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VARIATIONS OF THE STABLE ISOTOPIC COMPOSITION OF PRECIPITATION AND CAVE DRIP WATER AT ZHENZHU CAVE, NORTH CHINA: A TWO-YEAR MONITORING STUDY

Yunxia Li[1, 2, 3], Shengrui Zhang[4], Xiaokang Liu[1, 5], Yongli Gao[2], and Zhiguo Rao[3, C]

Abstract

We investigated potential factors influencing variations in isotopic composition of external precipitation and internal drip water at Zhenzhu Cave, Hebei Province, north China. Two meteoric precipitation sites and three cave drip sites were monitored monthly during April 2012–April 2014. The results of meteoric precipitation present: on monthly timescale, $\delta^{18}O_p$ and δD_p showed a significant seasonal double peak characterized by depleted isotopic values in both dry and wet seasons (but a significant seasonal variation in *d*-excess). There is a weakly negative correlation with monthly mean precipitation amount but no significant correlation with surface air temperature. Based on GNIP data, there are no significant correlations on an interannual scale between annual weighted mean $\delta^{18}O_p$ (and δD_p) and annual precipitation or annual mean temperature. This implies the variation of $\delta^{18}O_p$ (and δD_p) in the area cannot be interpreted solely as reflecting an "amount effect" or "temperature effect" on either seasonal or interannual scale, and may be affected by moisture sources. There isn't any significant change in $\delta^{18}O_d$ and δD_d values of cave drip water during monitoring period, demonstrating that cave drip water comprise a mixture of precipitation integrated over a long period rather than seasonal timescale. There were significant seasonal changes in cave air CO_2 concentration, with higher concentrations in summer and lower concentrations in winter that is well correlated with environmental variables such as surface temperature and precipitation, which may help improve the interpretation of stable carbon isotope records from speleothems ($\delta^{13}C_s$) in relation to cave air CO_2.

Introduction

Owing to the availability of various environmental proxies and precise chronology, speleothems are widely recognized as one of the most important archives of information for high-resolution paleoclimatic studies (Fairchild et al., 2006; Fairchild and Baker, 2012). In particular, the oxygen isotope and carbon isotope ratios of speleothems ($\delta^{18}O_s$, $\delta^{13}C_s$) have been widely used to reconstruct past climatic variability (Wang et al., 2001, 2008; Cosford et al., 2009; Cheng et al., 2009). $\delta^{18}O_s$ is often considered to inherit the $\delta^{18}O$ signal of atmospheric precipitation ($\delta^{18}O_p$) (Cheng et al., 2005; Luo and Wang, 2008), and thus, climatic factors effecting changes in $\delta^{18}O_p$ are usually regarded as drivers of variations in $\delta^{18}O_s$ (Wang et al., 2001; Dayem et al., 2010; Cheng et al., 2012; Tan, 2009, 2014). However, $\delta^{18}O_p$ is a function of air temperature, rainfall amount, moisture source, and other complex factors (Craig, 1961; Rozanski et al., 1982). Consequently, it is often difficult to separate the effects of these variables for the climatic interpretation of $\delta^{18}O_p$ records, although research has indicated that changes in $\delta^{18}O_p$ are mainly controlled by rainfall amount in low latitude regions and by temperature in high latitude regions (Dansgaard, 1964). Furthermore, $\delta^{18}O_s$ is not controlled solely by changes in $\delta^{18}O_p$. Cave conditions such as host rock lithology, ventilation, CO_2 concentration, and the in-cave process of calcite precipitation could also influence variations in $\delta^{18}O_s$ (Feng et al., 2012, 2014; Genty et al., 2014; Duan et al., 2016). Consequently, the interpretation of $\delta^{18}O_s$ is ambiguous, especially in the case of $\delta^{18}O_s$ records from the Asian monsoon region which are often interpreted as a proxy of local rainfall amount or monsoon intensity (Clemens et al., 2010; Pausata et al., 2011; Tan, 2009, 2014; Liu et al., 2015; Rao et al., 2015, 2016).

From the foregoing, it is evident that cave monitoring can help improve our understanding of the relationship between $\delta^{18}O_p$ and $\delta^{18}O_s$ (Luo and Wang, 2008; Ruan and Hu, 2010; Li et al., 2011; Genty et al., 2014; Duan et al., 2016). In the past decade, cave monitoring research has yielded valuable information (Luo and Wang, 2008; Ruan and Hu, 2010; Li et al., 2011; Genty et al., 2014; Duan et al., 2016), but monitoring results may be difficult to interpret and vary among different caves. Moreover, in some cases even results for different sites within the same cave are inconsistent (Duan et al., 2016). Therefore, more cave monitoring studies of longer duration are needed to clarify the significance of the $\delta^{18}O$ record of speleothems.

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The present study seeks to provide a reference for reconstructing past environmental changes in the Zhenzhu Cave region in north China and to better understand the cave environment. For this purpose, we conducted a two-year (April 2012–April 2014) on-site monitoring study of precipitation and drip water with a monthly sampling interval.

Study area and sampling sites

Zhenzhu ('Pearl' in English) Cave (38º15′ N, 113º42′ E, 990 m) is in the Tiangui Mountains in Pingshan County, Hebei Province, north China (Fig. 1). The current climate in the study area is dominantly monsoonal, characterized by two distinct seasons: a cool and dry winter from November to February and a hot and rainy summer from June to September. According to the meteorological data covering 1981-2010 from the nearest weather station (Pingshan city, National Meteorological Information Center, http://data.cma.cn/data/weatherBk.html), the mean annual temperature (MAT) is ∼13 °C, and the mean annual precipitation (MAP) is ∼539 mm, with ∼80 % of the annual precipitation occurring in summer (June to September). However, because Zhenzhu Cave is located in the high elevation mountainous area, the MAT in this cave area is ∼2-3 °C lower than that in Pingshan city, but the MAP in this cave area is ∼110 mm more than that in Pingshan city, with a final MAT of 8-11 °C and MAP of 649-690 mm (Chen, 2010). The bedrock of Zhenzhu Cave is upper Cambrian-lower Ordovician dolomite, lime-dolomite, and dolomitic limestone. The cave was formed along a NNW-SSE oriented fault zone. The thickness of the bedrock overlying the cave is very thin (∼10 m). At present, the natural vegetation above the cave is well-developed (Wang et al., 2011). According to the results of monitoring covering April 2012–April 2014, the thermohydrograph (hourly average output) showed that the relative humidity inside Zhenzhu Cave maintains 99%∼100% during almost the whole monitoring period. The average temperature inside the cave is 9 °C, with a standard deviation of 1 °C, consistent with annual air temperature in this area.

Two precipitation monitoring stations (W1 and W2) were established for this study. W1 was located on the rooftop of a water house at a scenic location near Zhenzhu Cave and W2 was located on the rooftop of the administration building near site W1. The linear distance between W1 and W2 is 600 m (Fig. 1B). Three drip monitoring sites (D1, D2, D3) were chosen from different localities along cave passages, with increasing distance from the cave entrance to the innermost part of the cave (Fig. 1C). Cave conditions, including cave air CO$_2$ concentration (CO$_2$ meter), cave air temperature (T_{in}) and inside cave relative humidity (RH) (Thermohydrograph), were also monitored (Fig. 1C).

