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Application of Stable Isotope Tracing Technologies in Identification of Transformation among Waters in Sanjiang Plain, Northeast China

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Abstract: In order to investigate the transformation among the precipitation, groundwater, and surface water in the Sanjiang Plain, Northeast China, precipitation and groundwater samples which were collected at the meteorological station of the Sanjiang Mire Wetland Experimental Station, Chinese Academy of Sciences and the surface water which collected from the Wolulan River were used to identify the transformation of three types of water. The isotope composition of different kinds of water sources were analyzed via stable isotope (deuterium and oxygen-18) investigation of natural water. The results show a clear seasonal difference in the stable isotopes in precipitation. During the cold half-year, the mean stable isotope in precipitation in the Sanjiang Plain reaches its minimum with the minimum temperature. The $\delta^{18}O$ and δD values are high in the rainy season. In the Wolulan River, the evaporation is the highest in August and September. The volume of evaporation and the replenishment to the river is mostly same. The groundwater is recharged more by the direct infiltration of precipitation than by the river flow. The results of this study indicate that the water bodies in the Sanjiang Plain have close hydrologic relationships, and that the transformation among each water system frequently occurs.

Keywords: hydrogen and oxygen stable isotopes; groundwater; precipitation; water cycle; recharge

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1 Introduction

Confirming the mechanisms of the hydrologic cycle to improve the utilization rate of water resources has become a hot research topic around the world in recent years (Boronina *et al.*, 2005; Breitenbach *et al.*, 2010; Baily *et al.*, 2011). Precipitation disperses on and near the surface through evaporation, runoff, and infiltration, causing changes in the stable isotope ratios of oxygen ($\delta^{18}O$) and hydrogen (δD) in distributed waters. This process of the change of oxygen and hydrogen was isotopic fractionation. The theory of the isotopic fractionation is that heavy isotopes concentrate in the remaining waters during the evaporation progresses. The changes in the stable isotopes among precipitation, surface water, and underground water indicate different stages of the hydrologic cycle (Gat, 1996; Gat and Airey, 2006). Stable isotope technologies are widely used in hydrological research. In a previous study on the changes in $\delta^{18}O$ and δD values in precipitation and surface mine water in Montana, USA, the evaporation volume of mine water in October, 2003 was found to exceed 50% (Gammons *et al.*, 2006). Similarly, fog vapor in Xishuangbanna, China mainly originates from evaporation from ponds, rivers, and soil, as well as from the plant respiration. Meanwhile, fog vapor is not quite related to the evaporation of streams, and most precipitation returns to the air through evaporation (Liu *et al.*,

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2007). Stable isotopes in each type of waters have their own specific values, and by analyzing the stable isotopes values can indicate the detailed information in the water cycling process.

In the 1960s, the Sanjiang Plain was the largest concentrated area of freshwater wetland in China, in which marsh vegetation was densely distributed (Wang *et al*., 2004). Over the last 50 years, the Sanjiang Plain was reclaimed four times, and most of the marsh wetland areas were turned into cultivated land. In particular, rice plantations have been rapidly developing since the 1990s. Farmlands of paddy rice reached 953 000 ha in 2000, accounting for 29.1% of the farmland area of the Sanjiang Plain. Large-scale land reclamation severely affects the local climate, and the average annual flow in swamp-type rivers has decreased and fluctuated since the 1960s, resulting in the overall decreasing trend of precipitation (Yan *et al*., 2004). Due to the extensive pumping of water for the irrigation of paddy rice fields, the underground water table in the major irrigated areas of the Sanjiang Plain has dropped over the years at a rate of 0.4 m/yr (Wang and Tian, 2003), and the decreasing trend in the groundwater level threatens the safety of water resources. The reserves and extraction of the underground water are the major factors that affect agricultural development in the Sanjiang Plain, and the way of underground water replenishment in the plain is always in dispute. Bai *et al*. (2004) proposed that underground water is mainly recharged by the vertical infiltration of precipitation, and the rainwater exceeds one half of the replenished underground water. Other researcheres thought that the extensive coverage of clay or loam layer (3–17 m) of the Upper Pleistocene Series on the surface of the Sanjiang Plain forms a wide and stable upper confining bed (Yin *et al*., 2004). By monitoring the underground water level, lateral replenishment at the alluvium and fissure water in the mountainous region was determined as the main channels for the replenishment of the underground water (Liu *et al*., 2001). Therefore, the studies on mutual transformation and replenishment between precipitation, surface water, and underground water are highly significant to the environmental protection and sustainable development of the Sanjiang Plain. Most researches on water circulation in the Sanjiang Plain focused on the changes in the groundwater table as well as the underlying reasons (Liu and Fu, 2008). However, little attention was paid on the transformation among precipitation, underground water, and surface water.

