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Stable Isotopic Characteristics and Influencing Factors in Precipitation in the Monsoon Marginal Region of Northern China

Peipei Zhao [1], Liangcheng Tan [2,3,4,*], Pu Zhang [1,*], Shengjie Wang [5], Buli Cui [6], Dong Li [2,7], Gang Xue [2,7] and Xing Cheng [2,7]

1. College of City and Environment, Northwestern University, Xi'an 710127, China; zppxbdx@163.com
2. State Key Laboratory of Loess and Quaternary Geology, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an 710075, China; lidong@ieecas.cn (D.L.); xuegang@ieecas.cn (G.X.); chengxing@ieecas.cn (X.C.)
3. Open Studio for Oceanic-Continental Climate and Environment Changes, Qingdao National Laboratory for Marine Science and Technology, Qingdao 266100, China
4. Institute of Global Environmental Change, Xi'an Jiaotong University, Xi'an 710054, China
5. College of Geography and Environmental Science, Northwest Normal University, Lanzhou 730070, China; geowang@126.com
6. School of Resources and Environmental Engineering, Ludong University, Yantai 264025, China; cuibuli@163.com
7. University of Chinese Academy of Sciences, Beijing 100049, China

* Correspondence: tanlch@ieecas.cn (L.T.); zhangpu@nwu.edu.cn (P.Z.); Tel.: +029-6233-6228 (L.T.)

Received: 1 January 2018; Accepted: 6 March 2018; Published: 8 March 2018

Abstract: Based on stable hydrogen and oxygen isotope data ($\delta^{18}O$, δD) and meteorological observation data for complete hydrological annual precipitation from 2016 to 2017 in the monsoon marginal region of northern China (Fengxiang and Ningwu), the isotopic characteristics of precipitation and the sources of water vapor in these two regions combined were studied. The results showed that $\delta^{18}O$ and δD values in the wet season (June through September) were higher than in the dry season (October to May of the following year) in Fengxiang and Ningwu. The intercept and slope of the meteoric water line in the two regions were somewhat low, revealing that the water vapor in the rainfall comes mainly from the tropical ocean. On a synoptic scale, significantly positive correlations among dry season precipitation, $\delta^{18}O$, and temperature manifested temperature effects, but in the wet season, the temperature effect was not significant. On a monthly scale, a relationship did not exist between the change in trend of the average value of monthly weighted $\delta^{18}O$ in precipitation and the average temperature change value in the two regions. However, in the wet season, significantly negative relationships can be found between the average monthly weighted $\delta^{18}O$ in precipitation and rainfall amount, which indicated a remarkable rainout effect. Further investigation revealed that continuous precipitation made the values of $\delta^{18}O$ and δD more negative under the same source of water vapor (the rainout effect). Because the annual rainfall in the monsoon marginal region of Northern China is mainly made up of monsoon rainfall, the oxygen isotope index of geological and biological records, such as stalagmites and tree rings, which inherit meteoric water isotope information, can be used to reconstruct past rainfall changes in northern China.

Keywords: northern monsoon marginal area; precipitation; stable isotope; water vapor source

1. Introduction

The use of stable oxygen isotopes in different geological biological carriers (such as stalagmites, tree rings, and ice cores) to reconstruct past climate changes has achieved remarkable results [1–7]. In particular, stalagmites have attracted a lot of attention from paleoclimatological researchers because of its high resolution proxies and accurate dating method. In China, use of the oxygen isotope composition of stalagmites has been very successful in the Asian Monsoon Study, although there are currently some disputes about its significance [8]. For example, Cheng et al. (2016) stated that the oxygen isotope composition of stalagmites in the Chinese monsoon area reflected changes in rainfall in the entire larger spatial extent from the marine source area to the cave site, and further reflected intensity changes in the Asian monsoon over a wide spatial range [5]. Maher and Thompson (2012) and Tan (2014) suggested that variations in the oxygen isotope composition of stalagmites was mainly controlled by variations in water vapor sources in the Indian and Pacific Oceans and did not reflect changes in local rainfall [9,10]. In addition, some researchers have suggested that the oxygen isotope composition of Chinese stalagmites is controlled by changes in monsoon intensity in the upper reaches of India. However, the oxygen isotope composition of stalagmites in southern China cannot reflects changes in rainfall in the northern China [8,11–13]. Tan et al. (2015) recently suggested that the indicative significance of the oxygen isotope composition of stalagmites in southern China was different at different time scales and in different regions [14]. There is a negative correlation between the oxygen isotope composition of stalagmites and rainfall changes in some areas and within local areas [14–19]. The oxygen isotope composition recorded by geological entities is essentially inherited information on $\delta^{18}O$ in precipitation. Therefore, to study the oxygen isotope characteristics and control factors of modern precipitation, one must not only understand the modern hydrological cycle, but also explain the significance of oxygen isotope geology in the carrier, and moreover, provide a scientific basis for reconstruction of past climatic change in different areas.