Methods And Data

Two precipitation-monitoring stations and three drip sites were visited every month from April 2012–April 2014 and corresponding water samples were collected. Drip water samples were collected in 100 mL high-density polyethylene (HDPE) bottles and stored at ∼4 °C until processing. Storage and processing of precipitation samples were the same as those for drip water. All samples were filtered using a 0.45 μm glass fiber membrane filter and stable isotopic composition of oxygen and hydrogen were measured using a liquid water isotope analyzer (Los Gatos Research, DLT-100) in the Key Laboratory of Western China's Environmental Systems (Ministry of Education), College of Earth and Environmental Sciences, Lanzhou University. Results are expressed relative to V-SMOW (Vienna Standard Mean Ocean

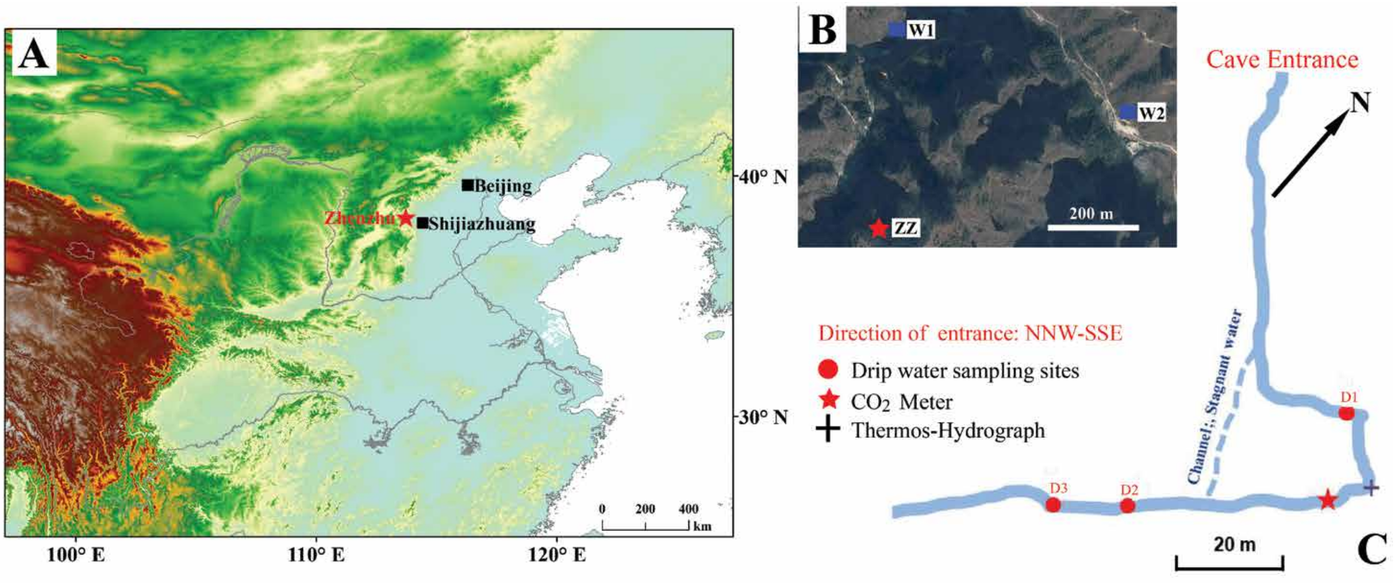

Figure 1. (A) Location of Zhenzhu Cave (red star) in Pingshan County, Hebei Province, China. The locations of Beijing and Shijiazhuang are also shown; (B) Location of rainfall sampling sites (blue rectangles, W1: 38.2643°N, 113.7149°E; W2: 38.2621°N, 113.7231°E), (Google Maps Image); (C) Plan view of the passages of Zhenzhu Cave, showing the locations of dripwater sampling sites (red circles), CO$_2$ meter (red star), and thermohydrograph (purple cross).

Water). Measurement errors of hydrogen isotope (δD) and oxygen isotope ($\delta^{18}O$) measurements were 0.3 ‰ and 0.1 ‰, respectively. Because meteorological data of a specific month in Pingshan city isn't available in the National Meteorological Information Center, meteorological data (monthly temperature and monthly precipitation) of Shijiazhuang (80 km away from Zhenzhu Cave) is downloaded from NOAA (https://gis.ncdc.noaa.gov/maps/ncei/summaries/monthly) to represent the local climate conditions.

Results and Discussion

Stable isotopic composition of precipitation

Seasonal variations

The hydrogen (δD_p) and oxygen ($\delta^{18}O_p$) isotopic compositions of precipitation samples from sites W1 and W2 during April 2012–April 2014 range from −84.4 ‰ to −16.9 ‰, and from −11.7 ‰ to −4.0 ‰, respectively. Because of the absence or limited amount of rainfall in some months, δD_p and $\delta^{18}O_p$ data for March 2013, December 2013, January 2014 and March 2014 are not available (Supplementary 1). In principle, precipitation isotopic values of sites W1 and W2 should be the same or similar. However, there are minor differences in the values in December 2012, January 2013, February 2013 and February 2014. Evidently, this difference occurred in months in which precipitation amount and temperature were very low, and thus water samples may have been affected by environmental factors such as evaporation and freezing. Minor variations in sampling and storage procedures during sample collection, as well as measurement errors, may also have been responsible for the differences. Nevertheless, precipitation isotopic values of the two sites were generally consistent during the monitoring period, suggesting that the data are valid.

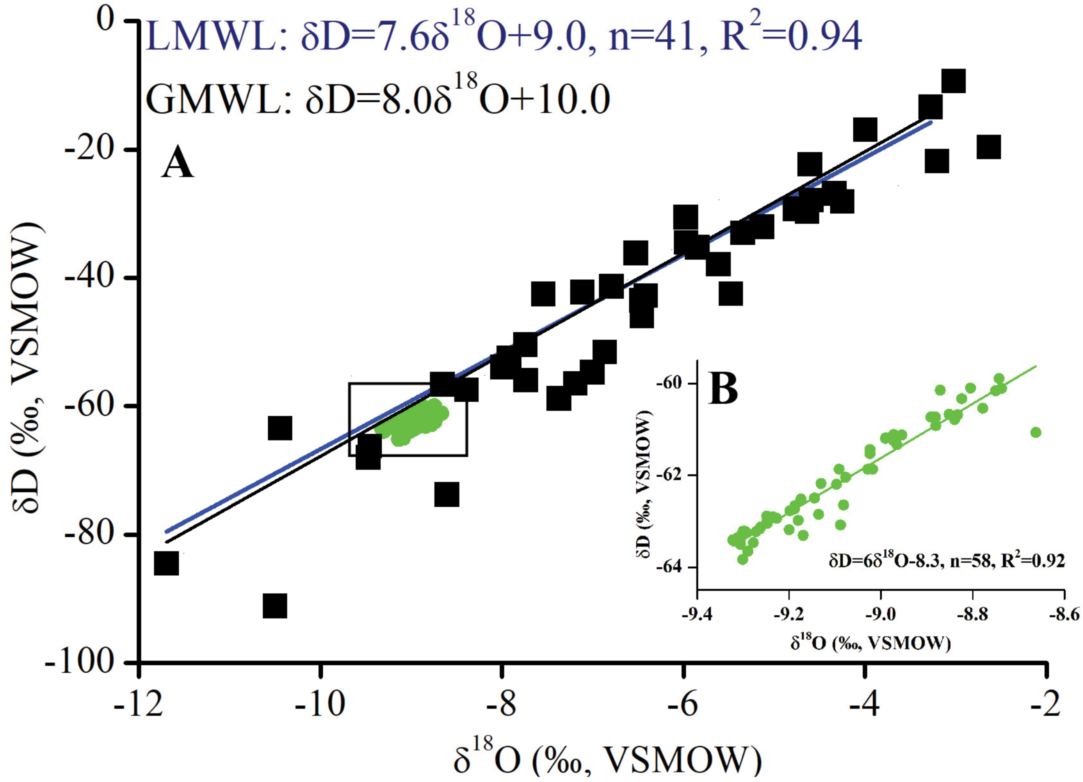

Figure 2. (A) Relationship between δD and $\delta^{18}O$ of drip water and local precipitation in Zhenzhu Cave. The Local meteoric water line (LMWL, blue line) is defined by all the precipitation data from two precipitation monitoring sites (W1 and W2). Within the analytical uncertainty, the Zhenzhu Cave drip-water samples fall on the Global meteoric water line (GMWL, black line) and below the LMWL; (B) Relationship between δD and $\delta^{18}O$ of drip water (grey box).

