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Influence of Evaporating Under the Clouds on the Precipitation Stable Isotope in the Transition Zone Between Tibetan Plateau and Arid Region of China

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Abstract: Consideration of stable isotopes in precipitation is valuable for investigating hydrological processes. Therefore, correcting the measured isotopic composition of precipitation under below-cloud evaporation is necessary. An accurate description of the underlying processes affecting stable isotopic composition of precipitation could help improve our understanding of the water cycle. The transitivity between monsoonal and arid climates was reflected by the evaporation rate of falling raindrops in precipitation in the Qilian Mountains, a typical transition zone between Tibetan Plateau and arid region of China. Considering 1310 precipitation event-scale samples, based on stable isotope analysis method, the mean below-cloud evaporation rate (*f*) in the study area was measured as 12.00% during the summer half-year (May-October). The evaporation rate on the northern slopes (12.70%) of the Qilian Mountains in China was significantly higher than that on the southern slopes (9.98%). The transition between monsoonal and arid climates was reflected in the evaporation rate of falling raindrops during precipitation in the Qilian Mountains of China. Below-cloud evaporation contributed to a noticeable enrichment of stable isotopes in the precipitation in the study area. The monthly precipitation $\delta^{18}O$ enrichment rate in the Qilian Mountains of China from May to October was 29.18%, 23.35%, 25.60%, 22.99%, 31.64%, and 14.72%, respectively. For every 1.00% increase in the evaporation rate of raindrops in Qilian Mountains of China, the changes in the concentration of oxygen isotopes from the bottom of the clouds to the ground increased by 0.92‰; however, with an evaporation rate of < 5.00%, for every 1.00% increase in the evaporation rate of raindrops the changes in the concentration of oxygen isotopes from the bottom of the clouds to the ground increased by 1.00 ‰ could also be observed. Furthermore, altitude was an important factor affecting below-cloud evaporation in the study area.

Keywords: below-cloud evaporation; stable isotopes; transition zone; Qilian Mountains of China

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1 Introduction

Precipitation is an important part of the earth's water circulation system (Aggarwal et al., 2016; Wang et al., 2016; Li, et al., 2016a). Stable isotopes of hydrogen and oxygen (^2H and ^{18}O) are the main constituents of water

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molecules (Gat, 1996; Jouzel et al., 2000), as they preserve information on fractionation, condensation, and exchange processes along the air mass pathway from a source region to the precipitation site (Dansgaard, 1964; Li et al., 2015; Shi et al., 2021). However, for arid areas, the isotopic composition of precipitation is susceptible to environmental factors (Craig, 1961; Li, et al., 2016b; Sun et al., 2019; Ansari et al., 2020). For example (Li et al., 2016c), below-cloud evaporation can significantly alter the stable isotopes of precipitation, making the recorded information inconsistent with reality. Below-cloud evaporation is the evaporation of raindrops under unsaturated water vapor pressure during precipitation from cloud-base to the ground, resulting in $\delta^{18}O$ enrichment (Wang et al. 2016). Therefore, it is necessary to correct the measured isotopic composition of precipitation under below-cloud evaporation, which could alter the ratios of stable hydrogen and oxygen isotopes in falling raindrops (Xiong et al. 2021). Accurate description of the underlying processes affecting stable isotopic composition could improve our understanding of the water cycle. If the influence of below-cloud evaporation on hydrogen and oxygen isotopes is understood, these changes can be used to obtain additional accurate information from the water cycle, which would be valuable to the research field. In general, heavy isotopes in precipitation are enriched, while the deuterium surplus is reduced due to secondary evaporation under clouds. During atmospheric precipitation, below-cloud evaporation occurs to varying degrees as raindrops fall from the cloud-base to the ground (Wang et al., 2016). Below-cloud evaporation is an important factor affecting isotopic variation in precipitation after condensation formation (Stewart., 1975). Moreover, it significantly impacts local small-scale rainfall events (Peng et al., 2007). Therefore, the evolution mechanism of stable isotopes, especially that of below-cloud evaporation of precipitation in arid and semi-arid areas, has become a core scientific issue in isotope hydrology (Li et al., 2019). In general, the drier the climate, the higher is the evaporation. Additionally, geomorphic features significantly influence below-cloud evaporation in plain areas, which is usually higher than that in mountainous areas (Froehlich et al., 2008). Many factors affect below-cloud evaporation (Ma et al., 2014), among which meteorological factors, such as temperature and relative humidity, play particularly important roles.

Remarkable progress has been achieved in research that considers below-cloud evaporation in some humid and sub-humid areas of China. It is mainly reflected in the following aspects: 1) The important effect of evaporation under cloud on precipitation was found. Below-cloud evaporation or recycled moisture controlled the spatial pattern during the wet season in the Chenghai Lake Basin, Southwest China (Wang et al. 2021). Wu et al., (2015) observed that below-cloud evaporation reduced the slope and intercept of the local meteoric water line (LMWL). Che et al., (2019) discussed the sub-cloud evaporation of precipitation isotopes in the Yellow River Basin of China. Xiao et al., (2020) observed that the hourly below-cloud evaporation was higher during daytime than at night; moreover, different regions had varying characteristics on a monthly scale in the Yangtze River Basin of China. 2) Below-cloud evaporation rates were calculated for some regions. Wen et al. (2017) concluded that below-cloud evaporation of raindrops can not occur at temperature < 7°C. Jin et al. (2015) calculated the degree of below-cloud evaporation in the Loess Plateau of China using the simplified formula of (Peng et al. 2010) and reported the possible existence of some errors in this backward calculation method. Zhou et al. (2019) studied the below-cloud evaporation of summer precipitation isotopes in Gansu of China based on an improved Stewart model (stratified hypothesis). Using a modified Stewart model, Jiao et al., (2019) simulated the variation for excess deuterium change (Δd) of a cloud-base and ground raindrops in Shanxi of China. It is also found that the annual average Δd in the northern Shaanxi and Guanzhong regions of China showed a linear decreasing trend over the previous 57 yr (Xiao et al., 2020). It can be found that below-cloud evaporation was considerable in summer and autumn in the northern subtropical and southern temperate regions, and in spring and summer in the central subtropical and plateau climate regions. Owing to the differences in meteorological conditions across regions, the linear relationship between δD and below-cloud evaporation rate (f) in precipitation varies between different meteorological elements (Che et al., 2019). According to the above analysis, significant progress has been achieved in this research field for some arid and semi-arid areas of China, but few studies have been conducted in arid or semi-arid regions. Arid and semi-arid regions may face more severe evaporation under clouds and thus deserve more attention.

The Qilian Mountains of China are located in the northeastern edge of Tibetan Plateau, with arid and semi-arid areas larger and serve as the transition zone between the Tibetan Plateau and arid region of China (Gui et al., 2020). Due to the complex terrain and hydrothermal conditions, the Qilian Mountains have a continental climate and precipitation is an important water resource in this region. Below-cloud evaporation affects falling hydrometeors during their descent from the cloud-base toward the ground, thereby modifying the size of the raindrops reaching the ground. Accurate description of the underlying processes affecting stable isotopic composition could improve our understanding of the water cycle. However, the changes in below-cloud evaporation of falling raindrops in unsaturated air in Qilian Mountains remain unclear. This study aimed to: 1) identify signs of evaporation under clouds in Qilian Mountains of China, 2) assess the variation of *d*-excess and $\delta^{18}O$ in precipitation from the cloud-base to the ground in Qilian Mountains of China, and 3) evaluate the influence of below-cloud evaporation on precipitation and its meteorological controls in Qilian Mountains of China.