The application of a stable isotope to obtain information on the internal processes of the hydrologic cycle has already been implemented. Oxygen-18 and deuterium levels in water bodies vary because of the different physical conditions (e.g., atmospheric temperature) during evaporation and condensation. Hence, the replenishing source of underground water can be determined by identifying the source of moisture through different isotope contents. Lacumin *et al*. (2009) monitored the changes in $\delta^{18}O$ and δD values in the rivers and underground water in the basins of the northern Italy. The results indicated that the underground water of the Po Plain was mainly replenished by surface water. The changes in $\delta^{18}O$ and δD values in the underground water and local precipitation in the southwestern Spain showed the spatial and temporal discontinuity of underground water (Vandenschrick *et al*., 2002). The results showed that shallow groundwater is mainly replenished by precipitation resulting from the condensation of vapor from the Atlantic. Deep underground water is fossil water replenished by precipitation, which results from the condensation of vapor from the Mediterranean. Oxygen-18 and deuterium in local volcanic and underground waters in the southern Argentina showed a strong hydrologic relationship, and that infiltration of lakewater is the main source of the replenishment of underground water (Mayr *et al*., 2007). Therefore, the method of monitoring $\delta^{18}O$ and δD values in underground water is reliable in identifying the transformation between the different types of water.

The present study aims to reveal the mechanism of these transformations to further improve the internal mechanism of water circulation in the Sanjiang Plain and provide the evidence on that whether the problem of underground water resource shortage exists after the land use changed.

2 Materials and Methods

2.1 Study area

The experiment was conducted in the Sanjiang Mire Wetland Experimental Station, Chinese Academy of Sciences (47°35′N, 133°31′E) in Heilongjiang Province, Northeast China. The Sanjiang Plain is a large water-storing basin slightly inclined to the northeast. The Heli, Baoquanling, and Fujin Formations of the Tertiary system, consisting of mudstone, sandstone, and sandy conglomerate, deposited in the basin during the Tertiary Period. A Quaternary formation composed of sand, gravel, and cobble then covered the basin. Pore water is abundant in this formation, with fissure-pore water existing in the sandstone and sandy conglomerate of the Tertiary formation. The fissure of the Quaternary bedrock, which is distributed in the monadnock and platform of the basin, also contains some water. Water in these formations composes the groundwater system of the plain (Yang and Wang, 2011). The altitude of the Sanjiang Plain is 56 m, with a monsoon climate of the temperate zone. The average annual temperature is 1.9℃, and the annual precipitation ranges from 550 mm to 600 mm. The rainfall is abundant in summer and autumn, and the total rainfall from June to September accounts for more than 65% of the annual precipitation. Water and soil of wetland are completely frozen from November to April of the next year, and are thawed only by early June (Jia *et al*., 2006).

The Honghe National Natural Reserve is located at the hinterland of the Sanjiang Plain and extends across two administrative regions (Tongjiang City and Fuyuan County). The reserve is adjacent to the Honghe Farm, Qianfeng Farm, and Yaluhe Farm and has a total area of 21 836 ha. It has a long, cold, and snowy winter, a windy and dry spring, a hot summer, and short autumn. The average annual temperature is 1.9℃, the sunshine hours amount to 2356 h, and the average annual precipitation is 585 mm. Approximately 50%–70% of the precipitation is concentrated on July, August and September. The average annual evaporation is 1166 mmp (Luan *et al*., 2003). The Wolulan River is a swamp-type river in the interior of the Honghe National Natural Reserve.