Precipitation isotopic data was used to construct the local meteoric water line in the Zhenzhu Cave region (LMWL): $\delta D = 7.6\ \delta^{18}O + 9.0$ ($n = 41$, $R^2 = 0.94$) (Fig. 2A). The slope of the LMWL is close to but slightly smaller than that of the global meteoric water line (GMWL: $\delta D = 8\ \delta^{18}O + 10.0$, Craig, 1961) and the Chinese meteoric water line (CMWL: $\delta D = 7.9\delta^{18}O + 8.2$, Zheng et al., 1983), implying that isotopic composition of precipitation in the area was weakly affected by unbalanced secondary evaporation and other fractionation factors, such as considerable seasonal temperature variations and relatively low condensation temperature (Liu et al., 2014).

A seasonal double-peak is evident in the data. δD_p and $\delta^{18}O_p$ were lower in the summer/rainy seasons and cold/dry seasons (Fig. 3A and 3B). This result differs from isotopic compositions of precipitation in South China, which are lower only in the summer/rainy season, but are similar to those in northeast China (Liu et al., 2008, 2009, 2014). Correlation analysis of the monthly mean $\delta^{18}O_p$ (and δD_p) and precipitation amount or temperature revealed a relatively weak negative correlation between isotopic composition ($\delta^{18}O_p$ and δD_p) and monthly precipitation (MP) (W1: $\delta^{18}O_p = -0.014P - 6.0$,

$n = 21$, $R^2 = 0.19$; $\delta D_p = -0.14P-34.4$, $n = 21$, $R^2 = 0.26$; W2: $\delta^{18}O_p = -0.011P-6.5$, $n = 20$, $R^2 = 0.11$; $\delta D_p = -0.12P-37.9$, $n = 20$, $R^2 = 0.2$) (Fig. 4). This demonstrates that $\delta^{18}O_p$ and δD_p in the Zhenzhu Cave area were either uninfluenced or only slightly influenced by the rainfall amount effect on a seasonal scale. There was no significant correlation between isotopic composition and monthly mean air temperature (W1: $\delta^{18}O_p = -0.011T-6.6$, $n = 21$, $R^2 = 0.003$; $\delta D_p = -0.22T-39.4$, $n = 21$, $R^2 = 0.019$; W2: $\delta^{18}O_p = 0.010T-7.3$, $n = 20$, $R^2 = 0.003$; $\delta D_p = -0.16T-43.4$, $n = 20$, $R^2 = 0.006$) (Fig. 4). Therefore, isotopic compositions of precipitation on a seasonal scale in the study area cannot be interpreted solely in terms of rainfall amount or temperature effect.

Changes in moisture source and vapor transport path have substantial effects on variations in isotopic compositions of precipitation (Welker, 2000; Liu et al., 2008, 2009, 2014). In studies of modern isotopic compositions of precipitation, deuterium excess (*d*-excess), defined as $d = \delta D - 8\delta^{18}O$, is an important parameter that may reflect climatic conditions in the moisture source area such as the distance from the source area to the precipitation location, and condensation and re-evaporation processes during vapor transport (Araguás-Araguás et al., 2000; Merlivat and Jouzel, 1979; Johnsen et al., 1989; Pfahl and Wernli, 2008; Pfahl and Sodemann, 2014).

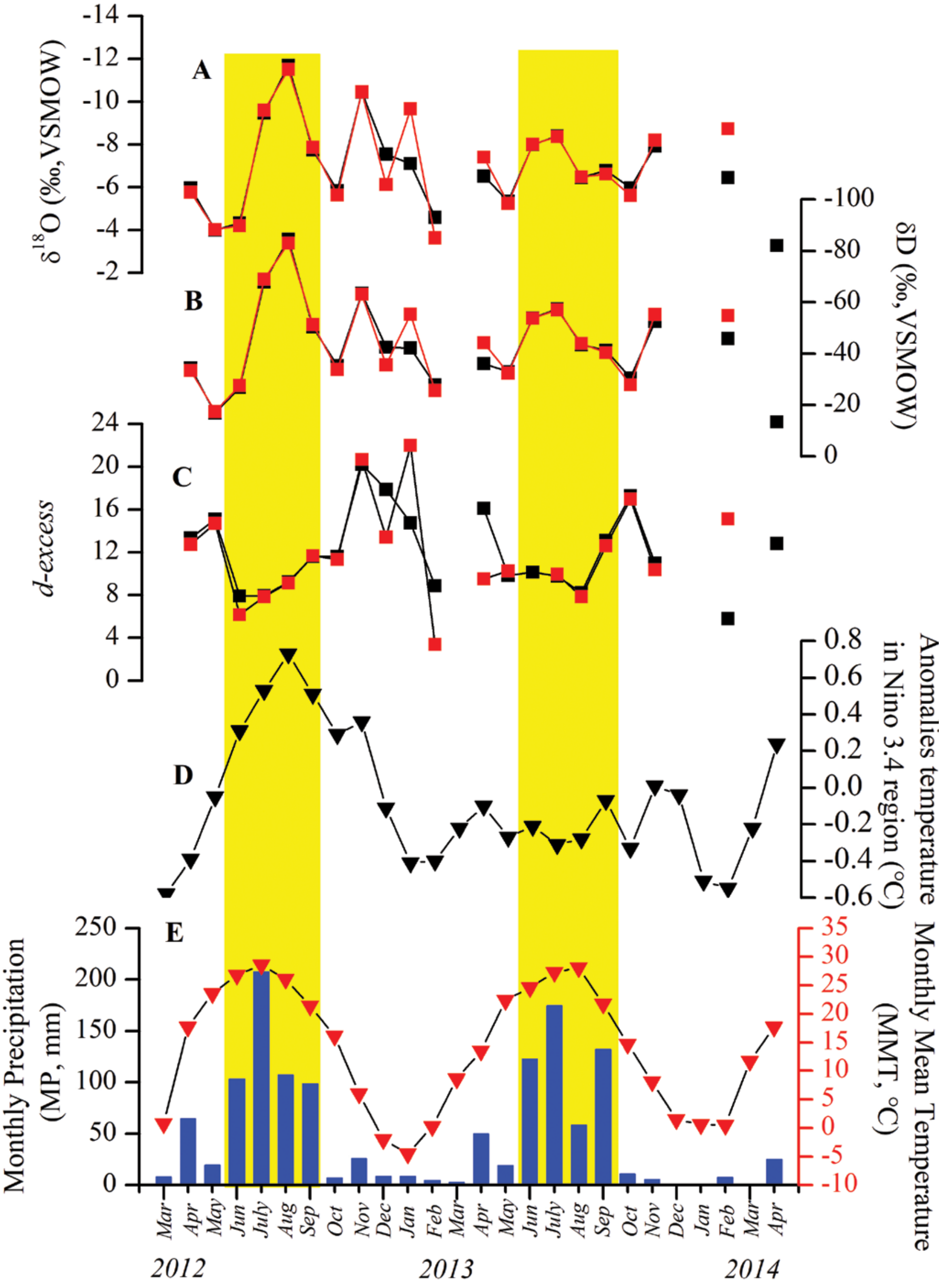

Figure 3. Time series of $\delta^{18}O$ (A), δD (B) and *d*-excess (C) from the W1 (black squares) and W2 (red squares) rainfall sampling sites in the Zhenzhu Cave area; (D) Temperature anomalies in the Niño 3.4 region (black triangles); (E) Variation of monthly precipitation (blue bars) and monthly mean temperature (red triangles) (the yellow bar represents the summer period).