2 Materials and Methods

2.1 Study area

The Qilian Mountains (36°N–43°N, 92°E–107°E) span 18.1×10^4 km^2; natural conditions of the region are complex, and hydrothermal conditions vary significantly. The region has a typical continental climate, with high humidity and precipitation in the eastern part and dry climate and low precipitation in the western part (Fig. 1). The eastern area is influenced by the southeast and southwest monsoons, while the western area is controlled by westerly circulation. The winter months are mainly controlled by the Mongolian high-pressure system; the weather is cold and dry, and the summer months are influenced by the southwest monsoon from the Indian Ocean and the southeast monsoon from the Pacific Ocean. Annual precipitation ranges from 400 to 700 mm, with more than half of the precipitation concentrated during May and August, and the precipitation decreasing from the southeast to the northwest. The ecosystem changes vertically from low to high altitudes, from dry shrubbery grassland to forest grassland, subalpine shrubbery meadows, alpine cold-desert meadows, alpine permafrost, and glaciers, respectively. The Qilian Mountains are the source and runoff formation areas of the inland rivers (Wu et al. 2015). Almost all major rivers in the region originate from mountain glaciers and depend on glacier recharge. The proportion of meltwater recharge increases from the east to the west. The mountain rivers are frozen annually from October to April. weak spring floods occur during April to May. The main flood season occurs during July–August because of the increase in precipitation and glacier melting.

2.2 Sample collection and analysis

Nineteen precipitation sample collection sites at different elevations and different slope direction in the Qilian Mountains of China were used, with a total of 1310 groups of samples collected (Fig. 1, Table 1). Sampling was done manually on an event basis using a wet col-

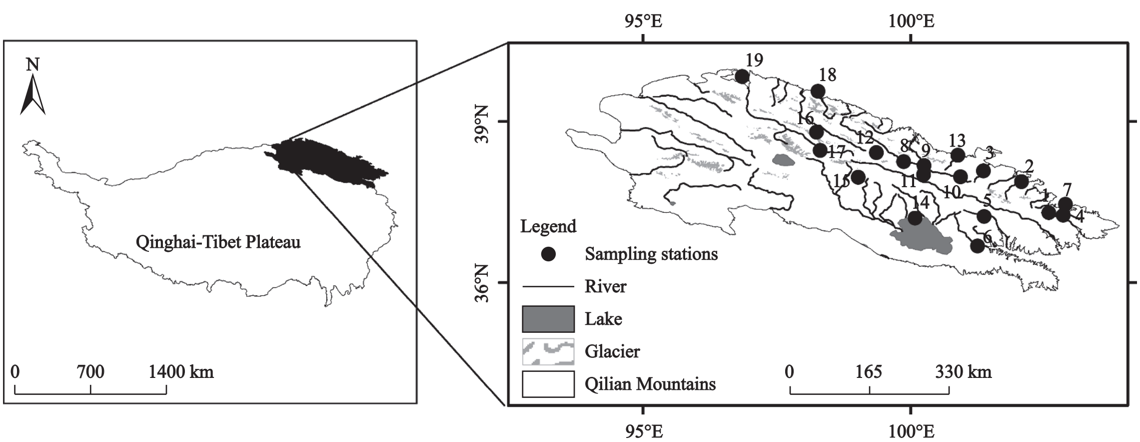

Fig. 1 Location of the Qilian mountains of China and sampling points

Table 1 Information of 19 sampling points in Qilian Mountains of China

No.	Sampling sites	Latitude / °N	Longitude / °E	Altitude / m	Samples number
1	Daiqian	37.30	102.58	3272	89
2	Jiutiaoling	37.88	102.07	2225	55
3	Xidahe	38.08	101.36	2897	57
4	Anyuan	37.25	102.85	2687	160
5	Mengyuan	37.23	101.37	2850	81
6	Huangyuan	36.68	101.25	2675	55
7	Gulang	37.46	102.89	2105	61
8	Hulugou	38.25	99.87	3260	56
9	Binggou	38.01	100.24	4146	143
10	Ebao	37.97	100.93	3452	95
11	Qilian	38.18	100.25	2787	65
12	Yeniugou	38.42	99.36	3320	79
13	Minle	38.37	100.88	2459	43
14	Gangcha	37.2	100.08	3301.5	66
15	Tianjun	37.96	99.02	3417.1	68
16	Tuole	38.80	98.24	3367	103
17	Suli	38.46	98.31	3841	35
18	Jiayuguan	39.80	98.27	1658	24
19	Changma	39.83	96.85	2112	30

lector, which consisted of a 5-L polyethylene collecting flask (at the bottom) fitted with a polyethylene bottle (26 cm diameter). Meteorological parameters such as precipitation, wind speed, and humidity were recorded simultaneously during sample collection. At the same time, all sampling points were divided into north and south slopes and east, middle and west sections of the Qilian Mountains according to reference Gui et al., (2020).

The analysis of stable isotopes of hydrogen (δD) and oxygen (δ^{18}O) in precipitation was conducted at the Key Laboratory of Eco-hydrology of the Inland River Basin at the Chinese Academy of Sciences. The stable isotopes in the precipitation samples were measured using a liquid water stable isotope analyzer (Model DLT-100, Los Gatos Research, Inc., Mountain View, CA, USA). The results were calibrated using the Vienna Standard Mean Ocean Water (V-SMOW) and laboratory working standards. The final results were expressed in the form of micro-difference relative to V-SMOW as follows:

$$\delta^{18}\mathrm{O}(\delta\mathrm{D}) = (R_{\mathrm{Sample}}/R_{\mathrm{V\text{-}SMOW}} - 1) \times 1000‰ \quad (1)$$

where R_{Sample} is the precipitation sample and $R_{\mathrm{V\text{-}SMOW}}$ is the ratio of oxygen or hydrogen stable isotope in V-SMOW.

d-excess is equivalent to the intercept when the slope of the regional meteoric water line in this region is 8, indicating the degree of imbalance in the evaporation process. It can be calculated as follows:

$$d\text{-excess} = \delta\mathrm{D} - 8\delta^{18}\mathrm{O} \quad (2)$$

2.3 Methods

2.3.1 Calculation on the stable isotopes of oxygen (δ^{18}O) *in precipitation of the cloud base*

Below-cloud evaporation increases δ^{18}O of precipitation, whereas moisture recycling decreases the value of this parameter (Pang et al., 2011; Kong et al., 2013). Therefore, it is necessary to correct the measured δ^{18}O in precipitation under the effect of sub-cloud evaporation. δ^{18}O of precipitation at the cloud-base can be determined reference to the Froehlich et al., (2008). The evaporation rate (*f*) of falling raindrops can be calculated reference to the Kinzer and Gunn (1951).

2.3.2 Calculation on the enrichment rate of δ^{18}O

The relative effect of below-cloud evaporation on the composition of δ^{18}O can be defined as the below-cloud evaporation enrichment rate of $\delta^{18}O$ (E), which can be estimated by using the following formula:

$$E = (^{18}F - \delta^{18}O)/\delta^{18}O \quad (3)$$

where ^{18}F is the composition of $\delta^{18}O$ of precipitation at the cloud-base, and $\delta^{18}O$ is the stable oxygen composition in precipitation of the ground sampling stations.