2.2 Methods

2.2.1 Sample collection

The samples of the precipitation were collected at the meteorological station of the Sanjiang Mire Wetland Experimental Station, Chinese Academy of Sciences from October, 2004 to December, 2005. Well water samples (well depth, 18 m) were collected from the meteorological station from March to December in 2005. Surface water samples were collected from the Wolulan River from May to October in 2005. The precipitation, surface water, and underground water samples were all collected once a month. The rainwater sample was a mixture of all precipitation for that month. Surface water was also the mixture from three different sites. The distance among the sites was approximately 200 m. Each precipitation sample was sealed in a 100 mL plastic bottle, and the samples were mixed together for a month to prevent the evaporation. After the snow sample was completely thawed at room temperature, the precipitation, underground water, and surface water samples from the Wolulan River were sealed in 100 mL plastic bottles after collection. The numbers of precipitation, surface water, and groundwater samples were 15, 6 and 18, respectively. The water samples were stored in a refrigerated chamber prior to analysis.

2.2.2 Sample analysis

Stable isotope values are expressed in the δ notation (Sun *et al*., 2011).

$$\delta = \left(\frac{R_{\text{sample}}}{R_{\text{standard}}} - 1 \right) \times 1000 \tag{1}$$

where δ is isotope ratio, which is usually used to denote a difference measurement made relative to standards during the analysis; R_{sample} is the ratio of the heavy (e.g., ^{18}O and D) to the light (e.g., ^{16}O and ^{1}H) isotope of sample; R_{standard} is the international standard ratio of the heavy (e.g., ^{18}O and D) to the light (e.g., ^{16}O and ^{1}H) isotope.

The oxygen-18 and deuterium values were measured by using Finnigan Mat 253 mass spectrometer (Finnigan, USA), and are expressed as δ values in parts per million relative to the international standard, the Vienna Standard Mean Ocean Water. The reproducibility of δD and $\delta^{18}O$ values is ±2‰ and ±0.1‰, respectively.

2.2.3 Underground water monitoring

The underground water tables in seven wells along the Bielahong River were monitored in September, 2009 to further prove the recharging process of underground water in the Sanjiang Plain. The locations of the monitored wells are shown in Fig. 1. The underground water was monitored by using Odyssey capacitance water level gauge (Odyssey, New Zealand) over a 6 h interval, and the resolution of the instrument is 0.8 mm.

2.2.4 Statistical analyses

The statistical analyses were carried out using SPSS software version 16.0. To test the normal distribution of $\delta^{18}O$ and δD values in underground water, surface water and precipitation, *Q-Q* probability plots were employed. The data of $\delta^{18}O$ and δD values in underground water,