We calculated *d*-excess during the monitoring period, using the monthly averaged $\delta^{18}O_p$ and δD_p data (Fig. 3C). Compared to the double-peak signal in the record of the isotopic composition of precipitation, a significant seasonal variation is evident, with higher values in winter and lower values in summer. For the Zhenzhu Cave area, which is very sensitive to changes in the summer and winter monsoon, the wind direction in summer is from the ocean towards the interior land, which is determined by the land-sea thermal gradient, or the local inland atmospheric circulation pattern. The temperature and relative humidity over the ocean result in a lower *d*-excess value than that of air masses originating from inland (Pfahl and Sodemann, 2014). By contrast, air masses transported by winter winds originate in high latitude regions, characterized by dry conditions and strong evaporation in the source area and by a long transport distance, resulting in a higher value of *d*-excess. In addition, based on NOAA/ARL HYSPLIT back trajectory modelling

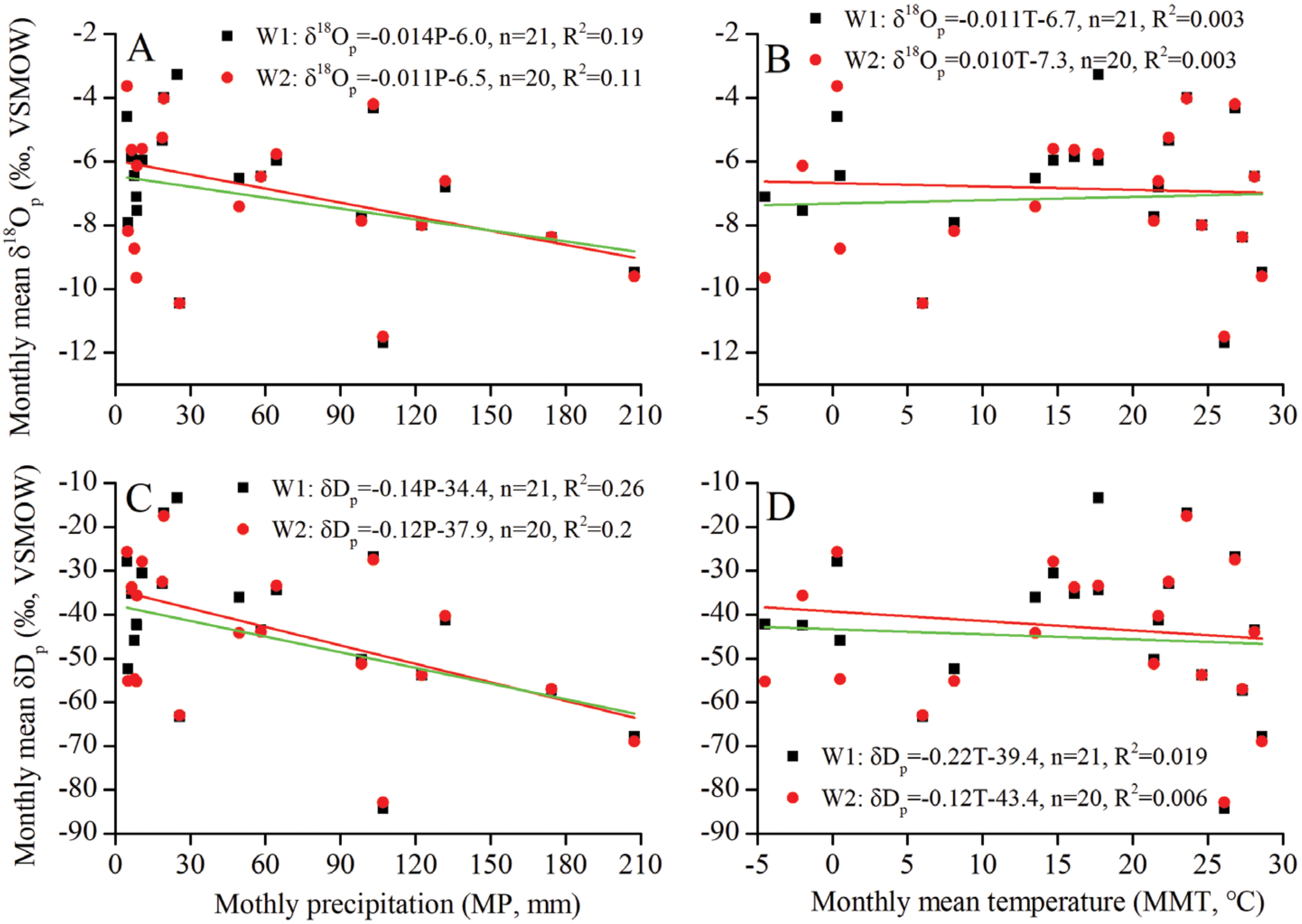

Figure 4. Relationship between monthly precipitation (MP, A) or monthly mean temperature (MMT, B) and monthly mean $\delta^{18}O_p$ for the Zhenzhu Cave area; C and D are the same as A and B, but for monthly mean δD_p.

(https://www.ready.noaa.gov/HYSPLIT_traj.php, Zhang et al., 2015), the air mass trajectory of a typical summer month (July 2013) and a typical winter month (January 2013) was determined weekly. Results demonstrated that the air mass trajectory in July 2013 was complex and mainly sourced from local inland circulation (Fig. 5). However, the air mass in January 2013 was mainly transported by northerly or westerly winds from high latitudes or by westerly winds far inland (Fig. 6), which is consistent with our *d*-excess data and results from previous research (Wang et al., 2003). On the other hand, this characteristic pattern of variation of *d*-excess is also in accord with the double-peak evident in the isotopic composition of precipitation mentioned above (Pfahl and Sodemann, 2014). Precipitation in the rainy season in the Zhenzhu Cave area is derived from local recycled vapor or oceanic sources, with lower values of precipitation isotopic compositions ($\delta^{18}O_p$ and δD_p) resulting from the rain-out effect. For the winter/dry season, precipitation is mainly sourced from remote inland air masses or polar air masses, with a long transport distance and evaporation, which finally result in lower isotopic values of precipitation.

Interannual variations

On an interannual scale, taking W1 for example, the amplitude of variation of $\delta^{18}O_p$, δD_p during April 2012 to February 2013 ($\delta^{18}O_p$: −11.7 ‰ to −4.0 ‰; δD_p: −84.4 ‰ to −16.9 ‰) was greater than that during the same period from 2013-2014 ($\delta^{18}O_p$: −8.4 ‰ to −5.4 ‰; δD_p: −57.4 ‰ to −30.5 ‰). In addition, the mean value of $\delta^{18}O_p$ (δD_p) weighted by monthly precipitation amount (MWP) for April 2012–February 2013 was −8.2 ‰ (−55.4 ‰), −8.6 ‰ (−60.0 ‰) for June 2012–September 2012 (warm/rainy season), and −8.4 ‰ (−50.5 ‰) for October 2012–February 2013 (cold/dry season). These values are lower than those for the same period in 2013–2014 (Table 1). On the other hand, meteorological data show that the total amount of precipitation during April 2012 to February 2013 (653.5 mm) was greater than that during April 2013–February 2014 (578.5 mm) (Table 1). Combining the corresponding MWP isotopic values for the aforementioned periods (Table 1), it suggests that the greater the rainfall amount, the more depleted the isotopic composition (i.e., the rainfall amount effect). This conclusion is tentative due to the relatively short monitoring period, however.

