Deuterium Excess in Precipitation Reveals Water Vapor Source in the Monsoon Margin Sites in Northwest China

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Abstract: The deuterium excess (d) in precipitation, determined by the stable hydrogen and oxygen isotopes (δ2H and δ18O), is a widely applied parameter in tracing the water vapor source. Based on the multiple-year observations of stable water isotopes in precipitation at four stations in the Lanzhou city, Northwest China, we analyzed the variations in deuterium excess in precipitation at the Asian monsoon margin region. The mean value of deuterium excess at the study region is 11.0% in the dry season and 8.0% in the wet season. The d value in precipitation negatively correlates with air temperature and vapor pressure. The low d value during the wet season reflects the monsoon moisture transported from long distances. During the dry season, the continental air masses correspond to the higher d value in precipitation. The moisture regimes based on reanalysis data are generally consistent with the findings using a stable isotopic approach, and the monsoon moisture is highlighted in summer precipitation at these monsoon margin sites.

Keywords: deuterium excess; water vapor source; precipitation; monsoon margin; Lanzhou

1. Introduction

Water resources and especially fresh water are indispensable for human survival and development [1,2]. However, fresh water accounts for 2.5% of the global water and only 0.3% of it can be easily used, indicating the water is unevenly distributed in space and time [3]. Since precipitation is the direct input of surface water, groundwater, glacier and other hydrologic components, it determines whether a region has abundant water resources or not. At present, atmospheric circulation is undergoing significant changes due to global warming and so is precipitation [4,5]. Stable water isotope ratios (δ2H and δ18O) in precipitation are sensitive to environmental conditions so they can be used as natural records of environmental changes [6–8]. Water bodies from different sources often have different isotope ratios; therefore, their isotopic signature allows us to trace these sources. Deuterium excess (d = δ2H − 8δ18O; [9]) in meteoric water reflects the evaporation degree for the water vapor source areas. The higher the evaporation degree, the larger the d value is; after a raindrop is condensed, the stronger the re-evaporative fractionation under the cloud, the smaller the d value is. The d value is not only affected by temperature, relative humidity as well as wind speed at the locations where the water vapor sourced but also by the fractionation due to re-evaporation of raindrops as they leave the cloud (below cloud evaporation) [10,11]. In the Mediterranean area, d values are generally higher in
autumn–winter precipitation than those in spring-summer precipitation, which can be attributed to
the influence of Mediterranean air masses [12,13].

Although a number of methods can be applied to trace the moisture sources, in recent years,
increasing attention has been paid to the stable isotopes in precipitation and water vapor [14–16].
As far as isotope techniques are concerned, knowledge has been accumulated worldwide regarding
the relation between the isotopic composition and the origin of the water vapor in various regions of
the world [17–20]. Since the \( d \) value in precipitation depends on meteorological conditions at the water
vapor source regions, changes in \( d \) values in precipitation can be used to trace the water vapor to a
certain area [21–24].

In western China, precipitation isotopes have been considered in the identification of water vapor
sources [25,26]. In the Tibetan Plateau, Yao et al. [27] identified three sub-regions with seasonal isotopic
features impacted by different large-scale moisture paths (the westerlies in the north, the transition in
the middle as well as the monsoon in the south), considering that the potential interaction between the
westerlies and monsoon is the critical driving force of different isotope characteristics. Liu et al. [28]
demonstrated that raindrops in northwest China are subjected to below cloud evaporation during
precipitation, so the precipitation vapor may be mixed with locally recycled vapor by means of both
Rayleigh and kinetic fractionation. Feng et al. [29] showed a case study linking the isotopic variation
to moisture sources, and found that there is a dominant effect of westerly moisture in summer and of
a combined influence of westerly and polar air masses in winter. Wang et al. [30,31] demonstrated
that the rainfall in westerly-dominated northwest China is related to locally recycled moisture and the
advected moisture via Europe and central Asia.

The Lanzhou city, located in the Loess Plateau of China, is impacted by both westerlies and
monsoon water vapor [32–34]. This region is also considered to be the western vertex of the Chinese
“Monsoon Triangle” defined by Li et al. [35], and is usually sensitive to modern and paleo climate
change. The geographic location at the monsoon margin makes the water cycle at the Lanzhou city
interesting for hydrological and climate studies. From 2011 to 2012, 243 samples were collected at
Lanzhou and the basic characteristics of \( d \) value were mentioned [36]. With further observation from
2011 to 2014 [33], the inter-annual variation of \( d \) value may provide more meaningful information
about the origin of water vapor over this the region. However, the moisture sources information in \( d \)
value in precipitation samples was not well treated for this region using multiple year event-based
observations in previous studies.