2.3.3 Calculation of the changes in oxygen isotope ratio and d-excess

The changes in oxygen isotope ratio and *d*-excess of the raindrops from the cloud-base to the ground (values at the ground minus values at the cloud-base) can be expressed as:

$$\Delta\delta^{18}O = \delta^{18}O_{gr} - \delta^{18}O_{cb} \quad (4)$$

$$\Delta d = d_{gr} - d_{cb} \quad (5)$$

where $\delta^{18}O_{gr}$ and $\delta^{18}O_{cb}$ are $\delta^{18}O$ values of a raindrop near the ground and below the cloud-base, respectively; d_{gr} and d_{cb} are d values of a raindrop near the ground and below the cloud-base, respectively.

3 Results

3.1 The signal of below-cloud evaporation

As rain falls from the cloud-base to the ground, it is strongly affected by below-cloud evaporation, which intensifies the dynamic fractionation of stable isotopes. Consequently, the slope and intercept of local meteoric water line (LMWL) decrease significantly (Peng et al., 2004; Peng et al., 2007). During precipitation, temperature and relative humidity primarily affect the influence of below-cloud evaporation on precipitation stable isotopes (Liu et al., 2008). As shown in Fig. 2a, the $\delta^{18}O$ values of precipitation (P) gradually increased with increasing temperature (T), while the slope and intercept of the LMWL showed a decreasing trend. The above facts indicate that at a precipitation temperature above 0°C (T > 0°C), below-cloud evaporation significantly affects the stable isotopes of precipitation, continuously enriching them. However, this effect weakens significantly at high precipitation with increasing temperature (Fig. 2b). For northwestern China, numerous studies have shown that below-cloud evaporation significantly affects stable isotopes at low precipitation; however, the influence is weak for snowfall or heavy precipitation (Lee et al., 2012).

For $\delta^{18}O$, at a precipitation of 0–1 mm, with increasing temperature, showing an overall increase of 5.32‰. However, when P >10 mm, $\delta^{18}O$ changed from −8.29‰ to −6.70 ‰, and ultimately reached 1.59 ‰ (Fig. 2b). With increasing temperature, the enrichment was only 1.59‰. Evaporation does not significantly affect snowfall or precipitation events below 0°C. For events with higher or continuous precipitation, the saturated atmospheric water vapor pressure is relatively higher; hence, the relative humidity increases, eventually causing the evaporation intensity to gradually weaken. Consequently, $\delta^{18}O$ is less enriched. As shown in Fig. 2b, when precipitation is > 5 mm, the $\delta^{18}O$ decreased significantly. These facts indicate that $\delta^{18}O$ is enriched with increasing temperature regardless of the precipitation range.

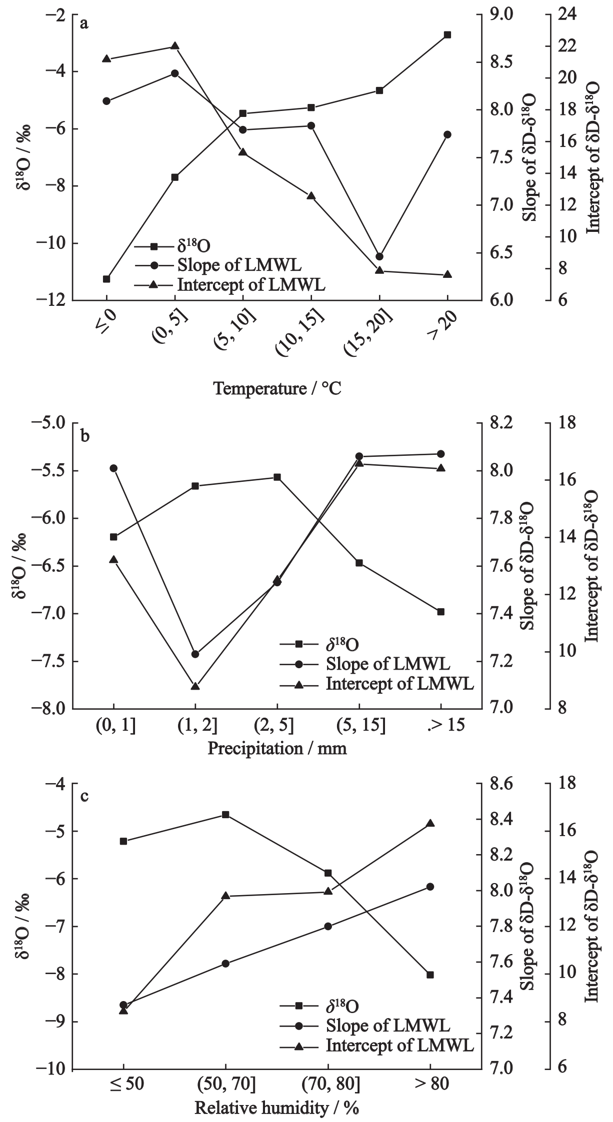

Fig. 2 Variation of $\delta^{18}O$ and local meteoric water line (LMWL) under different (a) temperature and (b) precipitation and (c) relative humidity in the Qilian Mountains of China

Stable isotopes in precipitation under low precipitation events are affected more by evaporation, compared with stable isotopes under high precipitation events. This mainly occurs because isotopic enrichment varies linearly with the evaporated mass fraction of the raindrop, resulting in higher values in the drier atmospheres and smaller raindrops.

The influence of relative humidity (H) on stable isotopes of precipitation was most noticeable during the transport of water vapor from the source to the study area (Gui et al.,2020). Stable isotope equilibrium fractionation occurred at a relative humidity of 100%; moreover, its value followed the Rayleigh fractionation law (Craig and Gordon, 1965). However, fluctuations in sea surface temperature at the water vapor source could lead to changes in relative humidity, wind speed, and initial evaporation, ultimately causing fluctuations in stable isotope concentrations (Bajjali, et al., 1997). Generally, in arid zones, owing to the lower relative humidity, the energy imbalance caused by evaporation and perturbations in molecular dynamics under non-equilibrium clouds during the phase change of the water column leads to a significant enrichment of $\delta^{18}O$ in the remaining precipitation. During the study period, $\delta^{18}O$ increased from −8.02‰ ($80\% < H$) to −5.22‰ ($H \leq 50\%$) with decreasing relative humidity (Fig. 2c), while the slope and intercept of the δD-$\delta^{18}O$ linear relationship were significantly reduced: the slope decreased from 8.02 to 7.36, and the intercept decreased from 16.29 to 8.42. The effect of evaporation under clouds on the stable isotopes in precipitation was demonstrated to be smaller at high relative humidity.

