

*Original Research*

# Characteristics of Hydrogen and Oxygen Stable Isotopes in Six Monsoon-Affected Cities in South China

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## Abstract

Hydrogen and oxygen stable isotopes are useful tracers for water cycle at local, regional and global scales. In monsoon climate zone, hydrogen and oxygen isotopes show different spatial distribution due to the influence of water vapor source, transport path and local meteorological factors. Based on the isotope data of precipitation and meteorological records in GNIP data set, this study investigated the temporal and spatial variation laws and influencing factors of hydrogen and oxygen isotopes in six cities in south China from 1987 to 1992 under the control of monsoon. The results show that the seasonal fluctuation of precipitation isotope in regions with single water source is slighter than those in regions with complex source. The synchronous seasonal variation of precipitation and temperature leads to the “temperature effect” of isotopes being masked. Water vapor source plays a leading role in isotope concentration, and its driving force for isotope change takes precedence over local meteorological factors. The local meteoric water line (LMWL) has significant differences among sites, and its slope is positively correlated with the percentage of urban-related land use, indicating that the change of isotopic fractionation coefficient caused by temperature and the contribution of recycled water vapor in urban areas may cause the characteristics of LMWL deviating from those in natural conditions.

**Keywords:** monsoon climate, water cycle, meteorological variables, moisture source, urban activities

## Introduction

Under the joint influence of global climate change and human activities, the water cycle pattern in urban environment is constantly changing, and the

hydrological conditions is becoming increasingly complex. The rapid population growth leads to the fast increase of industrial, agricultural and domestic water demand. The shortage of water resources is gr increasingly serious, which has become a major factor restricting social development. It is of great significance for understanding the evolution of regional water resources and promoting sustainable economic development to enhance the understanding of the

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distribution characteristics and change mechanism of urban water resources. As a natural tracer, hydrogen and oxygen stable isotope has stable chemical properties and sensitive response to environmental changes, which is widely used in hydrology, meteorology, ecology and paleoclimatology. In the study of water cycle, isotope tracer technology provides a good supplement to traditional hydrological methods, and plays an important role in tracing the source of precipitation water vapor, separating water components, evaluating the interaction between water bodies, estimating the response time of runoff and analyzing the water intake depth of plants [1-3]. The observation of isoscape characteristics in precipitation began in 1960s. Based on the measuring of  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  data in more than 400 different precipitation samples around the world, Craig [4] reported a significant positive correlation between hydrogen and oxygen isotopes, and established the classical global meteoric water line (GMWL) equation:  $\delta^2\text{H}=8\delta^{18}\text{O}+10$ . According to Rayleigh's Fractionation Model, for the isotopes in water, temperature is negatively correlated with the fractionation factor ( $\alpha$ ) from vapor to liquid and heavy isotopes is prone to enrich in the liquid water during the evaporation-condensation processes. Therefore, the spatio-temporal variation of stable isotopes in precipitation is dependent on the water sources and evaporation-condensation processes during the cloud formation and rainout, which can be influenced by both climatic and geographical parameters. On the basis of the observation of spatio-temporal variation and distribution characteristics of stable isotopes in precipitation, the influence of environmental conditions on isotope composition in precipitation was reported in many studies. Dansgaard [5] pointed out that the main factors affecting isotope change include land-and-sea position, elevation, latitude, temperature and precipitation through isotope observation of global precipitation. In the monsoon climate zone, the isotopic composition of precipitation is mainly affected by the transformation of water vapor sources [6, 7]. In some areas, the sub-cloud secondary evaporation and the contribution of recycled water vapor in the air mass has also been proved to be the factors affecting the composition of isotopes in local precipitation.

China is a country strongly influenced by monsoon climate, and its water resources distribution is uneven in both time and space, which is characterized by more in the south and less in the north and more in the east and less in west. In south China, the temporal and spatial characteristics of precipitation are closely related to the onset and decline of monsoon [8-10]. Under the background of climate change, extreme precipitation events driven by monsoon activities occurred frequently in recent years, leading to the risk of waterlogging in cities [8, 11, 12]. Therefore, understanding the spatial and temporal distribution of isotopes in precipitation and its influencing factors is of great significance to reveal the urban water cycle model. In the previous

studies, isotopic changes in precipitation at different time scales have been reported in various regions of south China [13-17]. However, most of these studies pay attention to the temporal trend of precipitation isotope in a single city, and only a few ones analyze the spatial variables affecting isotope characteristics based on the observation data from multi-sites [18, 19]. Based on the monthly monitoring data from GNIP database during the period 1987~1992, the stable isotope characteristics and local meteoric water lines (LMWLs) in six cities located in the monsoon region of South China were compared and analyzed. Combined with meteorological, monsoon activity and land use data, the dominant factors controlling the spatio-temporal differences of hydrogen and oxygen isotopes were revealed. The results of this study will help to understand the characteristics of regional water cycle in urban areas and provide new insights for analyzing the periodicity and driving factors of extreme hydrological events.

