, 90.0°E; 35.0°N, 88.0°E). Meteorological parameters are from NCEP/NCAR reanalysis data (Kanamitsu et al., 2002). For each of the four locations, 72-hour back trajectories were computed every 12 hours for the months of June, July and August from 2010 to 2013, during the years in which new river water samples were collected. These three months are chosen because they reflect the main rainy season on the Tibetan Plateau, which accounts for more than 70% of annual precipitation (Fig. 2). The initial parcel was set at 1 km above ground level, because most moisture in the atmosphere is in the

lower troposphere, within 0-2 km above ground. We note that there is no apparent difference in results for initiation elevations at 0.5, 1.0 and 1.5 km above ground level. For each location, there are 736 back trajectories; however, because the HYSPLIT back trajectory analysis deals with air parcels, and not vapor flux, we only consider precipitation-producing trajectories, which are considered as yielding precipitation within twelve hours before arriving at the location. We compile the end points of precipitation-producing back trajectories and contour them based on their spatial density. We also merge back trajectories that are near each other into clusters that are represented by their mean trajectory.

4. Results and interpretations

River water samples used in this study were collected during different months and years. In the eastern Plateau, Bershaw et al. (2012) collected samples in August of 2008, which is near the end of the rainy season. We collected samples along the same N-S transect as Bershaw et al. (2012) in early May of 2012, before the rainy season. Isotopic values of these two data sets are nearly identical (within 0.5‰ difference). This is also the case (similar isotopic values between different data sets regardless of the month of sampling) in the west-central Plateau. Given that the rainy season provides >80% precipitation of total annual precipitation on the Tibetan Plateau (Fig. 2), we infer that both the groundwater and river water are dominated by the isotopic composition of rainy season precipitation.

4.1 $\delta^{18}\text{O}$ and δD values and local meteoric water lines

The $\delta^{18}\text{O}$ values of river waters across the Tibetan Plateau range between -20.8‰ and -3.4‰, and the δD values range between -165‰ and -19‰ (Fig. 3, see auxiliary Table S1 for all the isotopic data). The local meteoric water line of Tibetan river water is $\delta\text{D}=(8.55\pm0.05)*\delta^{18}\text{O}+(16.53\pm0.67)$ ($R^2=0.97$, $n=1,056$), which is

steeper than the Southeast Asian precipitation line (including the Tibet area):
 $\delta D = 7.92 * \delta^{18}O + 9.20$ (Araguás-Araguás et al., 1998). Fig. 3 also shows the local lake
water line of the Tibetan Plateau: $\delta D = (6.40 \pm 0.24) * \delta^{18}O - (24.37 \pm 1.74)$, ($R^2 = 0.85$,
 $n = 121$; for data, see auxiliary Table S2). The lower slope of the lake water line can be
attributed to non-equilibrium (kinetic) evaporative fractionation between lake water
and water vapor (e.g., Gonfiantini, 1986). The majority of river water values plot on a
line of steeper slope compared to the lake water line, suggesting insignificant surface
water evaporation, compared with the strong evaporation of lake water.

Fig. 3. δD - $\delta^{18}O$ relationship of river waters (grey open diamonds) and lake waters (red solid triangles) on the Tibetan Plateau. Also shown are the local meteoric water line and local lake water line.

Following Yao et al. (2013)'s division of the Tibetan Plateau into three domains (Fig. 1), the local meteoric water lines for the northern, central and southern domains are: $\delta D = (8.42 \pm 0.20) * \delta^{18}O + (17.20 \pm 1.65)$ ($R^2 = 0.97$, $n = 67$), $\delta D = (8.32 \pm 0.10) * \delta^{18}O + (12.83 \pm 1.26)$ ($R^2 = 0.93$, $n = 615$), and $\delta D = (8.89 \pm 0.06) * \delta^{18}O + (23.02 \pm 1.02)$ ($R^2 = 0.98$, $n = 374$), respectively. The central domain can be further divided into the western (internally drained) and eastern (externally drained) parts (roughly along the 93°E), and the water lines for these two parts are $\delta D = (7.91 \pm 0.15) * \delta^{18}O + (4.09 \pm 1.97)$