3.2 Spatio-temporal variation of below-cloud evaporation

The above analysis confirmed that the effect of below-cloud evaporation on the stable isotopes in precipitation is significant. Therefore, the below-cloud evaporation rate of the landing raindrops (also known as the amount of water that evaporates as rain drops from the bottom of the cloud to the ground) and the values are expressed as percentages. According to Kong et al., (2013), the average radius of raindrops in the Qilian Mountains was 0.4 mm. Based on our calculation, the mean f in the study area was 12.00% during the summer half-year (May-October). The corresponding average monthly f values from May to October were 15.01%, 10.25%, 12.59%, 12.71%, 18.57%, and 11.80%, respectively (Fig. 3). The f value was relatively higher in May and September than in other months, mainly because of the rapid rise in temperature and the low precipitation in May, contributing to a higher evaporation rate. Compared with July and August, precipitation was lighter in September at high temperatures, resulting in high evaporation rates of raindrops. A low value of f mainly occurred in July and August, primarily because precipitation in the study area was concentrated during this period. As mentioned above, below-cloud evaporation had a

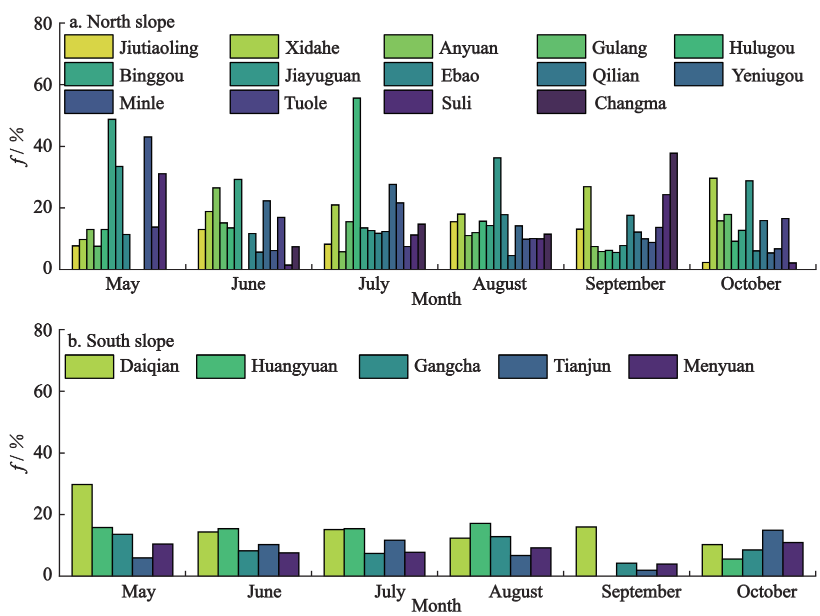

Fig. 3 Evaporation rate (f) varies with time on the (a) northern slope and (b) southern slope in the Qilian Mountains of China

greater impact on low precipitation events. Although the temperatures in July and August were higher, the high precipitation weakened the intensity of below-cloud evaporation to a certain extent.

The evaporation rate on the northern slopes (12.65%) of the Qilian Mountains was significantly greater than that on the southern slopes (9.98%) (Fig. 3). The spatial distribution of evaporation rates for the raindrops described above occurred due to three main reasons. 1) The climate on the northern slopes of the Qilian Mountains is relatively dry with little precipitation, resulting in strong evaporation. 2) The southern slope is influenced by monsoonal circulation in the summer, while the precipitation is relatively high. 3) The five stations on the southern slope had a higher average elevation and relatively lower temperatures. This also enables raindrops to evaporate over longer periods. In general, precipitation could be faced more raindrop evaporation in the plains than in the mountains, because the distance between the cloud-base and the sampling site was greater in the plains. As shown in Fig. 4, the evaporation rate of raindrops decreased gradually with increasing altitude. For every 100 m elevation, f decreased by 0.60%. This also explains the lower below-cloud evaporation rates at higher altitudes under similar conditions.

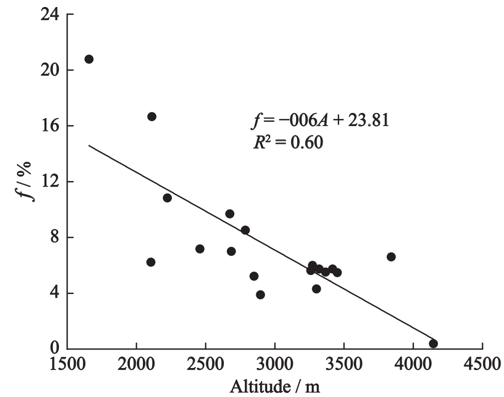

Fig. 4 Variation of evaporation rate (f) with altitude (A) in the Qilian Mountains of China

Below-cloud evaporation rate also varied greatly from month to month. As shown in Fig. 5, overall, f shows an increasing trend from the west to east and a decreasing trend from the north to south in the western section of the study area. It increased from the west to the east in June; however, the variation was smaller. In July, the Northwest corner of Qilian Mountains was taken as the low-value center, and f gradually increased from the central to the outer parts. In August, f increased from the south to north in general, and a lower value was observed in southeast corner of Qilian Mountains. Furthermore, a minimum value of f was observed for southeast corner of Qilian Mountains in September and October (Fig. 5). These spatial patterns reflect the differences in climatic conditions in the Qilian Mountains, thereby confirming that factors such as altitude, topography, and the water vapor source affect below-cloud evaporation.

3.3 Variation of $\delta^{18}O$ in precipitation from the ground to cloud

Fig. 6 shows that $\delta^{18}O$ in raindrops is evidently enriched due to the influence of below-cloud evaporation while raindrops fell from the cloud-base. Considering the measured $\delta^{18}O$ in precipitation, the results showed that the estimated $\delta^{18}O$ at the cloud-base was higher than that at ground level from May to October. The magnitude of the difference between the $\delta^{18}O$ value in the surface precipitation and that in the water vapor at the bottom of the cloud helps to determine the evaporation intensity of the raindrops below the cloud; a high magnitude indicates that the degree of evaporation enrichment of stable isotopes in raindrops is high, and vice versa. The monthly $\delta^{18}O$ values of precipitation at the cloud-base were –8.60‰, –6.30‰, –6.70‰, –7.30‰, –9.00‰, and –13.00‰ from May to October, respectively. The corresponding values considering ground precipitation were –6.00‰, –4.90‰, –5.10‰, –5.70‰, –6.40‰, and –10.90‰, respectively (Fig. 6). The monthly mean $\delta^{18}O$ variation between the cloud-base and the ground level ($\Delta\delta^{18}O = \delta^{18}O_{gr} - \delta^{18}O_{cb}$) demonstrated clear characteristics of time variation. The monthly mean $\Delta\delta^{18}O$ was 2.60‰, 1.37‰, 1.59‰, 1.53‰, 2.74 and 2.01‰. The greater the value of 1, the stronger the evaporation of precipitation from the cloud base to the ground. The change of $\Delta\delta^{18}O$ is also consistent with the change trend of f, that is, it shows a higher trend in May and September, and a lower trend in June. Confirmed once again that evaporation intensity of raindrops under clouds was higher in May and September, and lower in June.