## Methods and Materials

### Data Source and Study Area

Global Network of Isotopes in Precipitation (GNIP) was established in 1960 by the cooperation of the International Atomic Energy Agency (IAEA) and the World Meteorological Organization (WMO). As the longest-lasting global isotope network, GNIP has established 1178 monthly monitoring sites and 202 daily ones in more than 100 countries and regions, providing data basis for research in global precipitation isotope and water cycle, and effectively promoting the hydrological survey and water resources planning and management. Although the GNIP dataset has played an important new role in the study of both hydrology and other subjects, the stations distribute very unevenly and the observation periods vary to a large extent. Therefore, performing a spatial comparison of isotopes at regional or national scale is usually hard to achieve in some places with GNIP stations sparsely distributed, for example, in China. For performing the spatial comparison in south China, some "old records" are selected in our study, which are collected within a same period and have a high accuracy. Based on the monthly isotope records from GNIP, the data of stable isotopes in precipitation and meteorological variables (including precipitation, temperature and water vapor pressure) in six Chinese cities of Hong Kong, Nanjing, Changsha, Guilin, Zunyi and Kunming from 1987 to 1992 were obtained. The selected sites have relatively continuous and sufficient data during the study period, and their distribution can represent the geographic sub-zones of South (Hong Kong, Guilin), Central South (Changsha), Southeast (Nanjing) and Southwest (Zunyi, Guilin) affected by monsoon in China. The detailed information of selected sites is described in Table 1. The location information of the sites is shown in Fig. 1. To analyze

Table 1. Detailed information of the selected cities in south China.

Site Name	Portal Number	Elevation(m)	Longitude (°E)	Latitude (°N)	Period
Hong Kong	450400	66	114.17	22.317	1987-1992
Nanjing	5823800	26	118.18	32.18	1987-1991
Changsha	5767900	37	113.067	28.2	1988-1992
Guilin	5795700	170	110.08	25.07	1987-1990
Zunyi	5771300	844	106.88	27.7	1987-1991
Kuming	5677800	1892	102.68	25.02	1987-1992

the influence of water cycle processes related to urban activities on hydrogen and oxygen isotopes, land use data generated from Landsat 8 remote sensing images were obtained from Resource and Environment Science

and Data Center, Chinese Academy of Science. The land use of “urban, industrial and mining” type was used to represent areas directly related to urban activities.