A lot of modern rainfall monitoring was carried out over different regions of the world [20–22]. It was that in western Africa, the primary driver of the interannual variability of $\delta^{18}O$ in precipitation was ENSO [20]. Samuels–Crow et al. (2014) suggested that in the tropical Andes, tropical convection acts as a main controlling factor on precipitation $\delta^{18}O$ instead of temperature [21]. In the Asian Summer Monsoon region, the isotopic composition of precipitation is strongly related to the cloud-top height and convection in the dominant moisture source region and its transport paths [22]. In recent years, researchers also monitored precipitation isotopes in many areas of China. Studies in the Tibetan Plateau region showed that the northern limit of the summer monsoon is north of the Yalongzangbo River in the middle of the Tibetan Plateau, around 34° N–35° N [23]. North of 35° N, precipitation oxygen isotope composition was dominated by the westerlies (hereafter called "the westerlies domain") and depicted a close link between $\delta^{18}O$ and local temperature, there is a weak relationship with precipitation amount. South of 30° N, the precipitation oxygen isotope composition was dominated by the Indian monsoon (hereafter the "monsoon domain"), showing an antiphase between $\delta^{18}O$ and precipitation amount. In this case, the precipitation oxygen isotope composition exhibits a unique feature characterized by abrupt depletion of precipitation $\delta^{18}O$ around May, decreasing to the most severely depleted $\delta^{18}O$ value around August. In the monsoon domain between 30° N and 35° N, seasonal cycles show more complex $\delta^{18}O$ variations, and this region is defined as a transition domain, suggesting shifting influences between the westerlies and the Indian monsoon [24]. Studies of the eastern monsoon region have shown that this can be divided into three subregions. Among these, the northeastern region had the lowest isotopic values, north China had midrange values, and southern China had the highest isotopic values. In north China and northeastern China, temperature was the main factor affecting changes in precipitation $\delta^{18}O$, and a substantial effect existed only in the summer rainfall period. In fact, the summer monsoon can reach these two regions in summer. In the southern region, both an amount effect and a reverse temperature effect were found to exist. The amount effect may mask the temperature effect to some extent or even appear as a reverse temperature effect [25]. In the southern region, higher D-excess values during winter and early spring are considered to correspond to a lesser proportion of remote moisture, whereas lower D-excess values during summer and autumn correspond to larger amounts of remote moisture transported by summer monsoons [26]. Studies of the northwestern arid regions of China have shown that the slope of the local meteoric water line (LMWL) is lower than that of the global meteoric water line (GMWL). The low LMWL slope is associated with non-equilibrium conditions affecting falling raindrops under dry conditions, leading to a potential for significant subcloud evaporation. On a seasonal time scale, monthly δ^{18}Op values are more positive in summer and more negative in winter. This seasonal change is similar to that of temperature, reflecting continental climate characteristics [27]. Terrestrial moisture evaporated from Europe and central Asia may be the main moisture source around the Tianshan Mountains [28].

The scope of this paper is to enhance the understanding of the hydrological processes in the northern monsoon marginal region of China by emphasizing the isotope record of geological environmental. Here, we choose two sites, Ningwu and Fengxiang, which are located in northern China, to monitor the event-based precipitation stable isotope data (δ^{18}O, δD) for a complete hydrological year from 2016 to 2017. The controlling environmental factors are further analyzed. The temperature, precipitation amount, and relative humidity in the two sites during 2016–2017 are similar with their average values during 1982–2010, respectively, indicating their representativeness.

2. Data and Methods

2.1. Study Area

Fengxiang is located in the Western Guanzhong Plain (Figure 1), which is characterized by mountains to the north, the Southern Plateau, and the West River Valley. The annual average temperature is 11.4 °C, and the annual average precipitation is 625 mm. Ningwu is located in the north-central part of Shanxi Province (Figure 1). There are many mountains in this area, and the average annual rainfall is 445 mm. Because more than 70% of precipitation occurs from June to September in Fengxiang and Ningwu, these two areas are considered to belong to the North China monsoon marginal area.