($R^2=0.91$, $n=277$), and $\delta D=(8.46\pm0.07)*\delta^{18}O+(17.26\pm0.93)$ ($R^2=0.98$, $n=338$), respectively. We note that the slopes of meteoric water lines in the northern and central domains are lower (0.47 and 0.57 respectively) than that in the southern domain, which is also the case for the western central domain (0.55 lower), compared with that in the eastern central domain. Lower slopes suggest a stronger influence from evaporation, which we interpret as stronger sub-cloud evaporation (discussed in detail in section 6.2.3).

4.2 Spatial distribution of $\delta^{18}O$ and δD values

The spatial distribution of individual $\delta^{18}O$ and δD values of river waters and corresponding contour maps are presented in Fig. 4A–B and C–D, respectively (also auxiliary kmz file S1). We do not display the contour lines in the Himalaya Mountain (white regions in Fig. 4C–D) due to the lack of samples (Fig. 1). The Kriging gridding method results in contours that are inconsistent with observed patterns along well-sampled transects in this region (Bai et al., 2015; Garzzone et al., 2000b; Quade et al., 2011). On the contrary, in the eastern margin (Longmen Shan), where there are a lot of data (Fig. 1), the contours reflect theoretical predictions from Rayleigh distillation.

Fig. 4. A and B: 3,000 m altitude contour map (grey shades) of the Tibetan Plateau with locations of river water samples, that are color coded by $\delta^{18}\text{O}$ (A) and δD (B) values. Note that the samples used for these two isotopes are slightly different (see section 3.3). **C and D:** Contour maps of $\delta^{18}\text{O}$ (C) and δD (D) values of river waters on the Tibetan Plateau. The thick dark grey lines are 3,000 m altitude contours. The four black dots in C show locations for back trajectory analysis (see section 6.2.1 and Fig. 8).

The $\delta^{18}\text{O}$ values of river waters in the marginal high mountains of the Tibetan Plateau decrease with increasing elevation; for example, along the southern slope of the Himalaya (Fig. 5A) and the eastern slope of the Longmen Shan (Fig. 5B). This trend is less apparent in the northern slopes of the Qilian Shan and West Kunlun Shan. In the central and northern Plateau, there is a pronounced northward increase in the $\delta^{18}\text{O}$ values (from $\sim -18\text{‰}$ to $< -7\text{‰}$). This trend reflects a linear relationship between latitude ($^{\circ}\text{N}$) and $\delta^{18}\text{O}$ values (‰): $\delta^{18}\text{O} = (1.13 \pm 0.03) * \text{Lat} - (49.78 \pm 0.85)$ ($R^2 = 0.61$, $n = 1176$, Fig. 5E). The strength of the correlation improves when focusing on three north–south transects in the eastern ($\sim 96\text{--}98^{\circ}\text{E}$), middle ($\sim 92\text{--}93^{\circ}\text{E}$) and western ($\sim 88\text{--}89^{\circ}\text{E}$) Plateau ($R^2 = 0.84$, 0.85 and 0.84 , respectively). No correlations between longitude and $\delta^{18}\text{O}$ values are found.

4.3 Spatial distribution of d-excess values

The d-excess value ($\text{d-excess} = \delta\text{D} - 8 * \delta^{18}\text{O}$) of meteoric water reveals the difference in kinetic fractionation between $\text{HD}^{16}\text{O}/\text{HH}^{18}\text{O}$ and HH^{16}O during phase change under conditions where relative humidity is $< 100\%$ (Dansgaard, 1964; Froehlich et al., 2008). The spatial distributions of d-excess values of river waters and their corresponding contour map are presented in Fig. 6A–B (also auxiliary kmz file S1).