Here, based on 959 event-based samples from four stations in the Lanzhou city between April
2011 and October 2014 [33], we aim (1) to present new detailed information regarding the \( d \) value in
precipitation in the study region, (2) to clarify the relation between \( d \) value and meteorological factors
(especially temperature and vapor pressure) and (3) to discuss the linkage of stable isotopes and water
vapor origin for these monsoon margin stations.

2. Data and Method

2.1. Study Area

The Lanzhou city (Figure 1) is situated at a transition area between the three natural sub-regions
of China including the eastern monsoon region, the northwestern arid region and the Tibetan cold
region. Lying at the monsoon margin region, the Lanzhou city presents a cold semi-arid climate (BSk)
in Köppen–Geiger climate classification. The Indian and East Asia monsoons can be delivered to
this region, leading to an uneven intra-annual precipitation distribution; however, the mid-latitude
westerlies regime cannot be ignored. As recorded at the Lanzhou National Basic Meteorological Station
(36.05° N, 103.88° E, 1517.2 m) [33], the long-term mean precipitation amount is 293.9 mm, and the
fraction of precipitation is 85% during the wet season (from May to September) and 15% during the
dry season (from October to April); the pan evaporation amount is 1504.8 mm per year; the annual
mean air temperature is 10.4 °C, and the monthly temperature ranges from 23.1 °C in July to −4.5 °C in January.

![Map showing the four stations](image)

**Figure 1.** Map showing the four stations (Anning, Yongdeng, Gaolan and Yuzhong marked as black pins) in the Lanzhou city, China. The stations involved in Chen et al. [33] but not included in this study are also marked as white circles. The small map indicates the location of the Lanzhou city in China.

### 2.2. Sample Collection and Laboratory Work

The selected stations for precipitation sampling in this study are those of Anning, Yongdeng, Gaolan and Yuzhong from April 2011 to October 2014 [33] (Table 1). Samples in all stations were collected by accredited meteorological observers, except at Anning where sampling was performed by the members of the research group of Northwest Normal University. In order to prevent the possible evaporation in the collector, liquid samples (rain) were immediately gathered after the end of each event. Continuous (and usually with large amount) processes were selectively collected based on fixed time intervals (e.g., every hour). Liquid samples collected were put into approximately 50 mL plastic bottles with outer and inner caps as well as waterproof seals and then stored in a refrigerator. Solid samples (snow or hail) were firstly allowed to melt at room temperature in zip-lock plastic bags; once in liquid form, they were handled as described before. Air temperature, vapor pressure and relative humidity during precipitation events were recorded on an hourly basis using automatic weather stations, and arithmetically averaged for each precipitation event. Samples were analyzed using a liquid water isotope analyzer DLT-100 (Los Gatos Research, Inc., Mountain View, CA, USA) in the College of Geography and Environmental Science, Northwest Normal University, and the analytic precision was ±0.2‰ and ±0.6‰, for δ¹⁸O and δ²H, respectively. We presented the stable isotopic characteristics for the period from April 2011 to October 2014 in a previous study [33]. In this study, we focus on the second-order parameters deuterium excess, instead of the raw stable hydrogen and oxygen isotope compositions.

We acquired the historical data in Lanzhou (103°52′ E, 36°03′ N, 1517 m) from the Global Network of Isotopes in Precipitation (GNIP) [37]. The simulations of several isotope-enabled general circulation models (GCMs), including Laboratoire de Météorologie Dynamique Zoom (LMDZ), Isotope-incorporated Global Spectral Model (isoGSM) and Model for Interdisciplinary Research on Climate (MIROC), within 34–37° N and 103–105° E were selected (online available at [http:][37]).
The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model \cite{42} using the Global Data Assimilation System (GDAS) \cite{43} was applied in order to calculate the air mass trajectories. The weather systems giving the precipitation events were determined using the US National Centers for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) reanalysis \cite{44}.