A good linear relationship existed between f and $\Delta\delta^{18}O$, with a slope ($\Delta\delta^{18}O/f$) of ~ 0.12 for the Qilian Mountains (Fig. 7). Assuming that below-cloud evaporation does not occur (and that $\Delta\delta^{18}O = 0$) under conditions of low air temperature, high relative humidity,

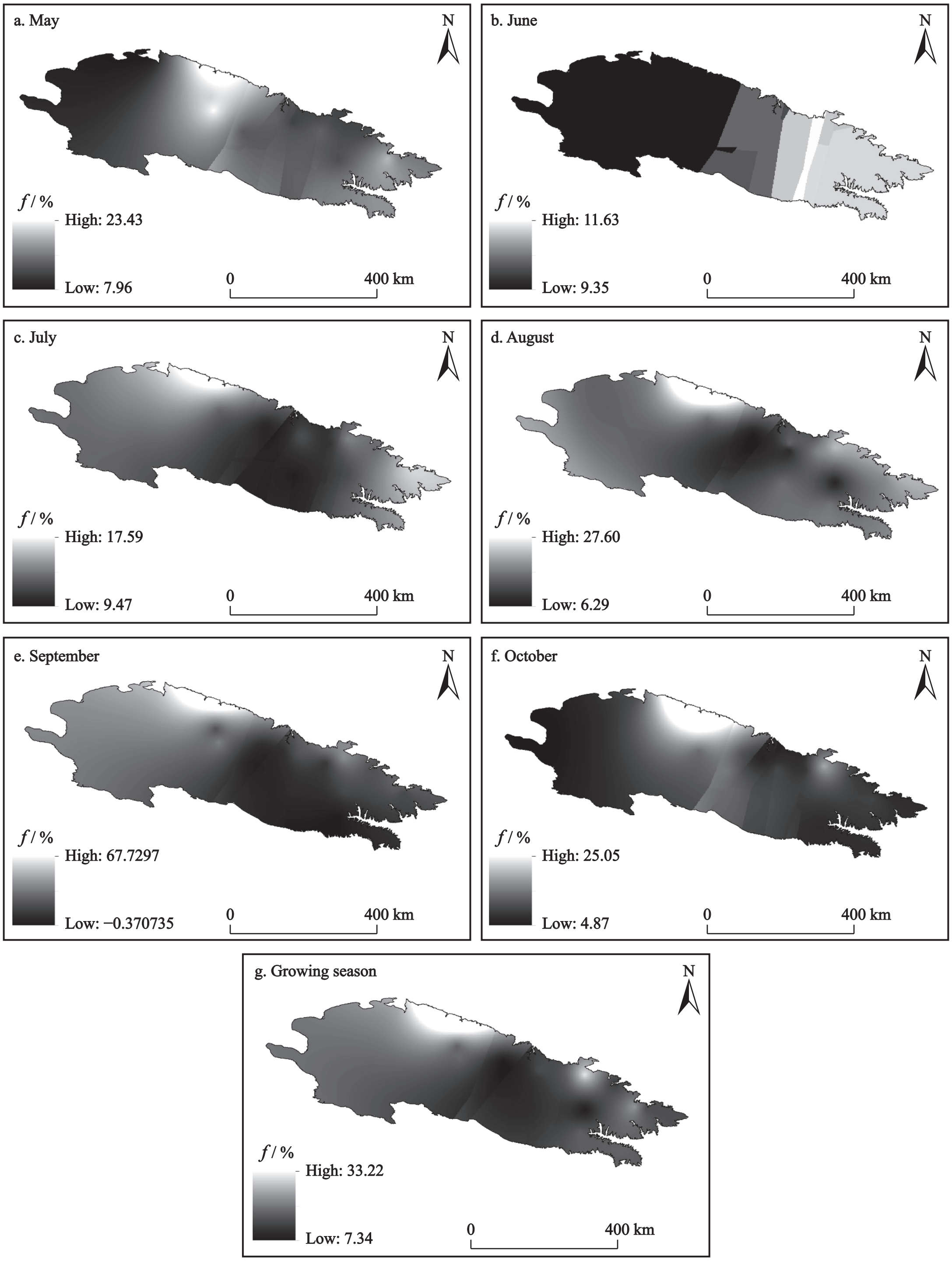

Fig. 5 Spatial variation of evaporation rate (*f*) in different months in the Qilian Mountains of China

high precipitation, and a large raindrop diameter, this linear relationship no longer exists. In fact, due to different altitudes and climatic conditions at different stations in the study area, the stable isotopes in precipitation are subject to and influenced by varying degrees of below-cloud evaporation; a linear correlation between f and $\Delta\delta^{18}O$ was observed. Slopes of 0.12, 0.12, and 0.14 were observed for the eastern, central, and western parts of the Qilian Mountains of China, respectively.

Although the $\delta^{18}O$ in precipitation observed on the ground differed from the corresponding value estimated under the cloud owing to the influence of below-cloud evaporation, a certain relationship still existed between them under specific evaporation and climatic conditions.

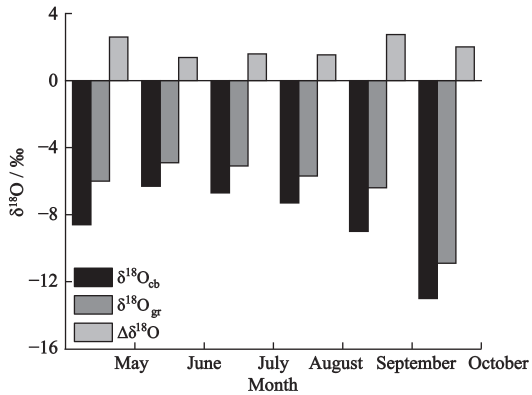

Fig. 6 The $\delta^{18}O$ values of cloud base ($\delta^{18}O_{cb}$), ground ($\delta^{18}O_{gr}$) and variation ($\Delta\delta^{18}O$) in the Qilian Mountains of China

If this relationship could be recognized, it is important to correct the measured isotopic composition in precipitation affected by below-cloud evaporation. This would facilitate a better understanding of the hydrological processes involved. The correlation coefficients between these parameters were 0.85, 0.86, and 0.74 for the eastern, central, and western parts of the Qilian Mountains, respectively (Fig. 8). The correlation coefficient was consistent with the spatial distribution characteristics of the evaporation rate *f*; it was highest in the central section, followed by the eastern and the western section. These facts again prove that below-cloud evaporation strongly influenced the stable isotopes in precipitation in the study area. The intensity of this influence varied spatially due to the differences in terrain and climatic conditions.

The influence of below-cloud evaporation on $\delta^{18}O$ was further evaluated and defined as the enrichment rate (*E*) of $\delta^{18}O$. The results show that the monthly average $\delta^{18}O$ values of precipitation at the cloud-base in the study area from May to October were –8.34‰, –6.36‰, –6.33‰, –6.60‰, –8.65‰, and –12.47‰, respectively. After the influence of below-cloud evaporation, the corresponding surface precipitation values were –6.32 ‰, –4.97‰, –4.90‰, –0.53‰, –6.16‰, and –10.74‰, respectively. $\delta^{18}O$ of precipitation on the ground was more positive than that at the cloud-base, which proved the strong influence of below-cloud evaporation on $\delta^{18}O$ in precipitation. The monthly precipitation $\delta^{18}O$ enrichment rate in the Qilian Mountains from May to October was 29.18%, 23.35%, 25.60%, 22.99%, 31.64%, and 14.72%, respectively (Fig. 9). In terms of temporal variation, the *E* of precipitation was similar to the evaporation rate of raindrops, i.e., it was highest in May and September. However, it varied insignificantly from June