Fig. 5. Plots of $\delta^{18}\text{O}$ -elevation (A and B) and d-excess-elevation (C and D) variations at four different transects in the southern Plateau Himalaya Mountain and eastern Plateau Longmen Shan. Note that for the Himalaya southern slope at 83.8°E, there are fewer data points for the d-excess plot compared with the $\delta^{18}\text{O}$ plot, due to lack of availability of δD /d-excess values. Data sources: Himalaya southern slope: at 83.8°E, Garzzone et al., 2000b; at 85.9°E, Bai et al., 2015; Longmen Shan eastern slope: at 31.1°N, Xu et al., 2014; at 30.0°N, this study. E and F, plots of $\delta^{18}\text{O}$ -latitude (E) and d-excess-longitude (F) variations for river water samples on the Tibetan Plateau.

Generally, the d-excess values range between -12‰ to 20‰. We group their spatial variations into three groups: 1) very high d-excess values between 12‰ and

20‰ in the marginal high mountains of the Tibetan Plateau (Fig. 6B), which show various trends (increase, decrease or remaining nearly the same) with increasing elevation (Fig. 5C–D). In the interior of the Plateau, two sub-regions can be clearly identified: 2) low d-excess values between -12‰ and 8‰ in the western Plateau; and 3) relatively high values (8‰–12‰) in the eastern Plateau (Fig. 5F and 6B).

Although the dominant river size is medium (59%) in the eastern Plateau and small (58%) in the western Plateau, we argue that the east–west contrast in d-excess values is not a result of sampling bias. Comparing any group (e.g., large, medium or small) of river water data, the d-excess values in the western Plateau are ~9‰ lower than rivers of similar size in the east. For example, comparing the d-excess values of medium-sized rivers (1.4‰) in the western Plateau with the average values of all samples (10.3‰) in the eastern Plateau (dominated by medium sized rivers), there is a ~9‰ difference between the east and west. 41 samples are excluded as a result of anomalous d-excess values (auxiliary Table S1), although inclusion of these data does not change the observation of the large contrast in d-excess values between the eastern and western Plateau.

Fig. 6. A: 3,000 m altitude contour map (grey shades) of the Tibetan Plateau with locations of river water samples that are color coded by d-excess values. **B:** Contour maps of d-excess values of river waters on the Tibetan Plateau. The thick dark grey lines are 3,000 m altitude contours. The four black dots in B show locations for back trajectory analysis (see section 6.2.1 and Fig. 8). The black dotted boxes show areas with large d-excess differences in the interior of the Plateau.

5. Validity of river water as a substitute for isotopic studies of precipitation

As most of the $\delta D/d$ -excess values of precipitation on the Tibetan Plateau are not reported in previous studies (e.g., Liu et al., 2008; Yao et al., 2013), we compare the oxygen isotopic composition to assess the validity of using river water as representative of the amount-weighted mean annual isotopic composition of precipitation (Fig. 7 and auxiliary Table S3). On the plateau scale, the mean annual oxygen isotopic value of precipitation increases from $\sim -18\text{‰}$ in the south (e.g. station Dui) to $\sim -7\text{‰}$ in the north (e.g. station Zhangye). A linear relationship between the $\delta^{18}O$ values of precipitation and latitude also exists: $\delta^{18}O = (0.96 \pm 0.17) * \text{Lat} - (43.47 \pm 5.35)$ ($R^2 = 0.63$, $n = 22$), which is very close to that of river water (Fig. 5E).

We compare the mean annual $\delta^{18}O$ values of precipitation with the river water $\delta^{18}O$ contour in which the precipitation station falls. Among the 22 stations (Fig. 7 and auxiliary Table S3), there are 12 (stations 1–12) that show good agreement (within $\pm 1\text{‰}$) between the $\delta^{18}O$ values of precipitation and the surrounding river water contour. If only consider the 14 high elevation stations ($> 3,300$ m), 10 of them show good agreement. The 8 stations that lie in the piedmont lowland regions (stations 15–22) show higher $\delta^{18}O$ values than adjacent river water. The lower $\delta^{18}O$ values of the river water likely reflect the integration of isotopic signals of precipitation that fell in the much higher elevation catchment areas ($> 1,000$ m elevation difference, auxiliary Table S3). There are only two stations (stations 13–14) that show lower $\delta^{18}O$ values than the contour defined by river water data, which suggests that the local river water may have experienced evaporation.