Table 1. Latitude (Lat), longitude (Lon), altitude (Alt), meteorological parameters (T—air temperature, P—precipitation amount, e—water vapor pressure and h—relative humidity) and their standard deviations ($\sigma$) at four sampling stations in the Lanzhou city from 2011 to 2014.

| Station | Lat (°) | Lon (°) | Alt (m) | T (°C) | P (mm) | e (hPa) | h (%) |
|---------|---------|---------|---------|--------|--------|---------|-------|
|         | Mean    | σ       | Mean    | σ      | Mean   | σ       | Mean  | σ    |
| Anning  | −28.3   | 23.4    | 5.1     | 8.7    | 7.9    | 4.1     | 9.1   |
| Yuzhong | −13.6   | 28.4    | 11.1    | 11.1   | 7.7    | 11.2    | 7.7   |
| Gaolan  | −36.4   | 33.3    | 9.3     | 10.2   | 7.5    | 8.9     | 11.6  |
| Yongdeng| −25.3   | 27.7    | 9.1     | 13.8   | 10.6   | 7.9     | 8.4   |

3. Results and Discussion

3.1. Deuterium Excess in Precipitation and Environmental Controls

Table 2 and Figure 2 show the long-term variations in $d$ value in precipitation at the four stations in the Lanzhou city from 2011 to 2014. Generally, the seasonal variability in $d$ value in precipitation can be seen, which is related to the meteorological conditions with higher temperature in the dry season (October to April) and lower temperature in the wet season (May to September). The regional mean $d$ value is 11.0% in the dry season and 8.0% in the wet season, respectively. Although four sampling stations are presented in Figure 2, the inter- and intra-annual variations are generally consistent. The historical data in the GNIP Lanzhou site are presented in Figure S1 in Supplementary Material; different sampling years are involved, but the basic characteristics of seasonal variability are generally consistent.

Table 2. Minimum, maximum, mean values and standard deviation ($\sigma$) of $d$ value for the wet (May to September) and dry (October to April) seasons at the four stations in the Lanzhou city.

| Station | Min (%o) | Max (%o) | Mean (%o) |
|---------|----------|----------|-----------|
|         |          |          | Annual    | Dry Season | Wet Season |
|         | Mean     | σ        | Mean     | σ        | Mean     | σ        |
| Anning  | −28.3    | 23.4     | 5.1      | 8.7      | 7.9      | 4.1      | 9.1    |
| Yuzhong | −13.6    | 28.4     | 11.1     | 11.1     | 7.7      | 11.2     | 7.7    |
| Gaolan  | −36.4    | 33.3     | 9.3      | 10.2     | 7.5      | 8.9      | 11.6   |
| Yongdeng| −25.3    | 27.7     | 9.1      | 13.8     | 10.6     | 7.9      | 8.4    |

Four simulations of isotope-enabled GCMs are shown in Figure 3, including LMDZ, LMDZ$_\text{free}$, isoGSM and MIROC. Although a slight fluctuation in $d$ value can be seen, the mean levels in the wet season are usually lower than that in the dry season, which is consistent with the observed results in this study. It should be mentioned that it is still necessary to improve the accuracy of isotope-enabled GCMs in China, because the differences do exist among these models.

Air temperature and vapor pressure influence the re-evaporation of the falling raindrops, affecting the isotopic signature in precipitation \cite{45–47}. According to event-based samples collected in this region, $d$ value is usually negatively correlated with local air temperature and vapor pressure (Table 3).
This shows that $d$ value in precipitation in a warm and humid environment is lower than that in a cold and dry environment. The $\delta^{2}H$ value during the wet season was higher than that during the dry season at the four stations from 2011 to 2014. The local meteoric water line (LMWL) slopes during the wet season were lower than those during the dry season. The $d$ value increased during the dry season, showing that a large amount of water vapor from local evaporation mixes with the westerly air masses and then impacts the study region [48,49]. Similar correlations are also presented using the GNIP Lanzhou station in Figure S2 in Supplementary Material.



Figure 2. Variations of $d$ value at the four stations in the Lanzhou city from 2011 to 2014.
Asia monsoons. Winter monsoon in China weakens from mid-February to mid-April, and strengthens from mid-September to early December; in contrast, summer monsoon starts and strengthens from May to September. This pattern is consistent with the seasonal variation of wind direction and atmospheric pressure systems. The monsoon flow brings significant amounts of water vapor to the study region, affecting local precipitation patterns. Similar seasonal variations are observed in other regions with monsoonal climates.

Table 3. Comparisons of mean d value, δ²H, correlation coefficients (r) between d value and air temperature (T) or water vapor pressure (e), and the local meteoric water lines (LMWLs) during the wet (May to September) and dry (October to April) seasons at the four sites in the Lanzhou city.