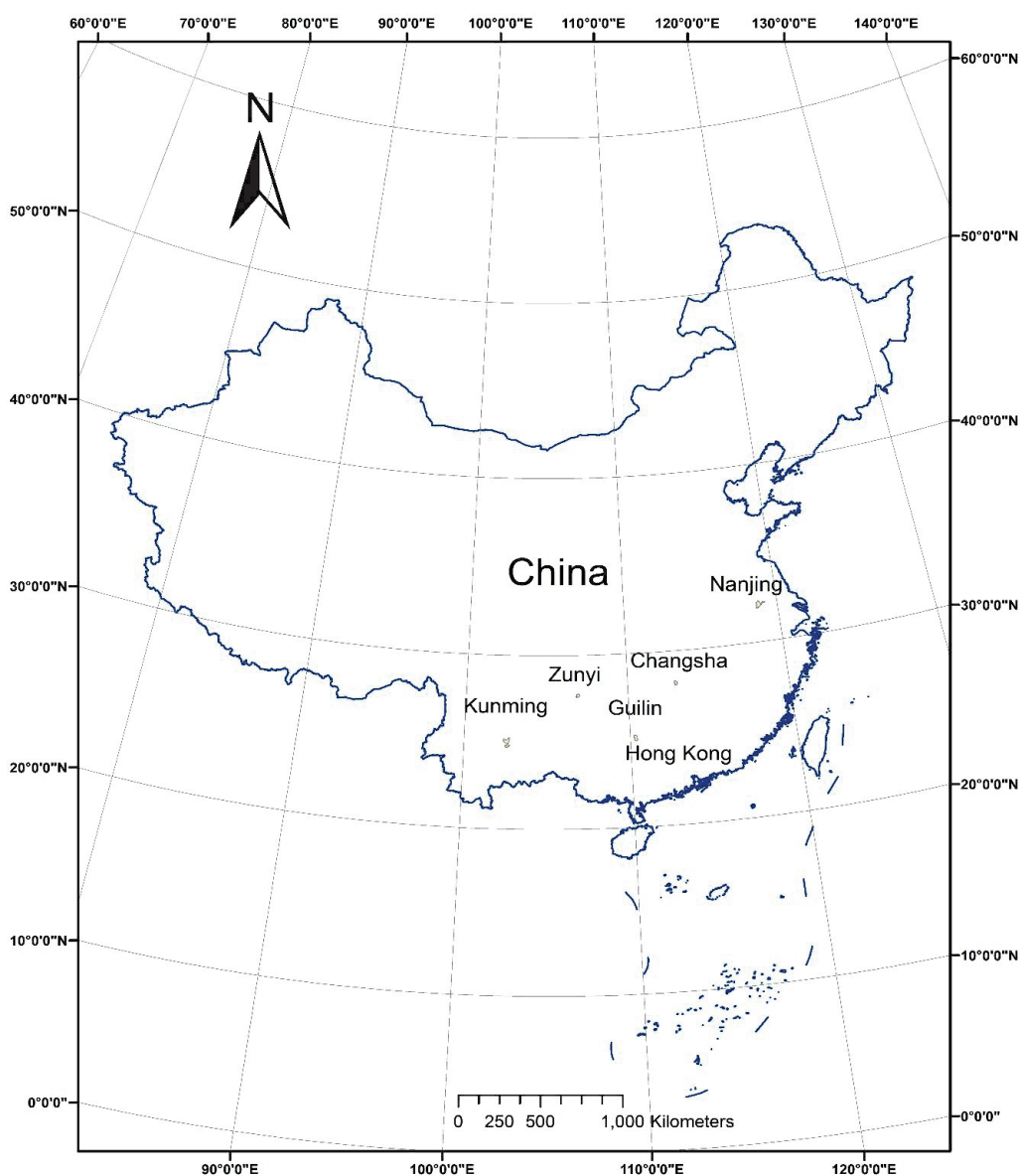


Fig. 1 Spatial distribution of the selected GNIP sites in south China.

## Analytical and Statistical Methods

The contents of  $^{18}\text{O}$  and D are expressed by the per mil deviation relative to Vienna Standard Mean Seawater (V-SMOW). The calculation formula is as follows:

$$\delta^{18}\text{O}(\delta\text{D}) = \frac{R_{\text{sample}} - R_{\text{standard}}}{R_{\text{standard}}} \times 1000 \quad (1)$$

The sampling and determination process and error control of  $\delta\text{D}$  and  $\delta^{18}\text{O}$  contents follow the laboratory analysis standards formulated by IAEA/GNIP strictly. The network maintains appropriate standards and precautions to ensure reliable isotopic data, which standardizes the approaches for evaporation prevention and considering the representative of the samples as well as logistical and safety aspects when establishing the stations. Stable isotope measurements are usually performed by isotope ratio mass spectrometry and by laser absorption spectrometry. The equipment for sampling (e.g., rain collector, buried sampler, sample bottles and basic laboratory equipment) also follows a specific strategy. These measures could satisfy the accuracy of data to a large extent. Detailed information of the methods of sample collection/analysis is available on the "IAEA/GNIP precipitation sampling guidebook" published in the website of IAEA (<http://www-naweb.iaea.org>). The local meteoric water line is established by the linear regression analysis, and the correlation between  $\delta^{18}\text{O}$  and  $\delta\text{D}$  is analyzed and calculated by least square method (LSR). The relationship between  $\delta^{18}\text{O}$  value in precipitation and environmental factors (precipitation, vapor pressure and temperature), and the relationship between slope of LWML and geographical factors are established by linear and exponential function regression. T-test is used to verify whether the correlation between variables meets the 95% confidence level.

## Results and Discussion

### Seasonal Trends of $\delta^{18}\text{O}$ and $\delta\text{D}$ in Precipitation

Temporal variation of hydrogen and oxygen stable isotopes in precipitation in six cities is shown in Fig. 2. For these cities,  $\delta^{18}\text{O}$  and  $\delta\text{D}$  change synchronously, and both tracers exhibit a significant seasonal trend. The maximum values of  $\delta^{18}\text{O}$  and  $\delta\text{D}$  usually occur between January and April, which corresponds to winter and spring in the northern hemisphere, while the minimum values mostly occur between July and September, when the northern hemisphere is in summer and is strongly affected by monsoon activity. In monsoon climate zone, the change of moisture source of local precipitation is the main driving force to control the seasonal isotopic fluctuation in precipitation. In summer months, south