Fig. 7. Comparisons between the $\delta^{18}\text{O}$ values of amount-weighted mean annual precipitation in 22 monitoring stations (red triangles), individual river waters (grey bars) and their average values (blue bars with vertical lines) within 50 km radius of monitoring station, and contoured $\delta^{18}\text{O}$ values around monitoring station (pink vertical bands). Inset: Geographic locations of precipitation observation stations (dots) on the Tibetan Plateau. The color codes indicate contoured $\delta^{18}\text{O}$ values of river water that are in agreement with (black), or higher (red), or lower (green) than the mean annual $\delta^{18}\text{O}$ values of precipitation.

Several lines of evidence suggest that evaporation of river water on the Tibetan Plateau is not significant. First, most of the river water data points fall to the left of the local lake water line, and only a few of them are located to the right of the line (Fig. 2), suggesting a small percentage of the water samples experienced surface water evaporation. Second, comparison of the $\delta^{18}\text{O}$ values of river water with precipitation stations shows that most of the precipitation values agree with adjacent river water values, especially in the high flat interior of the Plateau, with only 2 out 14 of the sites suggesting possible evaporation from river water (Fig. 7). Due to the lack of availability of δD and d-excess values of precipitation, we can not completely rule

out the possibility of some degree of surface water evaporation and/or sublimation of snow that contribute to the $\delta^{18}\text{O}$ signals of river waters in this region. However, it should be insignificant, considering the small difference in $\delta^{18}\text{O}$ values of river water with precipitation.

The comparable nature of $\delta^{18}\text{O}$ values of river water and precipitation, and the lack of evidence of significant surface water evaporation indicate that river water is a good substitute for the amount-weighted mean annual isotopic composition of precipitation in the high flat interior (elevation > 3,300 m) of the Tibetan Plateau.

6. Controls on the spatial isotopic variations of river water

6.1 Margins of the Tibetan Plateau

At the margins of the Tibetan Plateau, especially the southern and eastern margins (windward slopes of the Himalaya and Longmen Shan, Fig. 1), the $\delta^{18}\text{O}$ values of river waters decrease with increasing elevation (Fig. 5A–B). The d-excess values show differing trends in different locations; they either increase, or decrease, or are more complex (Fig. 5C–D) with increasing elevation. Decreasing $\delta^{18}\text{O}$ values with increasing elevation is a typical feature of the altitude effect of meteoric waters (Dansgaard, 1964; Rozanski et al., 1993). Early studies tended to consider d-excess values to be unchangeable after moisture evaporated from the ocean surface (Craig and Gordon, 1965; Merlivat and Jouzel, 1979). However, later model simulations following a Rayleigh distillation process show that the d-excess values of precipitation can either increase or decrease, due to the differences in relative humidity and temperature during precipitation (Gonfiantini et al., 2001; Jouzel and Merlivat, 1984).

We consider the Rayleigh distillation process a reasonable fractionation mechanism to account for the isotopic variations of river waters at the southern

through eastern margins of the Tibetan Plateau (See Bershaw et al. (2012) for Rayleigh distillation simulation in the Himalaya Mountains). We note that in the northern mountainous margins (Kunlun Shan and Qilian Shan, Fig. 1), the Rayleigh distillation process alone cannot explain the observed isotopic variations. Although there is still a decreasing trend in the $\delta^{18}\text{O}$ values of river waters with increasing elevation, this altitudinal isotopic gradient is greatly damped to $\sim 1\text{‰}/\text{km}$, compared with $\sim 3\text{‰}/\text{km}$ on the other margins, indicating that other fractionation mechanisms (e.g., addition of recycled surface water and sub-cloud evaporation) are required to account for the low $\delta^{18}\text{O}$ versus altitude gradient.