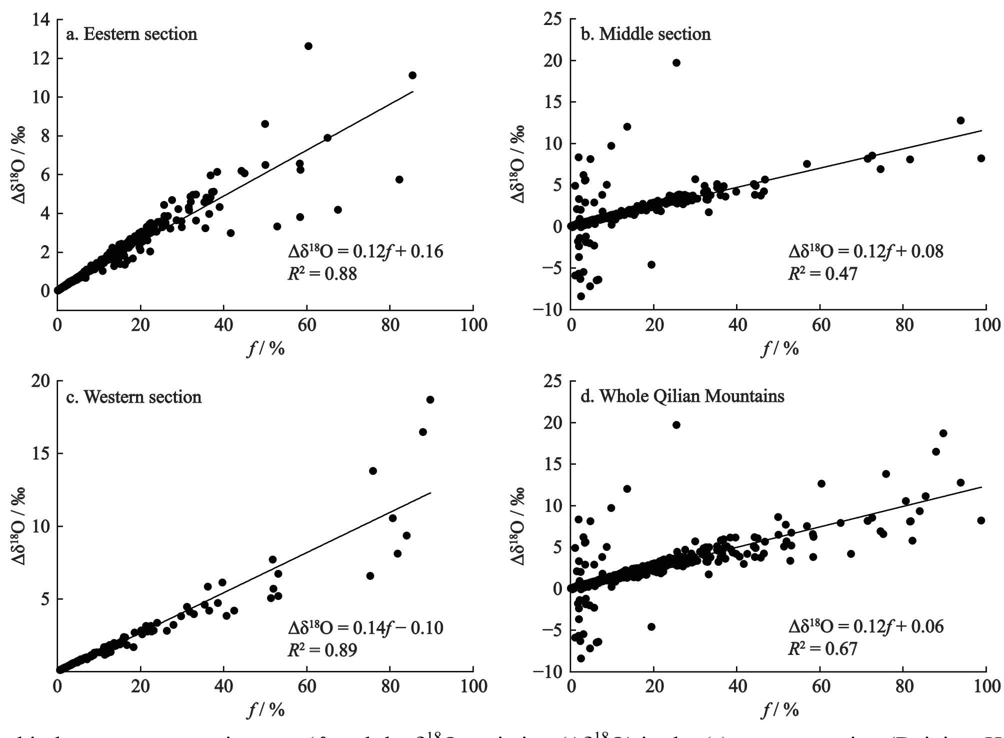

Fig. 7 Relationship between evaporation rate (*f*) and the $\delta^{18}O$ variation ($\Delta\delta^{18}O$) in the (a) eastern section (Daiqian, Huangyuan, Anyuan, Menyuan, Gulang, Jiutiaoling, Xidahe), (b) middle section (Hulugou, Binggou, Ebao, Qilian, Yeniugou, Gangcha, Tianjun, Minle), (c) western section (Changma, Jiayuguan, Suli, Tuole) and (d) whole of the Qilian Mountains of China

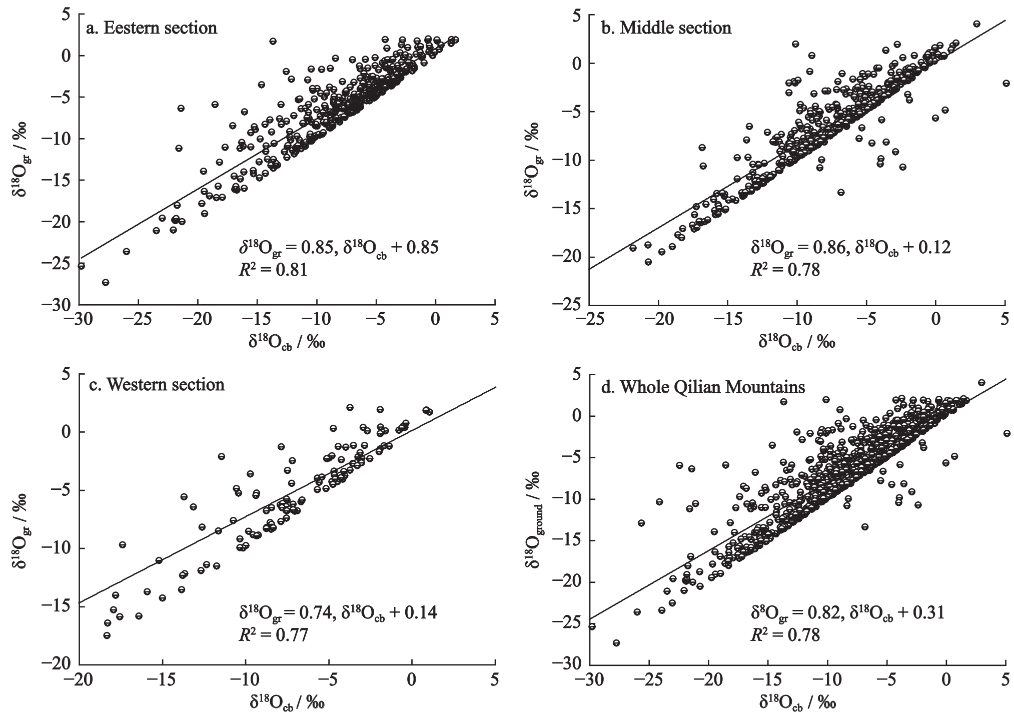

Fig. 8 Variation of $\delta^{18}O$ in precipitation from the ground ($\delta^{18}O_{gr}$) to cloud base ($\delta^{18}O_{cb}$) in the Qilian Mountains of China

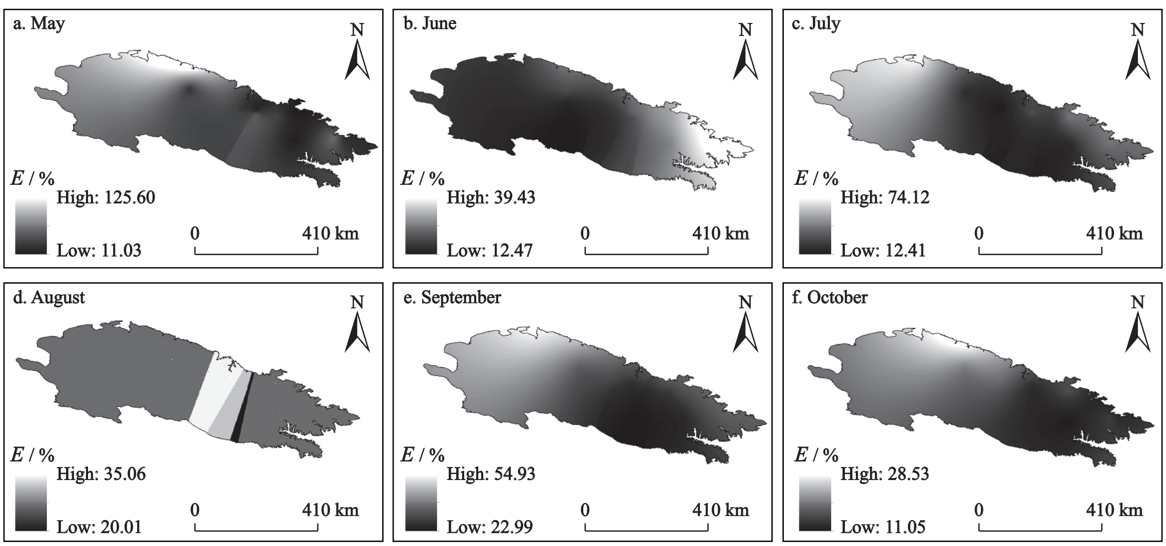

Fig. 9 Spatial distribution of $\delta^{18}O$ enrichment rate (E) in the Qilian Mountains of China

to August. Spatially, E increased gradually from the east to the west in May. E increased gradually from the central region to the outer region from the inside to the outside in June. In July, the central and southeastern parts of the Qilian Mountains showed low E values, while the maximum value was observed in the western part. In August, the spatial variation in E was small, and gradually decreased from the west to the east. The value of E gradually increased from the southeast to the northwest in September and October. This indicated that the evolution of stable isotopes in precipitation in the Qilian Mountains of China was influenced by below-cloud evaporation. As shown in the previous analysis, below-cloud evaporation rate of falling raindrops and the E value of $\delta^{18}O$ demonstrated similar trends in spatiotemporal variation. Further analysis showed that f and E were positively correlated, indicating that below-cloud evaporation significantly influenced the concentration of stable isotopes in precipitation.