China is mainly affected by the southwest monsoon from Indian Ocean and South Asia and the southeast monsoon from Northwest Pacific [20]. During the process of air mass moving inland from the ocean, a large amount of precipitation occurs along the way, and isotopes experience "rainout effect" and gradually depleted heavy isotope. In winter, the whole land of China is controlled by the northwest monsoon from Siberia, and the moisture mainly comes from the evaporation of inland lakes and rivers. During this period, the precipitation along the way is scarce and the heavy isotopes are relatively enriched. Among the six cities, the highest mean isotopic value appears in Hong Kong, which is the only site located in the tropics and close to the South China Sea. The relatively short migration trajectory of moisture weakens the isotope depletion caused by precipitation along the path. The the most depleted value of average isotope concentration is observed in Kunming, which may be due to the fact that Kunming is located on the plateau area and has significantly lower temperature than other cities. Temperature is positively correlated with the isotope concentration. Therefore, the isotopic composition of Kunming is not only controlled by the "land-sea effect", but also by the "elevation effect" and "temperature effect". The largest seasonal fluctuations occur in Zunyi and Kunming in southwest China, followed by inland cities of Guilin and Changsha. The smallest fluctuations are observed in two coastal cities of Hong Kong and Nanjing. The distance between Kunming and Zunyi with South China Sea and Indian Ocean is close, and thus these cities are influenced by both southwest monsoon and southeast monsoon in summer. Moisture from different oceans shows varying isotopic compositions due to the different characteristics of surface temperature and relative humidity above sea, which leads to large fluctuations of isotopic compositions in local precipitation. Nanjing and Hong Kong are located close to the western Pacific Ocean and the South China Sea, respectively, and far from inland. Precipitation is mostly controlled by moisture from a single ocean, so it shows slight isotope fluctuation.

### Correlations between $\delta^{18}\text{O}$ and Local Meteorological Parameters

The temperature and precipitation are also recorded by the GNIP database with the observing of isotopes. In order to explore the influence of local environmental factors on isotope change, the relationship between isotope composition and climate factors is established by simple and multiple linear regression models. Considering the consistency of behavior between  $\delta^{18}\text{O}$  and  $\delta\text{D}$ , only the analysis of  $\delta^{18}\text{O}$  is reported. The correlation coefficient R between  $\delta^{18}\text{O}$  and precipitation (P), temperature (T) and water vapor pressure (V) in precipitation of six cities and the determination coefficient  $R^2$  of multiple regression equation between  $\delta^{18}\text{O}$  and independent variable combination are shown