6.2 Interior of the Tibetan Plateau

6.2.1 Different moisture sources

There are three external sources providing moisture for precipitation on the Tibetan Plateau: the South Asian monsoon, the Westerlies, and the East Asian monsoon (Fig. 1; Araguás-Araguás et al., 1998). The $\delta^{18}\text{O}$ patterns of surface waters in the eastern Plateau do not support the East Asian monsoon as a major source of moisture to the Plateau, given the lack of observed east–west $\delta^{18}\text{O}$ gradients. Pronounced east–west $\delta^{18}\text{O}$ gradients are only observed in the eastern-most part of the Plateau, suggesting this region as the limit of influence of the East Asian monsoon. $\delta^{18}\text{O}$ values show significant gradients in the south–north direction across the entire Tibetan Plateau (Fig. 4C), suggesting the dominance of the South Asian monsoon and Westerlies. This conclusion is supported by studies of atmospheric vapor flux across all the margins of the Tibetan Plateau (Curio et al., 2015), and Lagrangian particle dispersion modeling work (Chen et al., 2012), which suggest the main moisture flux onto the Plateau comes from the south, west and north.

By determining the back trajectories at nine different locations in the east and central Plateau, Bershaw et al. (2012) show that in the southeast and east-central Plateau, external moisture is mainly derived from the South Asian monsoon. In the western Plateau, our back trajectory studies show that in the southern most site (Fig. 8A), moisture is mainly derived from the South Asian monsoon (>70%), whereas the remaining ~30% is from the north, transported by the Westerlies. Toward the north and west (Fig. 8B–C), air masses from the South Asian monsoon decrease, and moisture from the north increases. In the northern most site (Fig. 8D), Westerly moisture dominates to account for about 70%, whereas moisture from the South Asian monsoon decreases to ~30%.

Fig. 8. HYSPLIT back trajectory end points of summertime (June, July, August; 2010–2013) air parcels (black dots) 72 hour prior to their arrival at one of the four selected locations in the western Plateau (big grey/red solid circles). The value of “n” indicates the number of precipitation- producing trajectories. Color shades show spatial density of back trajectory end points. Red curves show clusters of back trajectories; red squares are locations for every 24 hours; the percentages indicate how many trajectories merged in this cluster. Black lines show the 3,000 m altitude contour of the Tibetan Plateau.

6.2.2 Surface water evaporation and recycling

Although there are two major external moisture sources that influence the isotopic variations of meteoric waters on the Tibetan Plateau, a simple mixing of these two different moisture sources can not fully explain the spatial isotopic variations across the Tibetan Plateau (Bershaw et al., 2012). As shown in Fig. 4C, in the central plateau (between 30°N and 35°N), the $\delta^{18}\text{O}$ values are nearly the same at the same latitude. However, back trajectory analyses for locations at similar latitude in the western (Fig. 8B–C) and eastern Plateau (Bershaw et al., 2012) show various contributions of moisture from different sources. If mixing of these two external moisture sources were the dominant factor to control the isotopic variations, different $\delta^{18}\text{O}$ values along lines of latitude, or more complex patterns, should be observed, which is not the case. In addition, the two moisture mixing model cannot account for the observed d-excess variations that show a large contrast between the eastern and western Plateau (Fig. 6B). Therefore, other factors must also contribute to the isotopic signals.

In continental settings, evaporation and transpiration of different surface waters (e.g., lake water, soil water and plant water) can be a significant contributor to atmospheric water vapor. A 12-year high-resolution climatology of the atmospheric vapor transport over and towards the Tibetan Plateau indicates that only ~37% of the moisture required for precipitation on the Tibetan Plateau is derived from outside of the Plateau, which requires that ~63% of moisture comes from local surface water evaporation and recycling (Curio et al., 2015). Stable isotopic studies of precipitation, near-surface atmospheric vapor and evapotranspiration in the central Plateau also indicate that locally recycled moisture plays an important role in contributing to precipitation (Kurita and Yamada, 2008). Based on these observations, Bershaw et al.