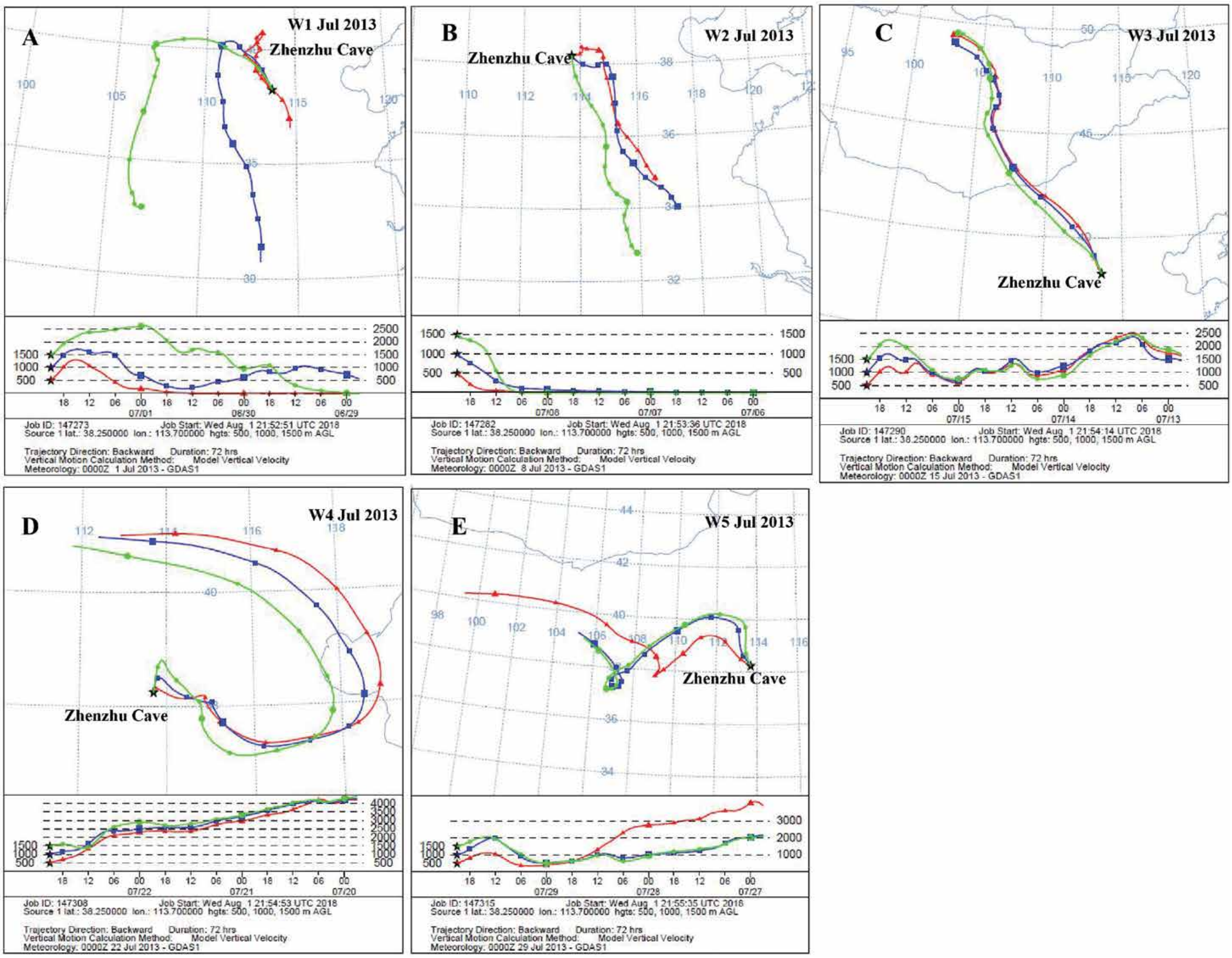

Figure 5. Weekly trajectory modeling of the air mass path in a typical summer month (July 2013) based on NOAA/ARL HYSPLIT.

To examine the relationships between $\delta^{18}O_p$ (and δD_p) and environmental variables (rainfall amount and temperature) on an interannual scale, due to the lack of monitoring data covering a longer interval, we referred to results of previous research (Li et al., 2015; Rao et al., 2016). Based on GNIP data from Shijiazhuang meteorological station, weighted mean annual $\delta^{18}O_p$ (and δD_p) spanning a 13-year interval was used to analyze the relationship between weighted mean annual $\delta^{18}O_p$ (and δD_p) and annual precipitation amount or annual mean temperature (Fig. 7). Results show that correlations between weighted mean annual $\delta^{18}O_p$ (and δD_p) and annual precipitation amount or annual mean temperature are weak (Fig. 7), and thus we cannot attribute the variation in $\delta^{18}O_p$ to a rainfall amount effect or temperature effect on interannual scales.

It has been suggested that the El Niño-Southern Oscillation (ENSO) cycle is the dominant control on the interannual variation in $\delta^{18}O_p$ in the monsoon regions of China, via its effect on the ratio of water vapor originating from distant oceanic sources (progressively depleted in $\delta^{18}O$) and local oceanic sources (relatively enriched in $\delta^{18}O$) at an observation site (Tan, 2014). Hence, we compared our precipitation isotopic values with an ENSO index, defined by the sea-surface temperature anomaly (SSTA) in the Niño 3.4 region (5°S − 5°N, 170°W − 120°W) in the tropical Pacific (NOAA, https://www.cpc.ncep.noaa.gov/data/indices/sstoi.indices). The results demonstrate that the pattern of variation of SSTA in the Niño 3.4 region is similar to that of the isotopic composition in precipitation, with the amplitude of variation in 2012-2013 greater than that during 2013–2014 (Fig. 3D), implying that the variation of the isotopic composition of precipitation during the monitoring period may be affected by the SSTA or by an atmosphere-ocean circulation anomaly, which can effect changes in moisture source or transport path, causing variations in the isotopic composition of precipitation. However, more research is needed to confirm this conclusion.