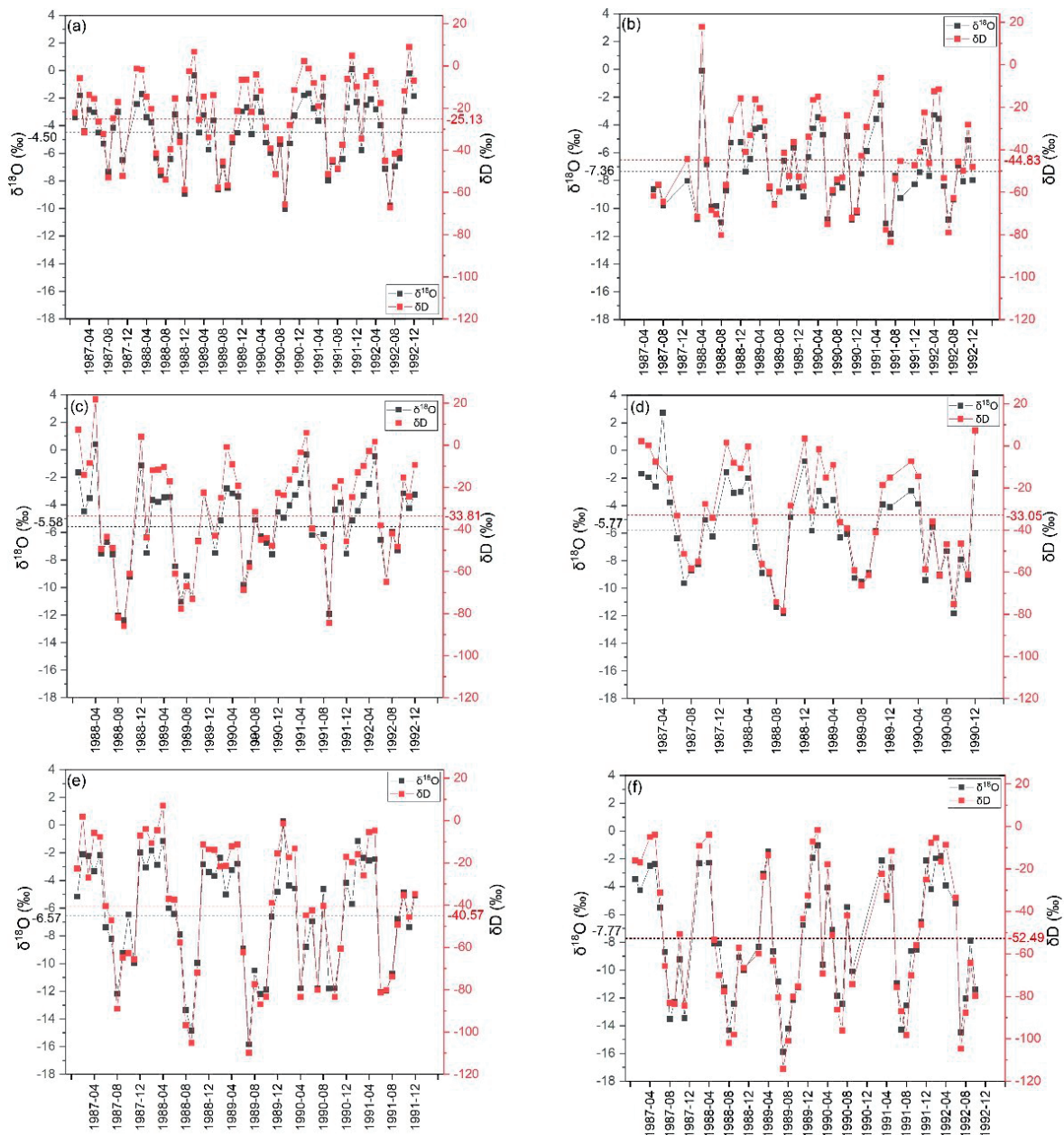


Fig. 2. Temporal variation of  $\delta^{18}\text{O}$  and  $\delta\text{D}$  in precipitation in Cities of a) Hong Kong, b) Nanjing, c) Changsha, d) Guilin, e) Zunyi and f) Kunming. The dash lines represent the arithmetic means of isotopes.

in Table 2. Since no water vapor pressure data were published at Guilin, Changsha and Kunming sites, only the correlation between  $\delta^{18}\text{O}$  and P, T and their combination is shown. According to the experience in other studies, the absolute value of  $r$  in 0.8~1 represents strong correlation, that in 0.4~0.8 represents good correlation, and that less than 0.4 represents weak correlation [21]. At most sites, good negative correlations can be observed between  $\delta^{18}\text{O}$  and precipitation, which is consistent with the „precipitation effect”. In Changsha and Guilin, which are cities located in inland areas, the correlation between isotope and precipitation is the weakest, which may be because that the time that ocean-sourced monsoon and land-sourced monsoon control these areas are similar on the annual scale, resulting

in the isotopic difference in water vapor carried by air masses from different sources. This masks the influence of local precipitation intensity on isotope fractionation. There are negative correlations between  $\delta^{18}\text{O}$  and temperature at all six sites, which is contrary to the „temperature effect” reported by Dansgaard [5]. This is partly due to the seasonal periodicity of isotopes in water vapor, which is low in summer and high in winter caused by monsoon switching. On the other hand, the precipitation and temperature change consistently due to the influence of monsoon activity while the correlations between these parameters and isotope are opposite. In low latitudes, “precipitation effect” is a more dominant control on isotopes [5], which may mask the effect of temperature. Such observation has

Table 2. Relationships between  $\delta^{18}\text{O}$  with single meteorological parameters and their combinations.

Site Name	$r$ ( $\delta^{18}\text{O}$ -P)	$r$ ( $\delta^{18}\text{O}$ -T)	$r$ ( $\delta^{18}\text{O}$ -V)	$R^2$ ( $\delta^{18}\text{O}$ -P+T)	$R^2$ ( $\delta^{18}\text{O}$ -P+T+V)
Hong Kong	-0.457	-0.650	-0.659	0.429	0.453
Nanjing	-0.447	-0.246	-0.390	0.161	0.375
Changsha	0.0039	-0.357	/	0.148	/
Guilin	-0.189	-0.715	/	0.489	/
Zunyi	-0.574	-0.635	-0.673	0.432	0.462
Kuming	-0.664	-0.503	/	0.433	/

also been reported in previous studies [18]. There is a good correlation between isotope and water vapor pressure, reflecting the influence of sub-cloud secondary evaporation. Under unsaturated atmospheric environment, isotopes are more likely to undergo dynamic fractionation due to evaporation in the process of raindrops falling, and lead to the enrichment of heavy isotopes. The higher the saturation degree of atmosphere is, the more negative the isotopic composition is. The results of multiple regression analysis show that the  $R^2$  values of the correlation equation between  $\delta^{18}\text{O}$  and combination “precipitation+temperature” are within a range of 0.148~0.489, and those of the correlation equation between  $\delta^{18}\text{O}$  and combination “precipitation+temperature+water vapor pressure” are within a range of 0.375~0.462, indicating that local meteorological parameters can only explain less than 50% of the total  $\delta^{18}\text{O}$  variance. This is because the effect of local

meteorological parameters on isotope occurs after the arrival of water vapor in monsoon climate zone, and the control of water vapor on isotopes is related to conditions of source and trajectories and may cover up the influence of meteorological parameters. Therefore, it can be inferred that the accuracy of predicting the future isotopic trend by meteorological records is not high in this region.