4 Discussion

4.1 Evaporation rate and changes of *d*-excess

The stable isotope ratios of the precipitation collected near the ground varied considerably from those of the precipitation at the cloud-base. It was assumed that the isotopes in precipitation at the cloud-base were in equilibrium with the surrounding water vapor isotopes. The change in *d*-excess in raindrops between the cloud base and the ground could be expressed as Δd (Stewart, 1975; Froehlich et al., 2008). The more negative the value is, the stronger the secondary evaporation effect is. From May to October, the mean Δd was –17.00‰, –10.80‰, –12.50‰, –12.10‰, –20.40‰, and –14.00‰, respectively (Fig. 10). This was consistent with the change in the evaporation rate *f*. In terms of the monthly average values, the maximum Δd was observed in May and September. At a single site, the Jiayuguan station on the north slope, had the smallest Δd. In September, Δd was as low as –117.30‰. This is closely related to Jiayuguan station's low altitude, relative humidity, and precipitation as well as high temperature. In contrast, some stations on the southern slope, such as Menyuan, had lower average Δd value of –7.40‰.

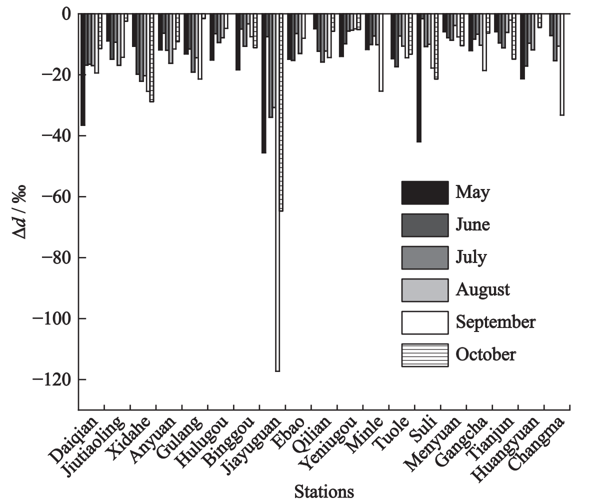

Fig. 10 The variation of *d*-excess from cloud base to ground (Δd) at different stations in the Qilian Mountains of China

Fig. 11 shows the spatial variation in Δd from May to October; Δd gradually increased from the northwest to the southeast, especially in May, September, and October, showing evident variation in spatial distribution. During these three months, Δd values were generally lower, especially in the western section of the study area, which was related to the intense evaporation during the study time. The spatial variation in Δd was not significant from June to August, gradually increasing from the central part of the Qilian Mountains to the eastern and western parts in June and July, respectively. In August, the variation in Δd was smaller, and increased from the west to the east. In September and October, Δd increased from the southeast to the northwest (Fig. 11).

Previous studies (Froehlich et al., 2008; Kong et al., 2013; Crawford et al., 2017) have shown that in some relatively humid areas with a low evaporation rate, the slope of the linear relationship between *f* and Δd was seen to always be close to 1 under low *f* conditions. Generally, in an arid environment with a high air tem-

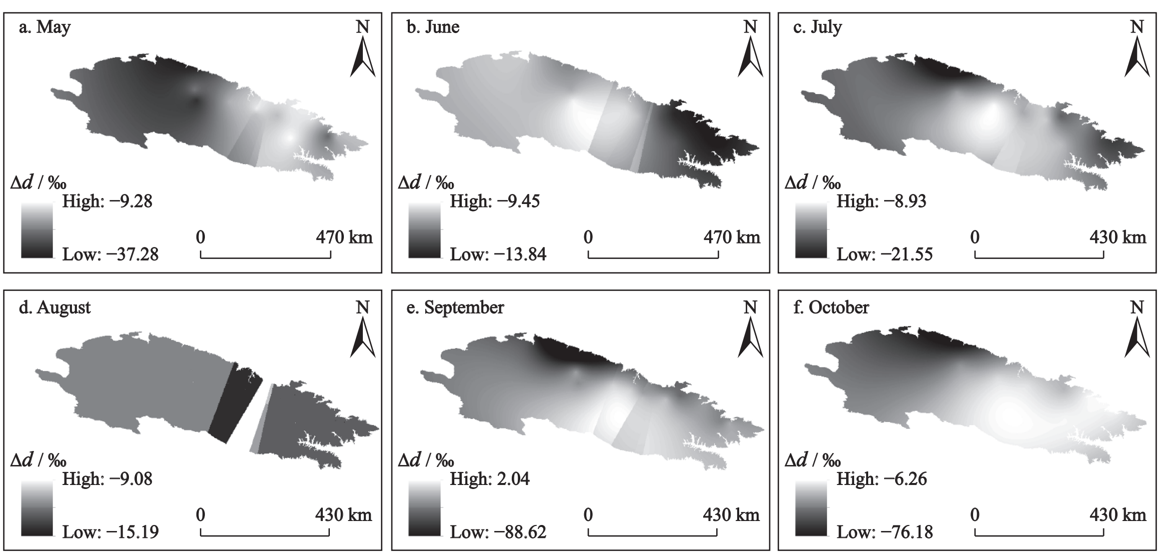

Fig. 11 Spatial variation of variation of *d*-excess from cloud base to ground (Δd) in the Qilian Mountains of China

perature, low relative humidity, low precipitation, and small raindrop diameters, the evaporation fraction is relatively large; moreover, the difference in Δd is also large. This could almost lead to total evaporation for small raindrops (Salamalikis, et al., 2016), with the slope deviating from 1. So, the slope of the linear relationship was not equal to 1 in every region. For example, Xiao et al., (2020) found that the slope of the linear relationship between *f* and Δd for the Shaanxi-Gansu-Ningxia provinces (regions) was < 1.00%. A similar linear relationship between the evaporation fraction and Δd was reported for Linfen (Sun et al. 2020). It was observed that Δd in the Qilian Mountains had a significant negative correlation with evaporation rate *f*. Furthermore, Fig. 12 demonstrate the correlation between these two parameters. In the summer half-year, for every 1% increase in the evaporation rate of raindrops in the Qilian Mountains, Δd decreased by 0.92‰. The north-south slope of the southern slope (1.02) was significantly higher than that of the northern slope (0.92) (Fig. 12). As evident from Fig. 12, the relationship between *f* and Δd gradually weakened as the evaporation rate increased. Moreover, as the evaporation rate increased to 20%, the scattering around the regression line strengthened. However, a linear relationship (~1.00 ‰ per 1.00%) was also observed for evaporation rates < 5.00%. It could be seen that at this time, regardless of the entire Qilian Mountains of China or their northern and southern slopes, the slope was approximately equal to 1.