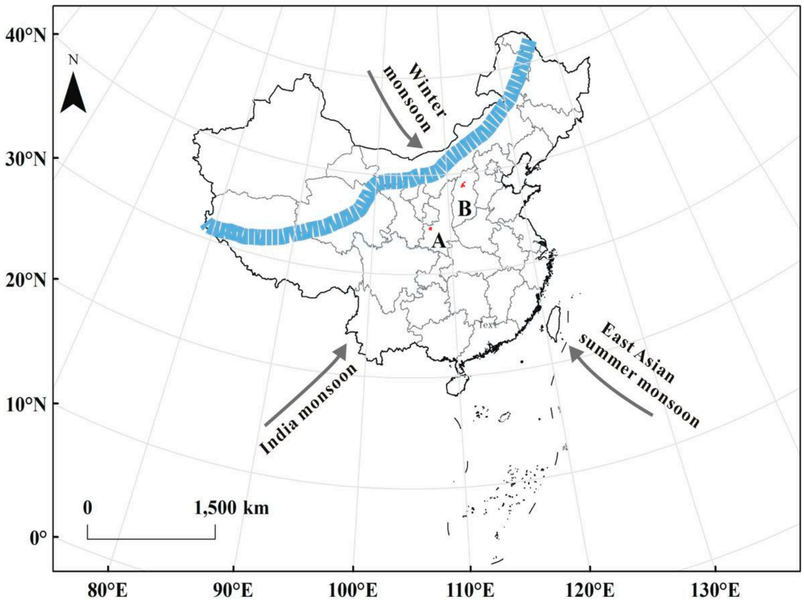

Figure 1. Precipitation sample collection site. (The blue dashed line denotes the northern limit of the Asian summer monsoon [29] (A,B) represent Fengxiang and Ningwu, respectively). The data of the map was downloaded from the National Geomatics Center of China (http:ngcc.sbsm.gov.cn/).

2.2. Sample Collection

Precipitation collection was carried out for a complete hydrological year from July 2016 to August 2017 in Fengxiang (107°27′20.1′′ E, 34°31′28.32′′ N, 767 m) and from August 2016 to August 2017 in Ningwu (112°18′14.55′′ E, 39°0′33.4′′ N, 1373 m). The collection method involved placing a water container in an open area. To prevent precipitation from evaporating, liquid samples were collected immediately after precipitation ceased, filtered using a filter head with an aperture of 0.45 m, and finally stored in 5-mL HDPE bottles with waterproof seals. Solid samples were melted at room temperature in bags before being sealed in bottles. The time and place of sampling were recorded for each sample. All samples were kept in cold storage (4 °C) [30–33]. In total, 49 and 52 precipitation samples were collected as individual events in Fengxiang and Ningwu, respectively.

2.3. Sample Analysis

All the samples were stored frozen before isotope ratio analysis using an L2130-i liquid water isotope analyzer and an IWA-35EP liquid water isotope analyzer (LGR, Institute of the Earth Environment, Chinese Academy of Sciences, Beijing, China). The results were expressed as δ-values relative to V-SMOW (Vienna Standard Mean Ocean Water). The precision of the L2130-i liquid water isotope analyzer measurement was <0.50‰ for δD and <0.20‰ for $\delta^{18}O$, and that of the IWA-35EP liquid water isotope analyzer measurement was <0.20‰ for δD and <0.03‰ for $\delta^{18}O$. Isotopic monthly precipitation values were calculated using the weighted mean value (VMA). Taking $\delta^{18}O$ as an example, the expression for the weighted mean was:

$$\delta^{18}\mathrm{O_e}\sum_{i=1}^{N} M_\mathrm{i}\delta^{18}\mathrm{O}_{\mathrm{m*i}}/\sum_{i=1}^{N} M_\mathrm{I} \quad (1)$$

where $\delta^{18}O_e$ was the calculated $\delta^{18}O$ deviation value, $\delta^{18}O_{m*i}$ was the actual measured $\delta^{18}O$ deviation value, and M_i was the amount of the i-th precipitation event.

In addition, the rainout effect of precipitation and the transfer process of atmospheric air mass for precipitation events involving $\delta^{18}O$ and δD stable isotopes were tested by the air mass trajectory method. The analysis was based on the HYSPLIT model (http://ready.arl.noaa.gov/HYSPLIT.php). Because the 850-hPa height (~1500 m asl) was approximately considered as cloud-base or precipitation height, an 850-hPa starting height (~1500 m asl) was used in the model [34–36]. The selection time was 00:00, and the backward duration was set as 120 h. Finally, the graph was visualized in ArcMap 10.2.

Related meteorological information for Fengxiang and Ningwu was provided by the Meteorological Bureaus of Fengxiang and Ningwu 5 km and 1.8 km from our sampling site, respectively.

3. Results and Discussion

3.1. Seasonal Variation of Stable Isotopes in Precipitation

From July 2016 to August 2017 in Fengxiang, δD varied from −85.11‰ to 14.23‰, with a mean value of −35.70‰. The $\delta^{18}O$ varied from −11.92‰ to 0.83‰, with a mean value of −5.52‰ (Figure 2A). Increased precipitation in the wet season (June to September) accounted for 70.3% of total precipitation. The average value of δD was −37.32‰, and the average value of $\delta^{18}O$ was −5.24‰. Reduced precipitation in the dry season (October to May of the following year) accounted for 29.7% of total precipitation. The average value of δD was −33.88‰ and the average value of $\delta^{18}O$ was −5.85‰.