### Characteristics of Local Meteoric Water Lines ( $\delta^{18}\text{O}$ - $\delta\text{D}$ plots) and Their Drivers

#### *$\delta^{18}\text{O}$ - $\delta\text{D}$ Correlations in Monsoon-Affected Urban Regions*

The correlation between  $\delta^{18}\text{O}$  and  $\delta\text{D}$  in precipitation isotopes of six cities was established by the least square method (Fig. 3). Although all sites are located

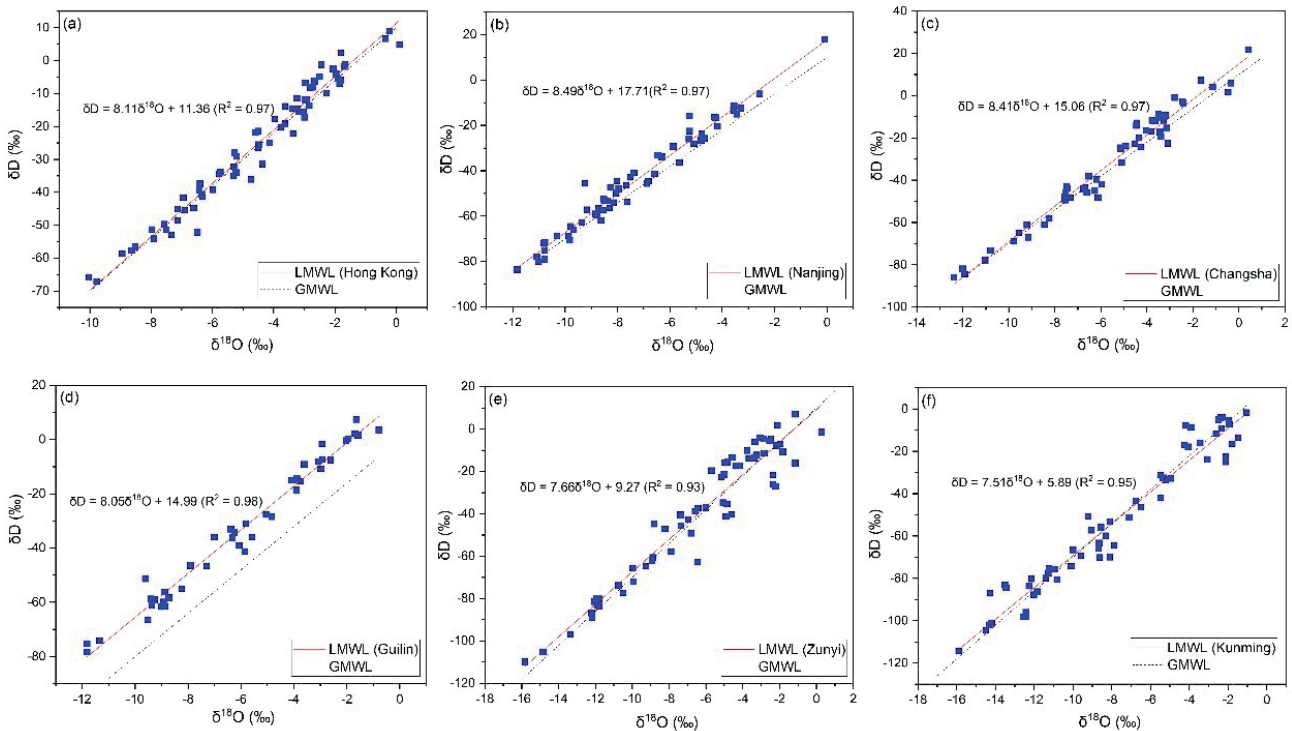


Fig. 3. Correlations between  $\delta^{18}\text{O}$  and  $\delta\text{D}$  in precipitation a) Hong Kong, b) Nanjing, c) Changsha, d) Guilin, e) Zunyi and f) Kunming. The dash lines represent the GMWL.