4.2 Factors influencing below-cloud evaporation

Changes in the concentration of stable isotopes in precipitation are mainly influenced by fractionation processes, which are also mainly influenced by phase transition processes, such as evaporation and condensation, controlled by temperature and humidity (Li et al. 2015). Considering that climatic conditions vary greatly at different altitudes, all precipitation events were divided into four groups based on the altitude of the sampling sites to analyze the effects of temperature and relative humidity on enrichment rates. The results showed that temperature and $\delta^{18}O$ enrichment had a significant positive correlation; the correlation coefficient was the highest in the region with altitude of 3000–4000 m, followed by the region with altitude of < 2000 m (Table2). The correlation coefficient between the isotope enrichment rate

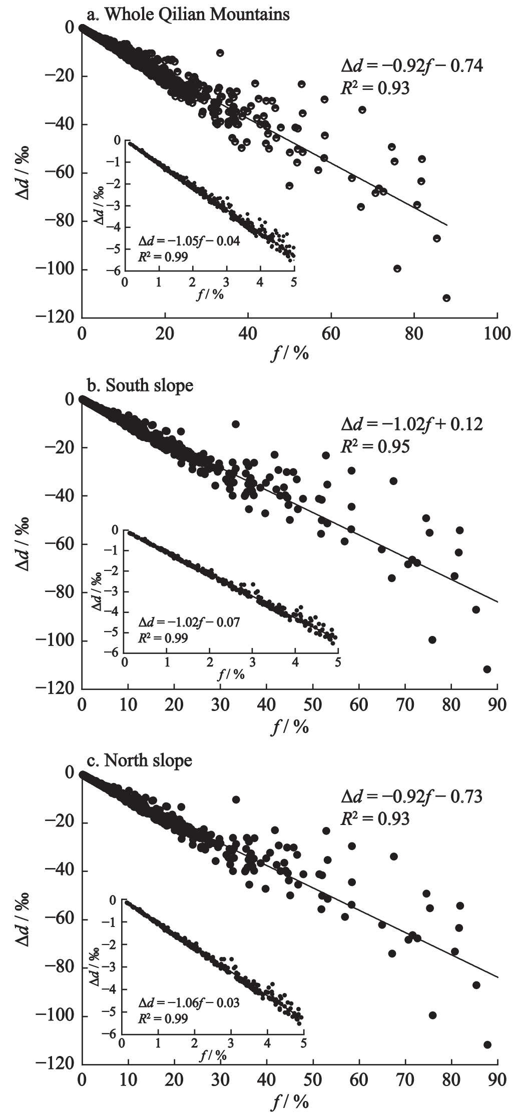

Fig. 12 Relationship between evaporation rate (*f*) and precipitation Δd in the (a) whole, (b) south slope, and (c) north sloppe of the Qilian Mountains of China

and air temperature was low in the region with altitude of 2000–3000 m and above 4000 m. The possible causes are as follows. 1) The high temperature in the region at an altitude < 2000 m could have caused intense subcloud evaporation. 2) The forest is concentrated in the Qilian Mountains at an altitude of 2000–3000 m, where the evapotranspiration process is intense. This is the zone with the highest precipitation in the mountainous area, indicating that the stable isotope enrichment caused by below-cloud evaporation was offset by the depletion caused by the LMWL. 3) The region at an altitude of 3000–4000 m is located in the transition region between the height zone with the second highest precipitation and the height zone with the highest precipita-

Table 2 The correlation between enrichment rate and temperature, precipitation and relative humidity at different altitude in the Qilian Mountains of China

Altitude / m	Temperature	Precipitation	Relative humidity
< 2000	**0.37**	**–0.61**	**–0.57**
2000–3000	**0.27**	**–0.44**	**–0.58**
3000–4000	**0.88**	**–0.41**	**–0.39**
> 4000	0.33	–0.28	**–0.75**

Note: boldface means $P < 0.05$

tion in the mountainous area, where the precipitation is low and the influence of secondary evaporation is relatively large. 4) The area above 4000 m is the second precipitation height zone, which has high precipitation and is less affected by secondary evaporation.

The enrichment rates of $\delta^{18}O$ and precipitation showed a significant negative correlation; moreover, the correlation coefficient decreased with elevation (Table 2). This indicated that in high-altitude areas, temperature had the greatest influence on secondary evaporation, whereas in low-altitude areas, with an increase in precipitation, the intensity of sub-cloud evaporation was weakened . The effect of relative humidity on the stable isotope enrichment rate is contrary to that of temperature. The former had the greatest impact on the area at altitudes > 4000 m, followed by the area at altitudes < 3000 m, and had the least impact on the area at the altitude of 3000–4000 m (Table 2).

4.3 Characteristics of below-cloud evaporation in the transition zone

Physical features can be characterized by transitivity in the Qilian Mountains; that is, the transition zone between the Tibetan Plateau and arid region of Northwest China, between monsoonal and arid climates, and between the inland and outland regions of China (Gui et al., 2020). The evaporation rate of raindrops under clouds also indicates that the Qilian Mountains are the transition zone between the Tibetan Plateau and the arid region of Northwest China. During the summer monsoon period (June to September), the evaporation rate of falling raindrops in the Yangtze River was significantly low, at only 4.35% (Ma et al., 2019); thereafter, it increased to 7.35% in the Yellow River Basin (Ma et al., 2019). On the southern and northern slopes of the Qilian Mountains, the corresponding values were 12%. Continuing to the north, in the Hexi Corridor of the arid region in Northwest China, the evaporation rate was 20% (Li et al., 2019). The evaporation rate was also higher in the Kunlun Mountains (18.98%) (Zhang et al., 2019) and Xihexiu, Shaman, Shiquanhe, Jiangka, and Hetian stations at the edge of the Taklimakan Desert, with average f values of 20.50%, 16.60%, 19.50%, 13.40%, and 19.50%, respectively (Sun et al., 2019).

5 Conclusions

The transitivity between monsoonal and arid climates was reflected by the evaporation rate of falling raindrops in precipitation in the Qilian Mountains, a typical transition zone between the Tibetan Plateau and the arid region of Northwest China. Based on 1310 samples of stable isotopes in precipitation, this study assessed below-cloud evaporation and variations in *d*-excess and $\delta^{18}O$ in precipitation from the cloud-base to the ground in the Qilian Mountains. For the summer half-year, the mean f value of the study area was calculated as 12.00%. The evaporation rate on the northern slopes (12.65%) was significantly higher than that on the southern slopes (9.98%), mainly because of evident variations in the temperature, precipitation, altitude, *etc*., between the northern and southern slopes. The stable isotopes in precipitation in the Qilian Mountains were enriched by sub-cloud evaporation. The monthly $\Delta\delta^{18}O$ precipitation enrichment rates in the Qilian Mountains from May to October were 29.18%, 23.35%, 25.60%, 22.99%, 31.64%, and 14.72%, respectively. A good linear relationship existed between f and $\Delta\delta^{18}O$ with a slope ($\Delta\delta^{18}O/f$) of ~ 0.12 in the Qilian Mountains. Slopes of 0.12, 0.12, and 0.14 were observed for the eastern, central, and western parts of the Qilian Mountains, respectively. These values are of great significance in correcting the measured isotopic composition of precipitation affected by sub-cloud evaporation. The Δd for the Qilian Mountains of China demonstrated a significant negative correlation with the raindrop evaporation rate f. In the summer half-year, for every 1.00% increase in the evaporation rate of raindrops, Δd decreased by 0.92‰, which is less than 1.00‰ per 1.00%, because of the higher temperature, lower relative humidity, and lower precipitation; however, a linear relationship (~1.00‰ per 1.00%) could also be observed at an evaporation rate of < 5.00%. By developing our understanding of these issues, the kinetic fractionation mechanism of stable isotopes can be explored more accurately. It can also help to improve our understanding of hydrology and water resources as reflected by the stable isotopes. Further efforts are required to provide a more accurate estimation of these influences below the clouds.

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