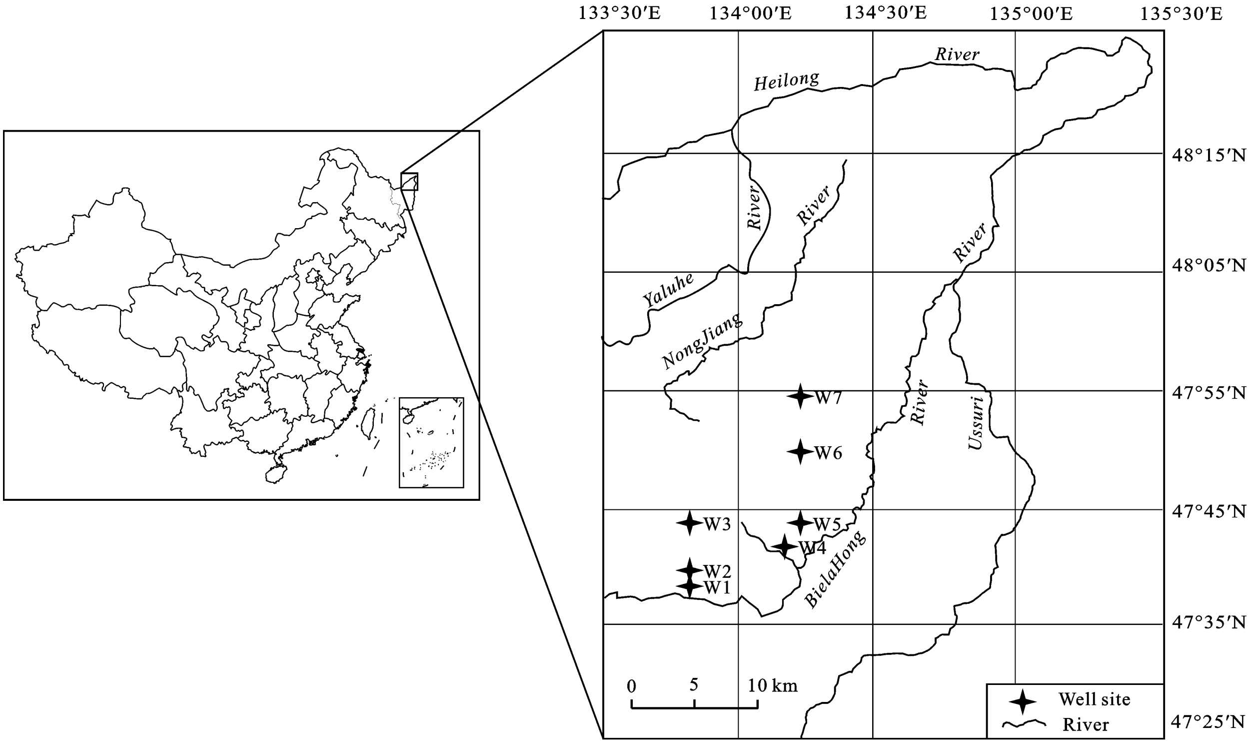

Fig. 1 Location of well sites

surface water and precipitation, were all at normal distribution. The ANOVA of δ^{18}O and δD values in underground water, surface water and precipitation was performed using one-way ANOVA, and the significance limit was set to $p < 0.05$. If p was lower than 0.05, it means there is the significant difference. The Least Significant Difference or Tamhane′s T2 procedure was used to determine the significant differences between each group of concentrations.

3 Results and Analyses

3.1 δ^{18}O and δD values in precipitation

Harmon (1961) revealed that the relationship between δD and δ^{18}O values in atmospheric precipitation around the world is $\delta D = 8\delta^{18}O + 10$ (global meteoric water line, GMWL). Based on the monitoring in 17 regions for two years, the Hydrogeology Engineering Research Institute of the Ministry of Geology and Mineral Resources deduced that the linear equation of precipitation in China is $\delta D = 7.83\delta^{18}O + 8.16$ (Chinese meteoric water line, CMWL) (Zhang, 1989). δD and δ^{18}O values in snow and rain, GMWL and CMWL are shown in Fig. 2. δ^{18}O and δD values in snow from the Sanjiang Plain were quite dispersed and generally above the GMWL, while those in most rain were more concentrated and below the GMWL. δ^{18}O and δD values in precipitation were basically distributed around the GMWL. Both δ^{18}O and δD values in snow and rain were above CMWL. Meanwhile, δ^{18}O and δD values in precipitation periodically changed with the season. Based on statistical analysis, δD values in snow was significantly lower than that in rain ($p < 0.05$), while δ^{18}O values in snow was not significantly different from that in rain ($p > 0.05$). Gener-

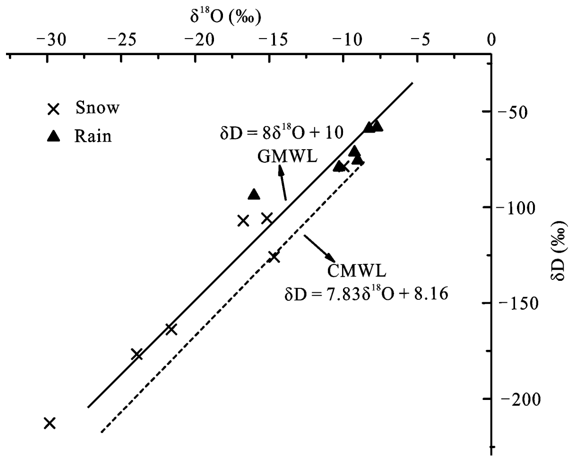

Fig. 2 δD vs. δ^{18}O values in snow and rain samples along with global meteoric water line (GMWL) and Chinese meteoric water line (CMWL)

ally, $\delta^{18}O$ and δD values were high in summer rain and low in winter snow, thereby exhibiting a periodic change in one year. Similar results were found in central Pennsylvania, USA (Driscoll *et al.*, 2005) and in the Gobi Desert of Mongolia (Kirsanow *et al.*, 2008).