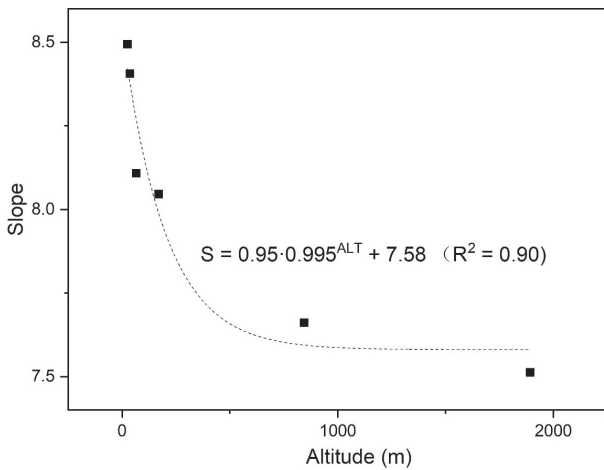


Fig. 4. Relationship between the slopes of LMWLs and altitude.

in the Yangtze River basin, and have similar climate patterns and monsoon transition regulations, their LMWLs present significantly different characteristics in slopes and intercepts. The slope of LMWL reflects the fractionation ratio of  $\delta^{18}\text{O}$  and  $\delta\text{D}$  and those in the six sites fluctuate from 7.51 to 8.49, which is close to that of GMWL, indicating the controlling role of marine air mass. Such range of slope is significantly higher than that of cities located in the monsoon region of north China reported in previous studies, such as Lanzhou (7.35), Fengxiang (7.05), Ningwu (6.88) and Yinchuan (7.21) [22-24]. For precipitation at the selected sites, most isotope scatters plot near LMWLs, and  $R^2$  values of regression equation are between 0.93 and 0.98, indicating that isotope mainly undergo equilibrium fractionation during the landing process of

raindrops. In Zunyi and Kunming, a number of isotope scatters plot below LMWL and deviate greatly from the line. This is likely because these two cities present the highest elevation (Fig. 1) and thus lowest air saturation among all selected sites, making them significantly influenced by the sub-cloud secondary evaporation. This is in agreement with the good correlation between  $\delta^{18}\text{O}$  and water vapor pressure observed in Zunyi site.

*Controls on the Slope Variations of LMWL*

Isotope fractionation is controlled by condensation temperature, moisture source, transport path and local climatic conditions, which are directly associated with the slope of LMWL [22]. Terrain change is one of the most decisive factors controlling the air mass migration path. The relationship between the elevation of sites and the slope of LMWL is shown in Fig. 4. By employing exponential function to fit the varying trend of slope with elevation change, a good negative correlation is observed ( $R^2 = 0.90$ ). However, a positive correlation between d-excess (an index positively correlated with the slope of LMWL) and elevation change was reported in previous studies, which was attributed to the change of isotopic fractionation coefficient caused by temperature variation [25]. The negative correlation between slope of LMWL and elevation may be due to the disturbance of water cycle caused by the existence of cities. All selected sites are located in urban areas, and the isotopic change patterns in precipitation are different from those in natural conditions due to the urban-related activities.

In urban areas, the uneven distribution of atmospheric heat and the change of material and energy