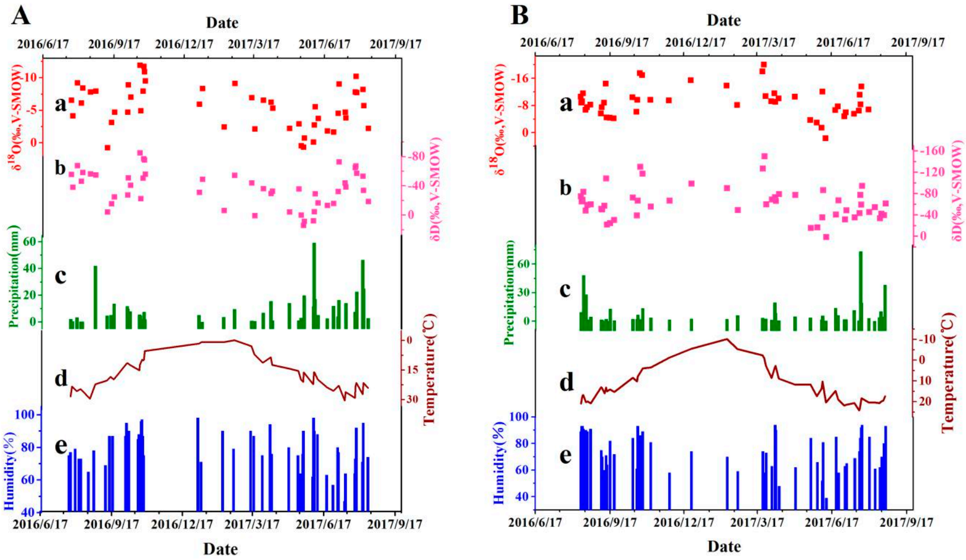

Figure 2. Seasonal variation of δD, $\delta^{18}O$, and meteorological information of precipitation. (**A**,**B**) represent Fengxiang and Ningwu, respectively. (**a**) indicates individual precipitation $\delta^{18}O$ (‰, V-SMOW); (**b**) indicates individual precipitation δD (‰, V-SMOW); (**c**) indicates individual precipitation amount (mm); (**d**) indicates daily surface air temperature (°C); and (**e**) indicates daily surface air humidity (%).

From August 2016 to August 2017 in Ningwu, δD varied from −149.71‰ to 2.00‰ with a mean value of −61.39‰. The $\delta^{18}O$ varied from −19.96‰ to 1.72‰ with a mean value of −8.72‰ (Figure 2B). Increased precipitation in the wet season (June to September) accounted for 78.7% of total precipitation. The average value of δD was −52.56‰ and the average value of $\delta^{18}O$ was −7.23‰. Reduced precipitation in the dry season (October to May of the following year) accounted for 21.3% of total precipitation. The average value of δD was −75.51‰ and the average value of $\delta^{18}O$ was −11.11‰.

Stable hydrogen and oxygen isotope values in Fengxiang and Ningwu precipitation showed seasonal variation and the patterns of variation for temperature and precipitation were similar (Figure 2). Seasonal variation of δD and $\delta^{18}O$ showed significantly more positive values occurring during summer and more negative values occurring during winter. The most negative values generally occurred in October, which may be related to the greater rainout fraction of precipitation falling on the ground during the monsoon period. Some strongly positive $\delta^{18}O$ values were found in some precipitation events in May before the monsoon; these may be related to high temperature, low precipitation amount, and high evaporation. The isotopic contents of precipitation were mainly controlled by the effect of subcloud evaporation.

3.2. Local Meteoric Water Line

Using all the event-based samples in this study, a local meteoric water line (LMWL) in Fengxiang was established as (Figure 3A):

$$\delta D = 7.05\delta^{18}O + 3.24\ (r = 0.944,\ n = 49) \quad (2)$$

Using the event-based values, the LMWLs for the dry and wet seasons, respectively, were:

$$\delta D = 7.13\delta^{18}O + 7.87\ (r = 0.968,\ n = 23), \quad (3)$$

$$\delta D = 7.20\delta^{18}O + 0.37\ (r = 0.943,\ n = 26). \quad (4)$$

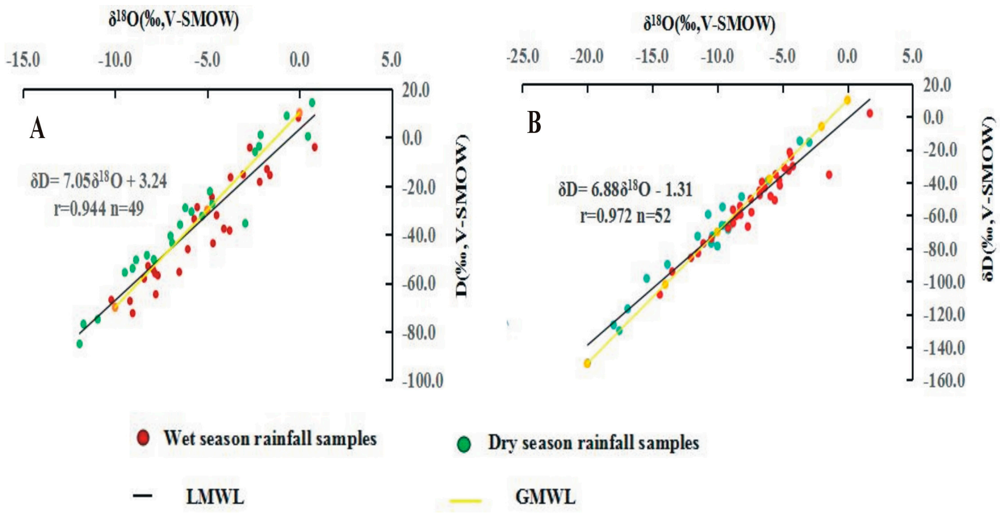

Figure 3. Distribution of δD and $\delta^{18}O$ of precipitation. (**A**,**B**) represent Fengxiang and Ningwu, respectively. LMWL = local meteoric water line; GMWL = global meteoric water line.