(2012) used a surface water evaporation modified Rayleigh distillation model to simulate the observed isotopic variations in surface waters and precipitation in the east-central Plateau.

6.2.3 Sub-cloud evaporation as a significant controlling factor

Neither mixing of different moisture sources nor addition of recycled surface water can account for the large difference in d-excess values between the eastern and western Plateau (Figure 5), as the second factor would lead to increase in d-excess values. Although lower condensation temperature can result in lower d-excess values, there is no observable difference in air temperature in the western compared to the eastern Plateau, during the summer time when most precipitation occurs (MauSSION et al., 2014). There is also no difference in the seasonality of precipitation (Fig. 2) that could contribute to the east-west d-excess contrast. We propose that sub-cloud evaporation of raindrops might explain the anomalously low d-excess values in the western Plateau.

Raindrops falling through an unsaturated air column will partially evaporate, leading to enrichment of heavy isotopes in precipitation (increase $\delta^{18}\text{O}$ and δD values; e.g., Lee and Fung, 2008). At the same time, kinetic fractionation due to low relative humidity will lead to more ^{18}O enrichment in the raindrop relative to ^2H (Gonfiantini, 1986), thus decreasing the d-excess values. Studies have shown that the intensity of sub-cloud evaporation increases with lower relative humidity and smaller raindrop radius (Lee and Fung, 2008), and that greater intensity of sub-cloud evaporation is also associated with smaller rainfall amount per precipitation event (e.g., Chen et al., 2015; Peng et al., 2007).

On the Tibetan Plateau, the amount of mean annual precipitation decreases from east (>400 mm) to west (<200 mm) (Fig. 9A). Available data suggests the

515 rainfall amount per precipitation event is also higher in the east (~7.5 mm; Tian et al.,
516 2001) than in the west (~5.0 mm in Gaize; Yu et al., 2007). We predict that smaller
517 rainfall amount (Fig. 9A) and lower mean relative humidity (Fig. 9B) in the western
518 Plateau would result in stronger sub-cloud evaporation (more fraction of raindrops
519 will be evaporated), leading to lower d-excess values of raindrops. In the eastern
520 Plateau, heavier rainfall that passes through a higher relative humidity air column
521 would reduce sub-cloud evaporation, and thus preserve higher d-excess values.
522 Similar observations (higher d-excess value - greater rainfall amount; lower d-excess -
523 less rainfall) also exist in the southeastern Plateau, on the northern and southern side
524 of the Yarlung Tsangpo River (Fig. 6B, small box). The spatial correlation coefficient
525 between d-excess values with mean precipitation amount and relative humidity are
526 0.44 and 0.40, respectively (Fig. 9C; both are significant above the 99% confidence
527 level using the r-test), further supporting the correlations between d-excess values and
528 these two climate parameters.

Fig. 9. Main rainy season (JJA: June, July, August for 2001–2013) precipitation amount (**A**) and relative humidity (**B**) for the Tibetan Plateau area (Maussion et al., 2014). Black arrows in A are wind direction and speed at 10 m above ground. Arrows A–A' and B–B' show simulated moisture transport routes in the western Plateau. Yellow boxes show east/high vs. west/low contrasts. **C**, correlations between d-excess values with annual mean relative humidity and precipitation amount.