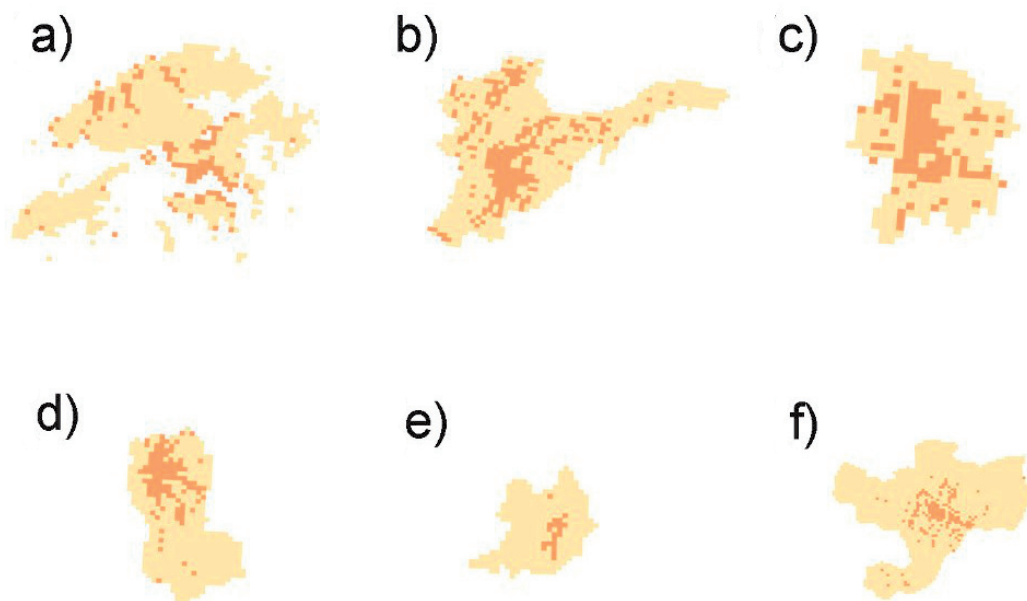


Fig. 5. Distribution of urban-related land uses in cities of a). Hong Kong, b) Nanjing, c) Changsha, d) Guilin, e) Zunyi and f) Kunming. The orange grids represent urban-related land uses and the light yellow ones represent non-urban types.

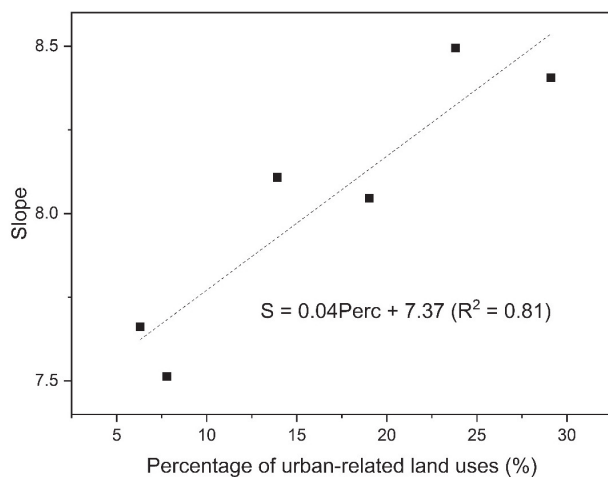


Fig. 6. Relationship between the slopes of LMWLs and the percentage of urban-related land uses in city area.

cycle caused by human activities lead to changes in the water cycle processes, which may impose impacts on isotope fractionation. Therefore, it is necessary to explore whether the urban environment is a driving factor of LMWL characteristics. In order to quantify the intensity of urban activities, the proportion of “urban, industrial and mining” land use types in the total urban area based on remote sensing images is calculated. The distribution of this land use in six cities is shown in Fig. 5. The correlation between the slope of LMWL and the proportion of urban-related land use is established by linear regression (Fig. 6). Significant positive correlation is observed between the two parameters, indicating that the water cycle related to cities may be one of the reasons for the regional differences in LMWL. On the one hand, the temperature rise in the areas with concentrated urban activities causes the equilibrium fractionation coefficient between D and  $^{18}\text{O}$  to increase. On the other hand, the formation of heat island effect in urban areas accelerates the rate of local evaporation, and increases the proportion of recycled water vapor characterized by higher D concentration in air mass, leading to the increase of LMWL slope. By comparing the differences of LMWLs in Chengdu, Southwest China between 1990s and 2018, Xia, et al. [26] reported that the slope increased from 7.64 to 8.63 during the 30-year time span, and attributed this change to the increase of urban-related land uses, which is consistent with our findings based on spatial comparison. However, the number of research samples these two studies based on is very limited, which can only be used as a speculation of influencing factors of LMWL in regional scale. In the future study, it is necessary to collect more samples on a larger (e.g., global or continental) scale and in a longer time scale to verify the comprehensive influence of the existence of cities on isotope fractionation.

## Conclusions

In this study, hydrogen and oxygen isotope characteristics of precipitation in six south China cities dominated by monsoon are analyzed.  $\delta^{18}\text{O}$  and  $\delta\text{D}$  show significant seasonal periodicity with valleys appearing in summer and peaks appearing in winter. Due to the alternating control of multiple monsoons, the isotopic variation is the largest in Kunming and Zunyi, which are located in the southwest China, while the most constant isotopic composition is observed in Hong Kong, which is closest to the sea. Meteorological parameters of temperature, precipitation and water vapor pressure all present negative correlation with isotope concentration, but the results of multiple regression show that the combination of meteorological elements can only explain less than 50% of the total variance, indicating the leading role of water vapor source on isotopes in local precipitation. The slopes of LMWLs at six sites are similar to GMWL, but there are differences among sites. At Kunming and Zunyi sites with higher elevation, the lower slope is observed because of the significant effect of sub-cloud secondary evaporation. There is a significant positive correlation between the percentage of urban land use and the slope of LMWL, indicating that the increase of isotope fractionation coefficient caused by temperature rise and the contribution of local recycled water vapor are the possible reasons for the LMWL change in urban areas.

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## Conflict of Interest

The authors declare no conflict of interest.

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