Similarly, using all the event-based samples in this study, a local meteoric water line (LMWL) in Ningwu was established as (Figure 3B):

$$\delta D = 6.88\delta^{18}O - 1.31\ (r = 0.972,\ n = 52). \quad (5)$$

Using the event-based values, the LMWLs for the dry and wet seasons, respectively, were:

$$\delta D = 7.59\delta^{18}O + 8.82\ (r = 0.982,\ n = 20), \quad (6)$$

$$\delta D = 6.59\delta^{18}O - 4.92\ (r = 0.958,\ n = 32). \quad (7)$$

The slope and intercept of the local meteoric water line (LMWL) in the two regions were different from the slope and intercept of the global meteoric water line (GMWL), reflecting the unique regional atmospheric circulation modes, water vapor sources, and evaporation modes for each region [37]. The slope and intercept of the LMWL in both regions were less than the slope and intercept of the GMWL (δD = 8, $\delta^{18}O$ + 10) [37] and the China meteoric water line (δD = 7.9, $\delta^{18}O$ + 8.2) [38]. The low LMWL slope was associated with non-equilibrium conditions affecting falling raindrops during dry conditions, leading to the potential for significant subcloud evaporation [39,40]. The drier the atmosphere, the lower the slope and intercept of the water line will be. This phenomenon reveals the arid climate in the northern monsoon marginal region of China. In the dry season, the slope and intercept of the LMWL in both regions were greater than in the wet season. This may have been related to differences between water vapor sources and dynamic fractionation.

Comparing the LMWL ($\delta D = 7.05\delta^{18}O + 3.24$, $r = 0.944$) in Fengxiang with values from Weinan (2012–2014) ($\delta D = 7.586\delta^{18}O + 10.514$ ($R^2 = 0.97$)), Xi'an (1985–1993) ($\delta D = 7.49\delta^{18}O + 6.13$ ($r = 0.958$)), and Changwu (2005, 2010, 2013) ($\delta D = 7.36\delta^{18}O + 3.59$ ($r = 0.94$)) [41–43], it was found that along the Weinan–Xi'an–Changwu–Fengxiang line from east to west, LMWL slope and intercept continuously decreased. This indicated that these places lie along the same water vapor transport path. In the water vapor transport process, the fraction of precipitation isotopes in the greater rainout portion was increased.

With reference to the GMWL (Figure 3, yellow line), the dry season rainfall points were found mostly above the upper GMWL line (low $\delta^{18}O$ values, $d > 10\%$), indicating that these points represented mainly winter precipitation at low temperature and low absolute air moisture content. Most of the rainfall in the wet season was located below the GMWL line (high $\delta^{18}O$ values), indicating that the water vapor comes mainly from the ocean with its high relative humidity and may be subject to subcloud evaporation [44–46].

3.3. Correlations between $\delta^{18}O$ and Climate Parameters

3.3.1. At the Synoptic Scale

At the synoptic scale, significantly positive relationships between precipitation $\delta^{18}O$ and dry season temperature were found to exist in both regions (Figure 4), showing a temperature effect [47]. Ningwu showed a temperature effect over the whole year (Figure 4B). However, in the wet season, no temperature effect existed in either place.