We model the isotopic variations during sub-cloud evaporation within a modified Rayleigh distillation process to determine whether simulations provide a match to observations in the western Tibetan Plateau. We consider two separate simulations, following two generalized trajectories: A–A' and B–B' (Fig. 9A–B), representing moisture transport from the South Asian monsoon and the Westerlies, respectively (section 6.2.1) (see auxiliary appendix for method description and auxiliary Table S4 for calculation spreadsheet). Our simulations show that along both

543 of the two routes (A–A', from south to north; and B–B', from north to south), the
 544 $\delta^{18}\text{O}$ values decrease with progressive rainout (Fig. 10, open diamonds and squares).
 545 However, after considering mixing of these two moisture sources (mixing ratios are
 546 estimated from back trajectory analysis), the modeled $\delta^{18}\text{O}$ values show a northward
 547 increasing trend (Fig. 10, solid triangles). Along both of the routes, the d-excess
 548 values first decrease and then increase (Fig. 10, plus signs and times signs), as a result
 549 of the variations of the percentage of sub-cloud evaporation associated with variations
 550 of relative humidity and rainfall amount (Fig. 9A–B). After mixing, the d-excess
 551 values show lower values in the central and higher values in the northern and southern
 552 ends (Fig. 10, solid circles). Both of the simulated $\delta^{18}\text{O}$ and d-excess values follow
 553 the observed variations in the western Plateau (Fig. 10, pink and green shaded areas).
 554 These simulation attempts suggest that in the western Tibetan Plateau, the spatial
 555 variations of $\delta^{18}\text{O}$ and d-excess values result from at least two major factors: 1)
 556 mixing of moisture from both the north (Westerlies) and south (South Asian monsoon)
 557 and 2) sub-cloud evaporation.

558 **Fig. 10.** Modeled isotopic variations (d-excess and $\delta^{18}\text{O}$ values) of precipitation in two idealized
 559 trajectories and their mixing in the western Tibetan Plateau: A–A', from south to north, moisture
 560

from the South Asian monsoon; B–B', from north to south, moisture from the Westerlies. The red and green shaded areas outline observed $\delta^{18}\text{O}$ and d-excess values between 30°N and 36°N, respectively. The mixing ratios (moisture from the south: moisture from the north) for each step are estimated from back trajectory analysis and are (from south to north): 0.65:0.35, 0.55:0.45, 0.40:0.60, 0.30:0.70, 0.20:0.80, 0.15:0.85.

Our simulations are simple and idealized, compared with the complex spatial variations of climatic parameters (relative humidity and rainfall amount) in the western Plateau (Fig. 9A–B). In addition, we did not consider the addition of recycled local surface water, which has been shown to be important on the Tibetan Plateau (Bershaw et al., 2012; Curio et al., 2015). The value of this modeling exercise is to demonstrate that through a simplistic set of calculations, the variations in $\delta^{18}\text{O}$ and d-excess values in the western Tibetan Plateau can be accounted for by sub-cloud evaporation of raindrops and moisture mixing; especially the anomalously low d-excess values, which are difficult to produce by other atmospheric processes (e.g., seasonality and condensation temperature).

Yet another process that might contribute to the low d-excess values in the western Plateau is sublimation of snow. Similar to kinetic fractionation during evaporation, sublimation can cause a decrease in the d-excess value of residual snow. As a potentially important supply of river water and groundwater, the isotopic composition of snowmelt could also influence river water composition. Although this study cannot rule out the potential contribution of snowmelt to river water in the western Tibetan Plateau, future studies focused on determining the isotopic composition of surface snow and rainfall in the western Plateau region could resolve the relative contributions of sublimation from snow versus sub-cloud evaporation from raindrops.

7. Implications for stable isotope-based paleoaltimetry

The application of stable isotope-based paleoaltimetry relies on assumptions that the isotopic evolution of rainfall follows equilibrium fractionation during Rayleigh distillation of an ascending vapor mass to produce uncertainties that allow for the detection of elevation changes (Rowley and Garzione, 2007; Rowley et al., 2001). Additional fractionation processes that vary strongly as a function of climate aridity or extreme continentality (e.g., surface water recycling, sub-cloud evaporation, and vapor mixing, etc.) result in uncertainties in stable isotope paleoaltimetry estimates that are too large to apply these methods with confidence. Considering that the observed systemic decreases in $\delta^{18}\text{O}$ values with increasing elevation in the southern through eastern margins of the Tibetan Plateau (Fig. 5A–B) can be characterized by Rayleigh distillation (sections 6.1), we infer that stable isotope-based paleoaltimetry is applicable in these regions. On the contrary, in the central–northern Plateau (north of 30°N), where isotopic compositions deviate from predictions of Rayleigh distillation, as a result of the combined effects of mixing of different moisture sources, addition of recycled surface water and sub-cloud evaporation (section 6.2), stable isotope-based paleoaltimetry cannot be applied without additional constraints and/or large uncertainties.