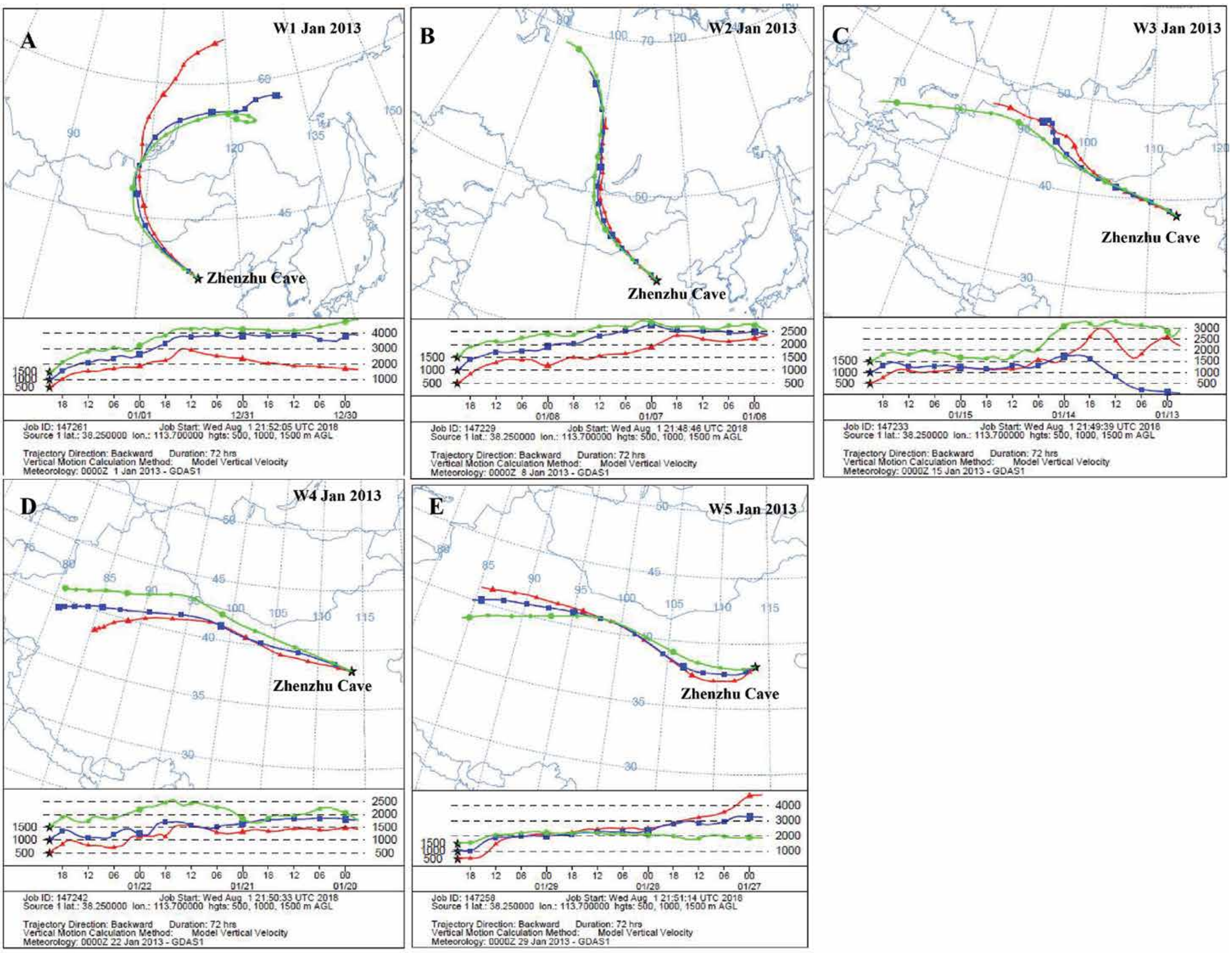

Figure 6. Weekly trajectory modeling of the air mass path in a typical winter month (January 2013) based on NOAA/ARL HYSPLIT.

Table 1. Summary of average temperature, total precipitation, mean $\delta^{18}O_p$ and δD_p, weighted by monthly precipitation amount (MWP), and average $\delta^{18}O_d$ and δD_d for specific periods for the Zhenzhu Cave area.

Projects for Statistics	April 2012–April 2014	April 2012–February 2013	June 2012–September 2012	October 2012–February 2013	April 2013–February 2014	June 2013–September 2013	October 2013–February 2014
Average temperature (°C)	14.4	14.55	25.7	3.2	14.8	25.4	5.1
Total precipitation (mm)	1260	653.5	515.9	53.8	578.5	486.9	23.3
MWP $\delta^{18}O_p$ (‰, VSMOW)	−7.7	−8.2	−8.6	−8.4	−7.3	−7.6	−6.9
MWP δD_p (‰, VSMOW)	−51.1	−55.4	−60.0	−50.5	−48.5	−50.3	−41.4
Average $\delta^{18}O_d$ (‰, VSMOW)	−9.1	−9.0	−9.0	−9.0	−9.1	−9.1	−9.1
Average δD_d (‰, VSMOW)	−62.1	−61.6	−61.5	−61.9	−62.4	−62.6	−61.3

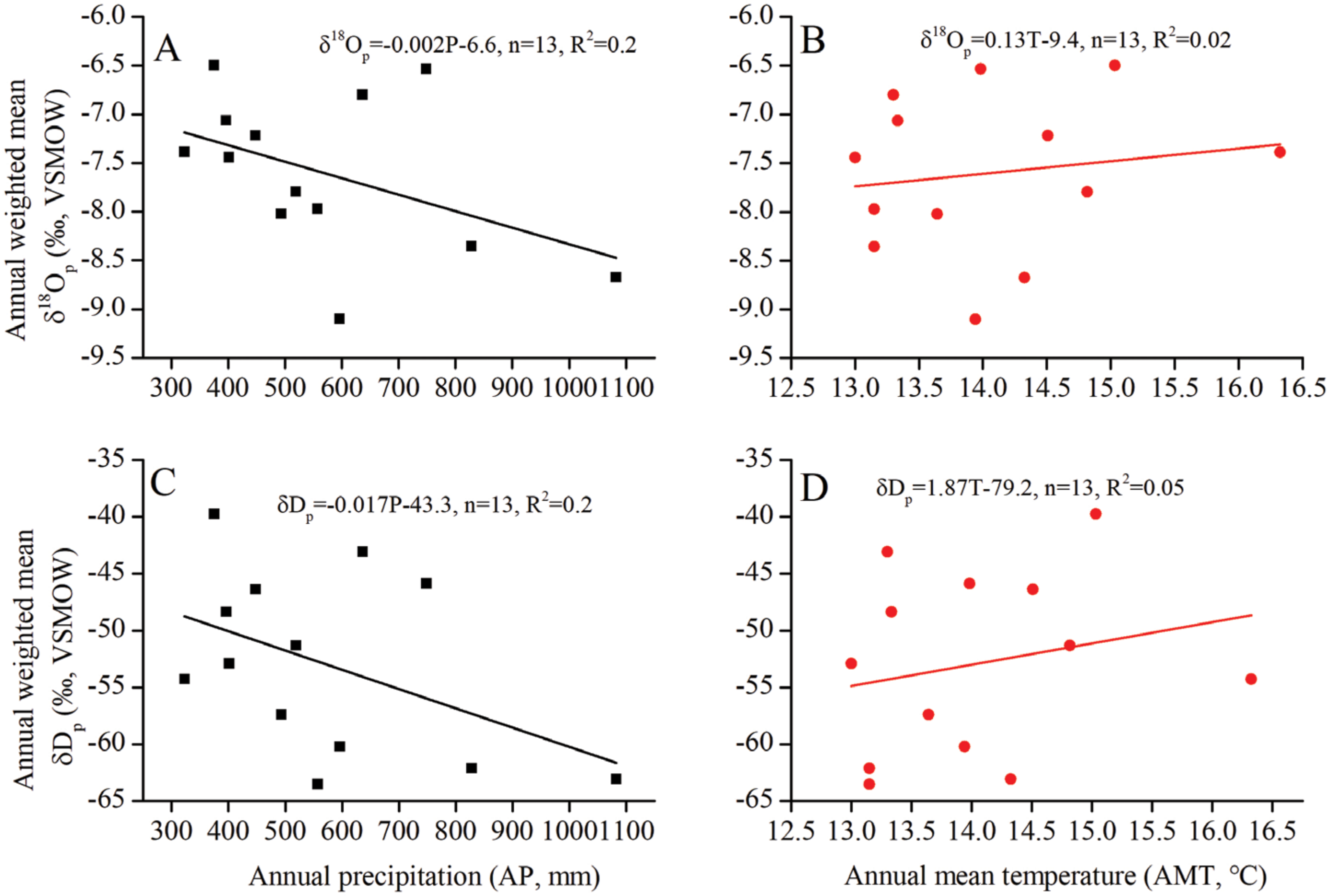

Figure 7. Relationships between annual weighted mean precipitation δ18O and annual precipitation (A) or annual mean temperature (B) at Shijiazhuang.

In summary, the variation of the precipitation isotopic composition in the Zhenzhu Cave area during the monitoring period cannot be attributed solely to the rainfall amount effect or temperature effect, on either seasonal or interannual timescales.

Stable isotopic composition of drip water

Three drip monitoring sites (D1, D2 and D3) were sampled monthly. Drip rate were measured at the time of sample collection from April 2012–April 2014. All three sites had perennial drips, but the drip rates of all sites were very low (>30 minutes per drip).