| Station | Season | d (%) | δ²H (%) | r (d vs. T) | r (d vs. e) | LMWL |
|---------|--------|-------|---------|-------------|-------------|------|
| Anning  | Wet    | 4.1   | -26.6   | -0.40       | -0.08       | δ²H = 7.035δ¹⁸O + 0.75 (r² = 0.95, p < 0.01, σslope = 0.13, σintercept = 0.87, n = 155) |
|         | Dry    | 8.7   | -61.2   | -0.21       | -0.10       | δ²H = 7.855δ¹⁸O + 7.63 (r² = 0.96, p < 0.01, σslope = 0.28, σintercept = 2.70, n = 34) |
| Yuzhong | Wet    | 11.2  | -46.3   | -0.15       | -0.04       | δ²H = 7.47δ¹⁸O + 7.10 (r² = 0.97, p < 0.01, σslope = 0.11, σintercept = 1.02, n = 154) |
|         | Dry    | 11.1  | -57.2   | 0.03        | 0.05        | δ²H = 7.48δ¹⁸O + 6.52 (r² = 0.97, p < 0.01, σslope = 0.17, σintercept = 1.68, n = 71) |
| Gaolan  | Wet    | 8.9   | -36.0   | -0.34       | -0.08       | δ²H = 7.40δ¹⁸O + 5.55 (r² = 0.88, p < 0.01, σslope = 0.19, σintercept = 1.34, n = 200) |
|         | Dry    | 10.2  | -41.6   | -0.02       | -0.15       | δ²H = 8.02δ¹⁸O + 10.13 (r² = 0.89, p < 0.01, σslope = 0.35, σintercept = 2.48, n = 63) |
| Yongdeng| Wet    | 7.9   | -26.3   | -0.11       | -0.17       | δ²H = 7.18δ¹⁸O + 4.17 (r² = 0.94, p < 0.01, σslope = 0.12, σintercept = 0.70, n = 233) |
|         | Dry    | 13.8  | -50.5   | 0.08        | 0.00        | δ²H = 7.12δ¹⁸O + 6.53 (r² = 0.92, p < 0.01, σslope = 0.29, σintercept = 2.77, n = 54) |

3.2. Stable Isotopes and Circulation Background

The climate regime in China is greatly influenced by monsoon moisture from the Indian and East Asia monsoons. Winter monsoon in China weakens from mid-February to mid-April, and strengthens from mid-September to early December; in contrast, summer monsoon starts and strengthens from...
mid-April to mid-June, and weakens from mid-August to mid-September. Although the study region lies at the monsoon margin, the monsoon moisture does play a vital role in local precipitation and water resources. To understand the circulation background relating to stable isotopes in precipitation, here, we applied the HYSPLIT model to calculate the backward trajectory to the Lanzhou city for each precipitation event.

The backward trajectories for the four seasons (March to May as spring, June to August as summer, September to November as autumn, and December to February as winter) from 2011 to 2014 are shown in Figure 4. Two different target altitudes—i.e., 1000 (Figure 4a,c,e,g) and 1500 m (Figure 4b,d,f,h)—are involved. To identify the main paths to the study region, we applied a cluster mean method incorporated into the HYSPLIT model. Since summer precipitation trajectories account for 53% in total, we firstly focused on the cluster mean in summer. Precipitation paths can be generally attributed into two directions—i.e., the northwest (T1 in Figure 4c,d; in yellow) and southeast (T2 in Figure 4c,d; in brown)—the southeast direction accounts for a slightly higher proportion than the northwest; although different target heights were calculated here, there is no significant difference in Figure 4c,d, and the southeast trajectories account for 56% and 58% for 1000 and 1500 m, respectively. Summer monsoon carries abundant moisture, which greatly contributes to the local precipitation. On the contrary, the contribution of the westerly trajectory to the summer precipitation is logically lower. The variations of the stable water isotopes in precipitation mentioned above and in previous reports [33], together with the advance and retreat period of monsoon, indicate the existence of a long-distance monsoon moisture transport to the Lanzhou city.

**Figure 4.** Clustered backward trajectories during different seasons to the Lanzhou city. The target altitudes are set as 1000 (a,c,e,g) and 1500 m (b,d,f,h), respectively. The percentages in brackets denote the proportions in trajectory amount for each cluster in total amount.
The shift of moisture sources may cause variability in $d$ value in precipitation. In spring (Figure 4a,b), the spatial pattern was generally similar to that in summer, but the contribution from the southeast path was much smaller; for example, for the target height of 1000 m, the contributions in spring and summer were 34% and 56%, respectively. In autumn (Figure 4e,f), as the monsoon retreated, the moisture trajectory via the southeast path became weak. In winter (Figure 4g,h), the monsoon moisture via the southeast direction could not be detected in the clustered trajectories, and the westerlies were considered as the dominant moisture paths; because of the low moisture loading in westerlies mass, the precipitation in winter is generally limited.

We also calculated the clustered trajectories for each year from 2011 to 2014 (Figures S3–S6 in Supplementary Material): the results are generally consistent with the above findings.