Two methods have been proposed to solve this problem in the central–northern Tibetan Plateau. The first method uses a different local isotope-altitude gradient (Ding et al., 2009; Polissar et al., 2009; Xu et al., 2013), whereas the second method corrects for the effect of latitudinal isotopic changes (Fig. 5E) (Bershaw et al., 2012; Quade et al., 2011). Although both methods may be applicable under the same climate and geography conditions as today, both methods suffer when applied to different paleoclimate and paleogeographic conditions (Botsyun et al., 2016; Roe et al., 2016).

For example, past differences in the position of India and the distribution of inland-sea and topographic growth over the Cenozoic have resulted in significant changes in the location of arid belts and high summertime precipitation (Roe et al., 2016). These climatic and geographic differences result in significant changes in moisture sources, recycling of surface water, and sub-cloud evaporation in different parts of the (modern) plateau, leading to large deviations in the pattern of isotopes across this region. Unless each of the processes can be constrained in the geologic past, the large deviations from Rayleigh distillation isotopic patterns, as observed in the Tibetan Plateau and Asian interior today, introduce large uncertainties in paleoaltimetry that make this method inapplicable outside of regions that have adhered to a simple Rayleigh distillation fractionation process.

These dilemmas signify the need to continue to develop new proxies for paleoelevation that rely on different atmospheric parameters, such as temperature and pressure, and to apply these multiple proxies where possible, although each with their own limitations. In addition, continued work toward accurate characterization of observations of both δD and $\delta^{18}O$ in precipitation within general circulation models is necessary to integrate both numerical simulation efforts and stable isotope-based paleoaltimetry.

8. Summary

To understand the controlling factors of modern surface water isotopic variations and caveats to applying stable isotope-based paleoaltimetry in the Tibetan Plateau, 1,340 modern surface water samples (458 newly collected) across a large area of the Tibetan Plateau were compiled. By comparing river water $\delta^{18}O$ values with the amount-weighted mean annual oxygen isotopic values of precipitation, we suggest that river water is a good substitute for isotopic studies of precipitation in the

high flat interior of the Tibetan Plateau on the mean annual timescale. The isotopic contour maps ($\delta^{18}\text{O}$, δD and d-excess) of modern surface waters on the Tibetan Plateau can provide validation criteria for isotopic simulations using general circulation models.

Different factors control the isotopic variations in different parts of the Tibetan Plateau. For the marginal regions, in the windward parts of the southern (Himalaya), southeastern (Yunnan Plateau) and eastern (Longmen Shan) margins, the decreasing $\delta^{18}\text{O}$ values with increasing elevations can be modeled as a Rayleigh distillation process, which suggests that stable isotope-based paleoaltimetry works well in the southern through eastern margins of the Plateau. The interior of the Tibetan Plateau is characterized by a northward increasing trend in $\delta^{18}\text{O}$ values, and east (high)-west (low) contrast in d-excess values. We conclude that these regional trends can be best explained by the combined effects of three dominant processes: 1) different moisture sources, 2) local surface water recycling, and 3) sub-cloud evaporation. Our modeling of Rayleigh distillation including the effects of sub-cloud evaporation and moisture mixing provides a means of reproducing the isotopic patterns in the western Tibetan Plateau, including observed anomalously low d-excess values. These observations indicate that in the interior of the Tibetan Plateau (north of 30°N), where the $\delta^{18}\text{O}$ values of meteoric waters deviate from predictions of Rayleigh distillation, stable isotope-based paleoaltimetry cannot be applied without additional constraints and/or large uncertainties.

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