The range of variation of the hydrogen (δD_d) and oxygen ($\delta^{18}O_d$) isotopic composition of drip water samples from the three sites was as follows. D1: δD_d: −62.9 ‰ to −60.1 ‰, and $\delta^{18}O_d$: −9.1 ‰ to −8.7 ‰; D2: δD_d: −63.7 ‰ to −60.1 ‰, and $\delta^{18}O_d$: −9.3 ‰ to −8.8 ‰; D3: δD_d: −63.8 ‰ to −59.9 ‰, and $\delta^{18}O_d$: −9.3 ‰ to −8.7 ‰. Isotopic compositions of drip water for some months are unavailable because of conditions such as limited drip water amount and evaporation during storage (Fig. 8, Supplementary 2).

Isotopic composition of the drip water samples generally plot close to or slightly below the LMWL (Fig. 2). In the meantime, the average value of $\delta^{18}O_d$ (δD_d) during the entire monitoring period is −9.1 ‰ (−62.1 ‰), which is close to or slightly more depleted than the $\delta^{18}O_p$ (δD_p) with values of −7.7 ‰ (−51.1 ‰) (Table 1), which indicates that in general $\delta^{18}O_d$ (δD_d) inherits the signal of the meteoric water above the caves (Luo and Wang, 2008; Pape et al., 2010; Genty et al., 2014). The drip water $\delta^{18}O_d$ (δD_d) plotting below the LMWL and the lower value of $\delta^{18}O_d$ (δD_d) may result from more depleted precipitation recharge of the drip water. Compared with the variation of the isotopic composition of precipitation (δD_p and $\delta^{18}O_p$), the $\delta^{18}O_d$ (δD_d) at the three sites had a low degree of variability (range of $\delta^{18}O_d < 1$ ‰), and a seasonal signal is absent (Fig. 8A), which is in accord with results in other cave sites (Genty et al., 2014; Duan et al., 2016). The pattern furthermore confirmed that the variation of $\delta^{18}O_d$ (δD_d) in drip water may be attributed to groundwater homogenization via the mixing of different timescales or multi-year timescales precipitation in the soil and epikarst zone above the cave (Williams and Fowler, 2002; Pape et al., 2010; Benton and Doctor, 2018; Eagle et al., 2015). The range

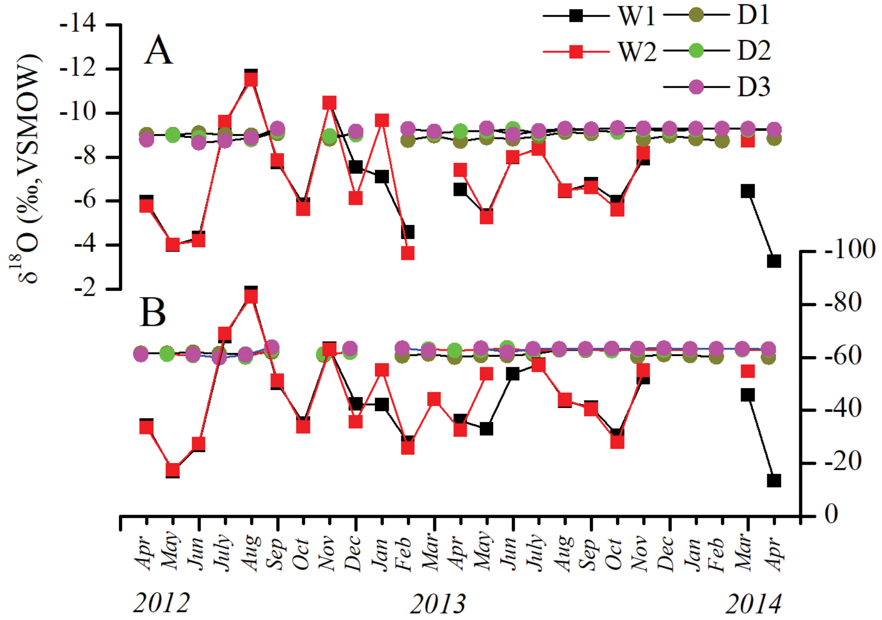

Figure 8. Time series of monthly water δ^{18}O (A), δ D (B) for two precipitation and three drip sites in Zhenzhu Cave

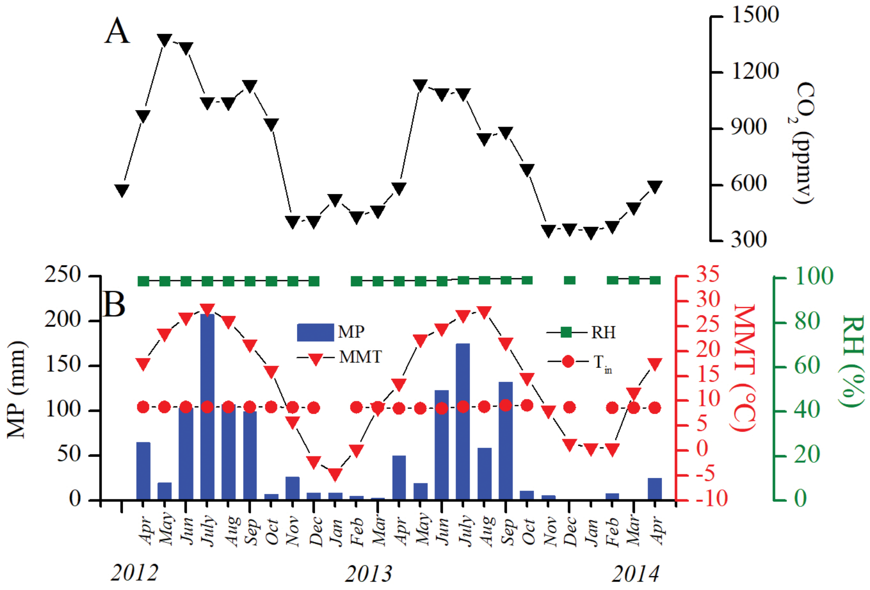

Figure 9. (A) Time series of cave air CO_2 concentration at Zhenzhu Cave; (B) variability of monthly precipitation (blue bars), monthly mean temperature (red triangles), and relative humidity (RH, green rectangle) inside the cave and cave air temperature (T_{in}, red dot).

of variation of the isotopic composition of the drip waters was small, implying that the resulting speleothem δ^{18}O ($\delta^{18}O_s$) could not reflect seasonal-scale variations in the isotopic composition of precipitation in the Zhenzhu Cave region.

Cave air CO_2 concentration, cave air temperature and relative humidity

In contrast to δD_d and $\delta^{18}O_d$, cave air CO_2 concentration exhibits a clear seasonal pattern of variation, ranging between 352 to 1383 ppmv, with lower values in the winter/dry season and higher values in the summer/wet season (Fig. 9). The average temperature inside the cave is 9 °C, with a standard deviation of 1 °C, consistent with annual air temperature in this area (Fig. 9). The RH in Zhenzhu Cave is 99 % to 100 % during the entire monitoring period. These characteristics of cave air CO_2 concentration, T_{in}, and the RH in Zhenzhu Cave were also observed in other studies (Hu et al., 2007; Li et al., 2011; Pu et al., 2015).

The variability of cave air CO_2 concentration is mainly affected by CO_2 sources: including the overlying soil, bacterial oxidation of organic matter in the cave system, outside atmosphere CO_2, and deep gas diffusion or transport (Liñán et al. 2008; Pu et al., 2015). However, contributions of other factors (e.g., the concentration of the outside atmospheric CO_2 and deep gas diffusion or transport) were minor, compared to the ~1000 ppmv range observed in the CO_2 level of Zhenzhu Cave. Thus, we conclude that the variation of cave air CO_2 concentration in Zhenzhu Cave was dominated by variations in CO_2 generated in the overlying soil, which were controlled by vegetation conditions and biological activity. In addition, biological productivity of vegetation and soil above a cave are influenced by climatic variables such as precipitation and temperature.