These $\delta^{18}O$ and δD values of the different types water for the Sanjiang Plain were used to obtain the local meteoric water line (LMWL) and to determine the relationships among the different types of water systems. The straight line for the Sanjiang Plain in Fig. 3 obtained via linear regression has a slope close to 7 (6.81, $R = 0.9766$, $p < 0.01$) and thus it can be considered as LMWL. The results in Fig. 3 show that the $\delta^{18}O$ and δD values of the underground water and the surface water in the Sanjiang Plain were distributed on both sides of the LMWL. In addition, the slope and intercept of the LMWL were both lower than those of GMWL and CMWL.

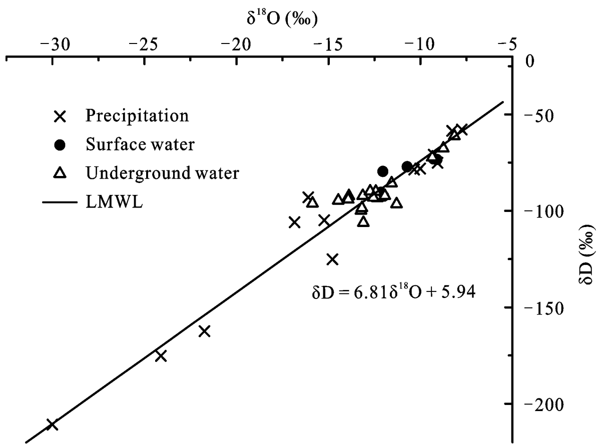

Fig. 3 δD vs. $\delta^{18}O$ values of of different types of water along with local meteoric water line (LMWL)

3.2 $\delta^{18}O$ and δD values in underground water and surface water

$\delta^{18}O$ and δD values in underground water and precipitation varied within a large range, while those in the surface water were quite stable (Table 1). The mean values of $\delta^{18}O$ and δD decreased in the following order: surface water, groundwater, and precipitation. The minimum $\delta^{18}O$ and δD values in underground water, precipitation, and surface water were in December, January, and October, respectively, and the maximum values were in August, June, and September, respectively.

Figure 4 shows the variability of δD values in the Wolulan River and rain from May to October, 2005. For the Wolulan River, δD values reached the peak in August and September. The δD values decreased in October. By contrast, the δD values in rain increased from May to June, and reached its peak in June, and then decreased and reached its minimum value in August. From August to October, δD values increased again. The rainy season in the Sanjiang Plain is from July to September, and the rainfall is the highest in July with 135.9 mm. Based on the statistical analysis, δD values in the Wolulan River and rain samples did not show a significant difference ($p > 0.05$).

4 Discussion

4.1 Changes in $\delta^{18}O$ and δD values in precipitation

According to the Rayleigh equation, vapor from water bodies has lighter isotopes, and the remaining water has heavier isotopes (Li *et al.*, 2007; Wen *et al.*, 2008). Therefore, snow in the Sanjiang Plain has lighter isotopes compared with rain. In addition, $\delta^{18}O$ and δD values are low in winter and high in summer, conforming to the seasonal effects of the isotopes. This result could be attributed to the strong effect of the monsoon in the temperate zone of the Sanjiang Plain. The temperature in winter and summer are quite different, with the average temperature of 21.18℃ in July and –20.17℃ in January. The vapor for precipitation comes from different sources. The northerly wind from the high-altitude hinterland (Mongolia-Siberia) contributes to the low temperature in winter; eastern and southeastern winds prevail in summer, and the southeastern monsoon blowing from the tropical ocean brings abundant precipitation. When vapor freezes to snow in winter, the isotope values are above the GMWL because of fractionation.

Table 1 Eigenvalues of $\delta^{18}O$ and δD values in different types of water in Sanjiang Plain

Water sample	δD (‰)			$\delta^{18}O$ (‰)		
	Mean	Minimum	Maximum	Mean	Minimum	Maximum
Precipitation	–107.4 ± 48.1	–210.8	–57.8	–14.9 ± 6.9	–30.0	–7.8
Surface water	–81.0 ± 8.5	–92.4	–73.0	–10.9 ± 1.4	–12.2	–9.1
Underground water	–89.7 ± 11.5	–106.1	–61.2	–12.3 ± 2.0	–15.9	–8.2