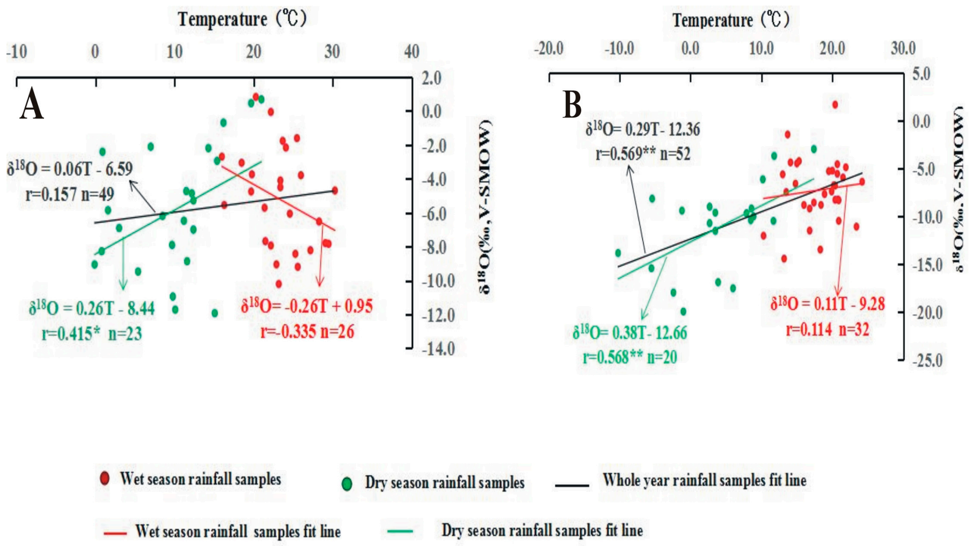

Figure 4. Relationship between $\delta^{18}O$ precipitation and temperature. (**A**,**B**) represent Fengxiang and Ningwu, respectively. ** means a significant bilateral correlation at the 0.01 level; * means a significant bilateral correlation at the 0.05 level.

The relationship between precipitation $\delta^{18}O$ and precipitation amount was not significant over the whole year or in the dry season in Fengxiang (Figure 5A). This showed the rainfall amount effect did not exist at a synoptic scale in the northern monsoon marginal region.

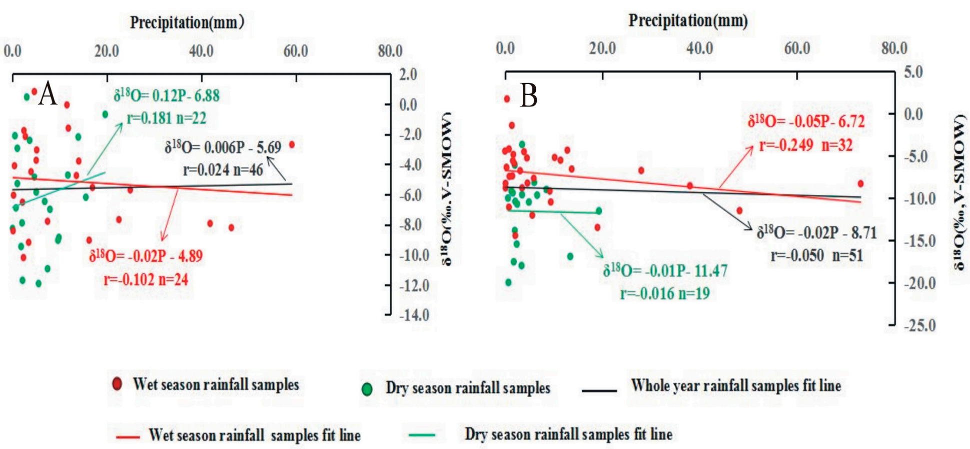

Figure 5. Relationship between $\delta^{18}O$ precipitation and precipitation amount. (**A**,**B**) represent Fengxiang and Ningwu, respectively.

3.3.2. Monthly Average Scale

Although the temperature effect of precipitation $\delta^{18}O$ existed at a synoptic scale in Fengxiang and Ningwu, as did the amount effect of precipitation $\delta^{18}O$ on monthly weighted average, the relationship between $\delta^{18}O$ weighted average monthly precipitation change trend and monthly average temperature change was not significant in Fengxiang and Ningwu (Figure 6). For example, in Fengxiang, the temperature decreased in January 2017, but $\delta^{18}O$ in precipitation increased. In Ningwu, the temperature decreased in November 2016, but $\delta^{18}O$ in precipitation also increased.

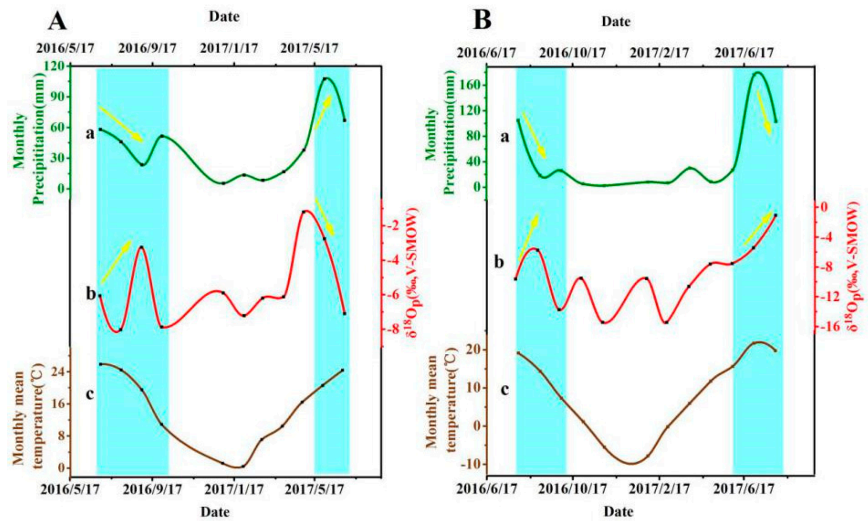

Figure 6. Relationship between monthly weighted mean precipitation $\delta^{18}O$ and related meteorological factors. (**A**,**B**) represent Fengxiang and Ningwu, respectively. (**a**) indicates monthly precipitation amount (mm); (**b**) indicates monthly weighted mean $\delta^{18}O$ value in precipitation (‰, V-SMOW); and (**c**) indicates monthly average surface air temperature (°C).