As shown in Fig. 9, seasonal variations of cave air CO_2 concentration in Zhenzhu Cave are highly consistent with the surface monthly mean temperature (MMT) and monthly precipitation (MP). There is also a significant positive correlation between CO_2 concentration and MP or MMT (Fig. 10). During the warm/wet season, when vegetation and soil biological activity are flourishing, the biogenic production of CO_2 would increase. As the outside air temperature increases, cave ventilation effects resulting from the barometric pressure (or density) difference between the outside air and cave air begin to decrease and the soil CO_2 yield begins to increase. In addition, an increasing amount of CO_2 from the overlying soil enters the cave with dripwater (or seepage water) that passes through the zone, maintaining the cave air CO_2 concentration at a higher level because of the continuous supply from the soil (Buecher 1999; Fernandez-Cortes et al. 2009). In the dry/cold season, when biological activity and the outside air temperature are lower, the soil CO_2 yield would decrease. Colder and denser air with lower CO_2 concentration enters the cave via the entrance and karst fissures, diluting the cave air CO_2 and leading to lower cave air CO_2 concentration (Li et al., 2012; Liñán et al.

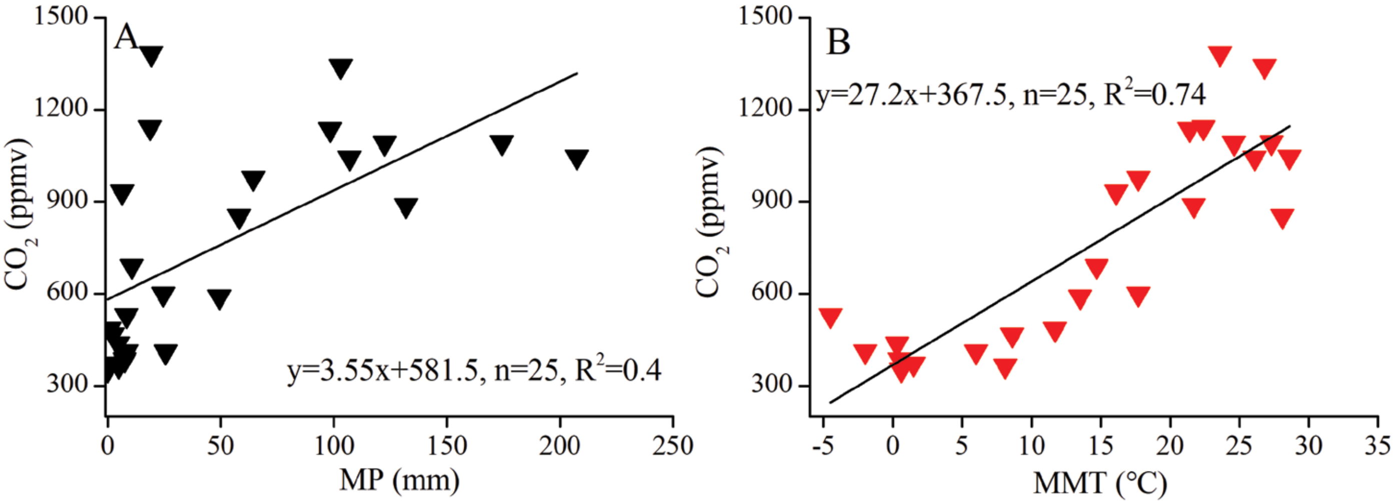

Figure 10. Linear regression between cave air CO_2 and MP (A) or MMT (B) for Zhenzhu Cave.

2008; 2015). However, statistical results including average temperature, total precipitation amount, and monthly mean CO_2 concentration during April–October of 2012 and 2013 (Table 2) show that the monthly mean CO_2 concentration during April–October of 2012 was greater than that during 2013, and was also the case for total precipitation, although the temperature was not significantly different between 2012 and 2013. The cave air CO_2 concentration may be more sensitive to precipitation variations. More sites for long-term monitoring are needed to verify these inferences.

In summary, climatic conditions (temperature and especially precipitation) outside the cave are likely responsible for the seasonal changes in cave air CO_2 concentration, which may control cave ventilation and biological activity in the vegetation and soil overlying the cave.

Table 2. Summary of average temperature, total precipitation, and monthly mean CO_2 concentration during April − October in 2012 and 2013 for the Zhenzhu Cave area.

Year	Average temperature (°C)	Total precipitation (mm)	Monthly mean CO_2 (ppmv)
2012	22.9	606.2	1122.6
2013	21.8	565.9	906.6

Implications of results for paleoclimatic studies

Our findings have several implications for the use of stable isotope values of speleothems from Zhenzhu Cave for paleoclimatic reconstruction. First, in the study area, there is no significant correlation between the stable isotopic composition of precipitation and rainfall amount or air temperature, on either seasonal or interannual scales. Therefore, variations in precipitation isotopic composition cannot be interpreted as a proxy of local rainfall amount or air temperature on these timescales. Second, because the relatively constant isotopic composition of drip water is caused by the mixing of meteoric waters in the epikarst zone above Zhenzhu Cave, the isotopic composition of drip water should reflect the long-term weighted average composition of local precipitation above the cave. Therefore, paleoclimatic records based on the oxygen isotopic composition of speleothems growing from such drip sites should reflect multi-year variations in the climate of the Zhenzhu Cave area.

Cave air CO_2 is closely related to speleothem $\delta^{13}C$ ($\delta^{13}C_s$), which is demonstrated by the following: $\delta^{13}C_s$ is mainly influenced by the $\delta^{13}C$ of soil CO_2 and CO_2 degassing of the drip water, which are also the dominant factors controlling cave air CO_2 concentration. Conversely, cave air CO_2 concentration plays an important role in the degassing of CO_2 and carbonate reprecipitation, which in turn affect variations in $\delta^{13}C_s$ (Paulsen et al., 2003; Fairchild et al., 2006, 2009; Spötl et al., 2005). Thus, the study of cave air CO_2 complements and reinforces our interpretation of $\delta^{13}C_s$ records. The cave air CO_2 level in Zhenzhu Cave during April 2012–April 2014 exhibited distinct seasonal changes and were well-correlated with outside climate conditions especially precipitation. Climatic conditions outside the cave substantially influenced the variations of cave air CO_2 concentration. Furthermore, the history of vegetation changes and deforestation based on the $\delta^{13}C_s$ record covering the past millenium from Zhenzhu Cave has been successfully reconstructed (Yin et al., 2017). CO_2 results with seasonal variation may be useful for interpreting the significance of $\delta^{13}C_s$ on seasonal timescale.

Conclusions

The major conclusions of our two-year monitoring study of Zhenzhu Cave in north China are

1. In the study area there was no significant correlation between $\delta^{18}O_p$ and δD_p and precipitation and temperature on the seasonal and interannual scales. Variations in $\delta^{18}O_p$ (δD_p) cannot be interpreted solely by the temperature effect or amount effect. They could be influenced by other factors such as changes in moisture sources caused by changes in atmospheric circulation.
2. The narrow range of variation and absence of seasonality in the drip-water $\delta^{18}O_d$ (δD_d) values suggest that the drip water is derived from the mixing of different timescales or multi-year meteoric waters in the epikarstic zone. Thus, the $\delta^{18}O_s$ records of speleothems fed by this type of drip water should reflect multi-year variations in the isotopic composition of the local precipitation.
3. The CO_2 concentration of Zhenzhu Cave exhibited significant seasonal changes and was well-correlated with climate conditions especially precipitation, implying seasonal variations in CO_2 concentration likely impact the signatures of $\delta^{13}C_s$, and possibly providing a seasonal signal that does not appear in $\delta^{18}O_s$ data.

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