Usually, as the precipitating air mass transports, the stable hydrogen and oxygen isotopes present a depleting trend due to the rainout effect [9]; however, this is not always true for a specific small domain. Here, we analyzed four typical sequentially rainfalls in April, June and August in 2014 without negative trends [33]. The water vapor paths before the four typical events provide a spatial perspective to identify the linkage between isotopic variability and moisture transport. Figure 5 shows the 24-h backward trajectories in which the green, blue and red lines denote the target heights of 500, 1000 and 1500 m, respectively. The slightly different air mass trajectories for the three heights may also impact the fractionation of stable isotopes in precipitation, although the moisture paths are generally similar for different heights. Taking August 3rd, 2014 as an example, the closer to the ground the air mass was, the further south the trajectory extended, and the farther from the ground the air mass was, the further east the trajectory was.

**Figure 5.** Backward trajectories for four typical rainfall events during the past 24 h: (a) 15 April 2014, (b) 18 June 2014, (c) 28 June 2014, and (d) August 3, 2014. The green, blue and red lines denotes the target heights of 500, 1000 and 1500 m, respectively.

### 3.3. Stable Isotopes and Water Vapor Transport

The precipitable water ($PW$) and water vapor stream can be used to identify the moisture transport, which is different from the HYSPLIT model in that the water loadings in the air are not well considered. Here, we calculated precipitable water and water vapor stream from the surface to the 300 hPa isobaric level during the four seasons during 2011–2014 based on the global reanalysis (Figure 6, Figure 7, Figure 8, and Figure 9). Although the water vapor stream presented generally similar patterns—i.e., from the west to the east—the precipitable water amount showed spatial incoherence for each season. Different from other seasons, the monsoon moisture could be greatly delivered to the study region in summer, which can be easily seen in Figure 8. The westerlies mass is associated to the high values of $d$
in winter. The seasonal characteristics of water vapor stream are related to the seasonal variations of stable isotope compositions in precipitation.

**Figure 6.** Water vapor stream and precipitable water amount (PW) surrounding the study region in January for different years.

**Figure 7.** Water vapor stream and precipitable water amount (PW) surrounding the study region in April for different years.
Figure 8. Water vapor stream and precipitable water amount ($PW$) surrounding the study region in July for different years.

Figure 9. Water vapor stream and precipitable water amount ($PW$) surrounding the study region in October for different years.
We also examined the water vapor streamline for the four events mentioned in Section 3.2 (Figure 10). Compared to Figure 5 showing the backward trajectories, the moisture diagnosed moisture transport in Figure 10 generally exhibited similar but slightly different information. For example, in August 3rd, 2014 (Figures 5d and 10d), the two methods exhibited a very similar pattern that moisture is delivered from the southeastern direction; however, in other cases—in Figures 5 and 10—the dominant directions were not always the same, indicating the air mass transport cannot always be treated as moisture transport.

Figure 10. Water vapor stream and precipitable water amount (PW) surrounding the study region during four typical rainfall events.

4. Conclusions

The spatial incoherence and seasonal variability in $d$ value in precipitation provide a useful method to understand regional moisture sources. Here, we presented a case study at the monsoon margin, and investigated the $d$ value in precipitation as well as the possible linkage to water vapor sources. In the study region, the regional mean $d$ value is 11.0‰ in the dry season and 8.0‰ in the wet season, respectively. The $d$ value in precipitation usually presents a negative correlation with local air temperature and vapor pressure. The low $d$ value during the wet season generally coincides with the high values in air temperature and water vapor pressure, and is influenced by the monsoon moisture transported from long distances. During the dry season, the continental air masses lead to a relatively dry background and higher $d$ value in precipitation, which is also consistent with the strong below-cloud evaporation under dry conditions. The Lagrangian method was applied to diagnose the air backward trajectory, and the water vapor flow was used to identify the moisture transport. The moisture regimes based on reanalysis data are generally consistent with the findings using the stable isotopic approach in the study, and the monsoon moisture is highlighted in summer precipitation at these monsoon margin sites.
Supplementary Materials: The following are available online at http://www.mdpi.com/2073-4441/12/12/3315/s1, Figure S1: Variation of monthly $d$ value at the GNIP Lanzhou station. Figure S2: Correlations between $d$ value and air temperature (a) or water vapor pressure (b) at the GNIP Lanzhou station. Figure S3: Clustered backward trajectories during different seasons to the Lanzhou city in 2011. Figure S4: Clustered backward trajectories during different seasons to the Lanzhou city in 2012. Figure S5: Clustered backward trajectories during different seasons to the Lanzhou city in 2013. Figure S6: Clustered backward trajectories during different seasons to the Lanzhou city in 2014.

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