From the monthly precipitation amounts, the negative correlation between weighted average monthly precipitation $\delta^{18}O$ and monthly precipitation amounts was significant. For example, in Fengxiang, the precipitation amount increased in July 2017, but precipitation $\delta^{18}O$ decreased. In Ningwu, the precipitation amount increased in August 2016, but precipitation $\delta^{18}O$ also decreased. Even during the dry season in Fengxiang, monthly precipitation weighted average $\delta^{18}O$ was negatively correlated with monthly precipitation amount, showing a remarkable precipitation amount effect. For example, precipitation amount increased in October 2016 and in February 2017, but precipitation $\delta^{18}O$ decreased.

Annual rainfall in the northern monsoon marginal area of China is mainly made up of monsoon rainfall, which accounts for more than 80% of annual rainfall. Oxygen isotopes in stalagmites are usually recorded in monthly, annual, and even multiple years of weighted mean precipitation oxygen isotopes. From this point of view, the oxygen isotope records of stalagmites from the northern monsoon fringe area can reflect changes in local monsoon rainfall amounts. A good negative correlation was recently obtained between 165-year-old stalagmite oxygen isotope records in Shihua Cave and rainfall amounts obtained in Beijing [48], in agreement with our interpretation. This result was also consistent with the simulation results obtained by Liu et al. (2014) [49].

3.4. *Rainout Effect*

The effect of continuous rainfall on isotope composition was also examined. HYSPLIT backward trajectory analyses showed that on 20 and 21 October 2016, rainfall water vapor from Central Asia was carried by the westerly wind to Ningwu (Figure 7). During two days of continuous rainfall, the precipitation $\delta^{18}O$ values decreased from −6.10‰ to −9.65‰, and δD values decreased from −38.80‰

to −66.11‰; δ^{18}O and δD decreased by 3.55‰ and 27.31‰, respectively. When water vapor came from the South China Sea on 23 and 24 July 2017 (Figure 7), the precipitation δ^{18}O values decreased from −8.30‰ to −13.47‰, and δD values decreased from −58.65‰ to −94.17‰; δ^{18}O and δD decreased by 5.17‰ and 35.52‰, respectively. On 5 and 6 October 2016, during two days of continuous rainfall in Fengxiang, the precipitation δ^{18}O values decreased from −4.72‰ to −8.86‰, and δD values decreased from 27.33‰ to −50.50‰; δ^{18}O and δD decreased by 4.14‰ and 21.73‰, respectively. On 4 and 5 June 2017, the precipitation δ^{18}O values decreased from −2.69‰ to −5.54‰, and δD values decreased from −4.28‰ to −28.81‰; δ^{18}O and δD also decreased by 2.85‰ and 24.53‰, respectively. From this point of view, when the source of water vapor is constant, continuous heavy monsoon rainfall will lead to a gradual negative bias of oxygen isotopes in the north China monsoon marginal area, which also explains the negative correlation between oxygen isotope concentration and rainfall amount in stalagmites in this area [15–17,48]. In addition, if rainfall occurs mainly in winter in a region and the water vapor source is relatively stable, the rainout effect can be expected. Indeed, the oxygen isotope concentration of stalagmites from Fukugaguchi Cave in North Central Japan had a significant negative correlation with rainfall amount in winter. The rainfall in this area mainly occurs in wintertime and is caused by winter wind [50].

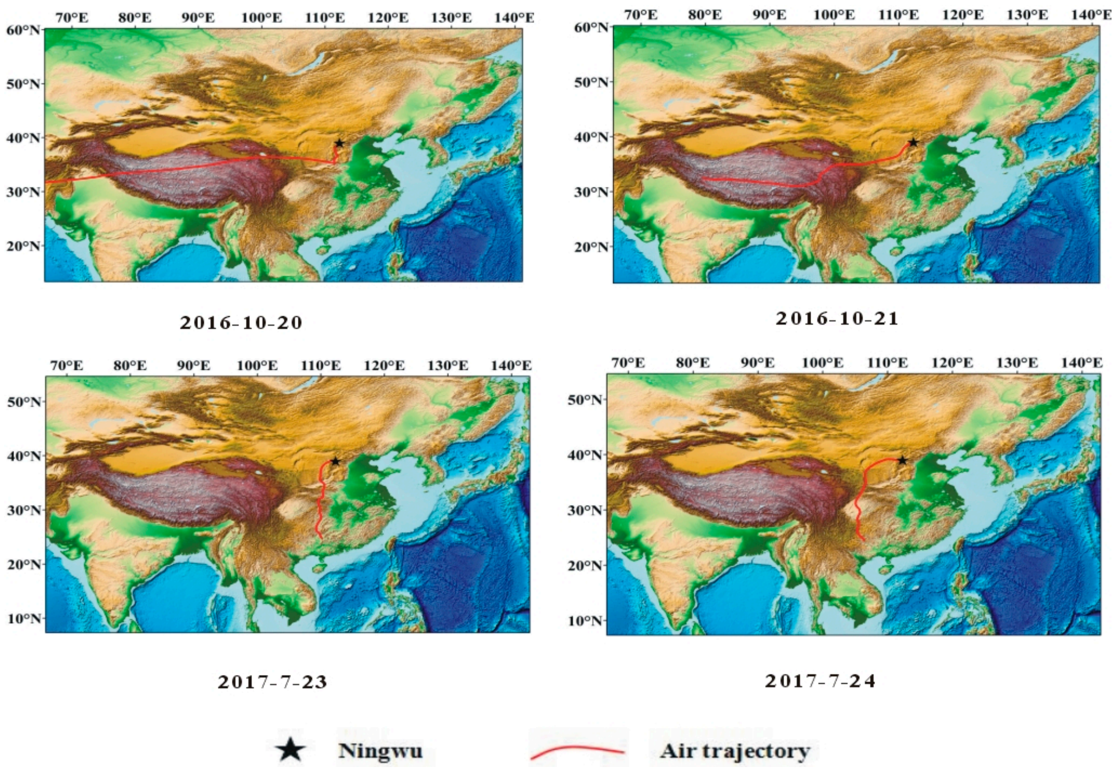

Figure 7. Tracking results for water vapor transport in Ningwu.

4. Conclusions

From the research described above, the following conclusions can be drawn:

(1) The δ^{18}O and δD values for individual precipitation events in the wet season (June to September) were higher than in the dry season (October to May of the following year) over one year in Fengxiang and Ningwu. The most negative values generally occurred in October and may be related to the greater rainout fraction of precipitation falling on the ground during the monsoon period. Some of the most strongly positive δ^{18}O values occur in precipitation events in May before the monsoon. These may be related to high temperature, low precipitation amount, and high evaporation. The isotopic fraction of precipitation was mainly controlled by the effect of subcloud evaporation.

(2) Using all the event-based samples in this study, local meteoric water lines (LMWLs) in Fengxiang [$\delta D = 7.05\delta^{18}O + 3.24$ ($r = 0.944$, $n = 49$)] and Ningwu [$\delta D = 6.88\delta^{18}O - 1.31$ ($r = 0.972$, $n = 52$)] were established. The intercept and slope of the local meteoric water line in both regions were small, which revealed that the water vapor in the rainfall came mainly from the moist ocean area. In the dry season, the slope and intercept of the LMWL in both regions are greater than in the wet season. This may be related to the difference between water vapor sources and dynamic fractionation. It was also found that along the Weinan–Xi'an–Changwu–Fengxiang line, from east to west, LMWL slope and intercept continuously decreased, indicating that these places lie along the same water vapor transport path. In the water vapor transport process, the fraction of precipitation isotopes in the greater rainout portion was increased.

(3) At the synoptic scale, significantly positive relationships among precipitation $\delta^{18}O$ and dry season temperature were observed. However, in the wet season, the temperature effect was not significant. At a monthly average scale, the relationship between monthly weighted average $\delta^{18}O$ precipitation change trends and average temperature change trends was not observed, whereas in the wet season, significantly negative relationships between monthly weighted average precipitation $\delta^{18}O$ and rainfall amount were found, indicating an amount effect.

Monsoon rainfall makes up more than 80% of the annual rainfall in the north monsoon marginal area of China. Oxygen isotopes of geological records, such as stalagmites, are usually recorded in monthly, annual, and even multiple years of weighted mean precipitation oxygen isotopes. Hence, oxygen isotope records in stalagmites from the northern monsoon fringe area can reflect changes in local monsoon rainfall amounts. Long-term observations will help to explain the oxygen isotopes in geological records [51,52] on a decadal or even longer timescale.

Acknowledgments: This work was supported by the National Key Research and Development Program of China (2017YFA0603401), the Natural Science Foundation of China (41372192), Science Foundation for Distinguished Young Scholars of Shaanxi Province, China, Youth Innovation Promotion Association of Chinese Academy of Sciences, and West Light Foundation of Chinese Academy of Sciences. Related meteorological information for Fengxiang and Ningwu was provided by the Meteorological Bureaus of Fengxiang and Ningwu, respectively.

Author Contributions: Liangcheng Tan designed the experiments, analyzed the data and revised the paper. Peipei Zhao performed the experiments, collected data and wrote the paper. Pu Zhang revised the paper. Shengjie Wang and Buli Cui analyzed and interpretated data. Dong Li, Gang Xue and Xing Cheng contributed in sample collection and paper preparation.

Conflicts of Interest: The authors declare no conflict of interest.

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