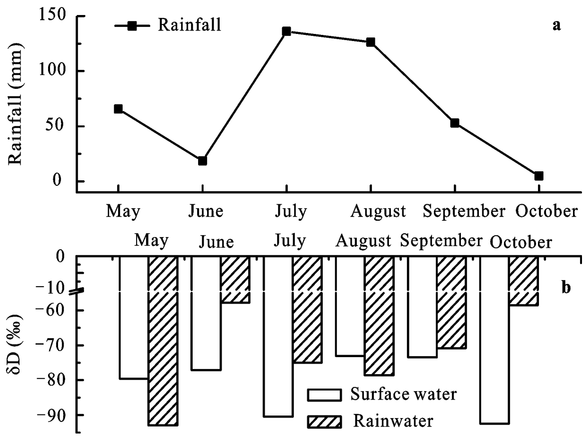

Fig. 4 Rainfall (a) and variability of δD values (b) in surface water and rainwater from May to October, 2005

By contrast, summer precipitation has a higher number of heavy isotopes because of evaporation. Similar results were found in other researches. By comparing the changes in $\delta^{18}O$ values in precipitation along major paths of vapor transport that affect precipitation in China, Zhang *et al*. (2005) concluded that $\delta^{18}O$ values changes with the season and the precipitation volume. For example, $\delta^{18}O$ values in the precipitation along the vapor path in the south of China remarkably differ over the seasons (Zhang *et al*., 2005).

The $\delta^{18}O$ and δD values in snow and rain were above the CMWL, reflecting the climate characteristics (such as humid air, a total evaporation lower than the precipitation, *etc*.) of the Sanjiang Plain. Meanwhile, the slope and intercept of the LMWL are both lower than those of GMWL and CMWL, indicating that the evaporation in the Sanjiang Plain is higher than the mean values of GMWL and CMWL. The results in Fig. 5 show that the

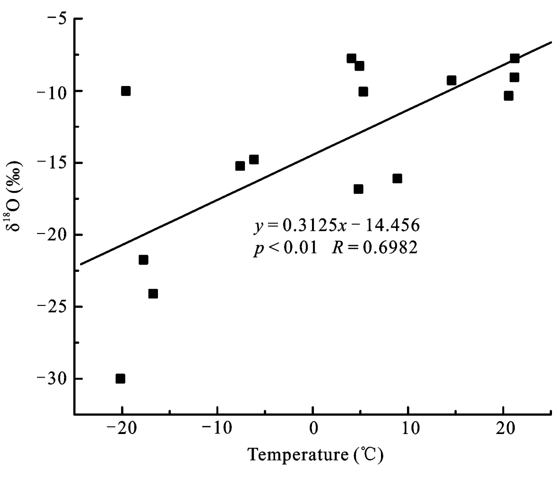

Fig. 5 Changes of $\delta^{18}O$ values in precipitation with temperature

$\delta^{18}O$ values in 15 precipitation samples collected monthly from October, 2004 to December, 2005 has significant positive correlations with the temperature. In other words, the changing trend of $\delta^{18}O$ values is consistent with that of temperature and conforms to the temperature effect on the isotope levels.

4.2 Replenishment of underground water

Figure 3 shows that $\delta^{18}O$ and δD values in the underground water and the surface water in the Sanjiang Plain are distributed at both sides of the LMWL, indicating the apparent hydrologic link among the three types of water systems. The results of Table 1 show that the $\delta^{18}O$ and δD values in underground water were between those of the precipitation and surface water. It proved that the underground water was replenished by the precipitation and the surface river water. As shown in Fig. 6, the underground water level significantly increased when the precipitation was high; the water level also increased from early April to middle May, when the precipitation was abundant. The underground water level reached the minimum burial of the year in early April, but remarkably dropped when the underground water was pumped for irrigation in the middle of May; the water level reached its lowest value at the end of June. The underground water level steadily increased during the rainy season (from early July to the end of September). The groundwater in the Sanjiang Plain was pumped from around the 25th of April to the 25th of June in 2005. The most concentrated and highest precipitation in the Sanjiang Plain occurs from July to September, and the precipitation reached 315 mm in 2005. The underground water level was restored to that from January to May and became stable. This phenomenon suggests that the precipitation recharges the underground water to some extent.

The results in Fig. 7 show that the $\delta^{18}O$ values in the underground water in the meteorological station of the Sanjiang Station remarkably increased in July and September because of the high $\delta^{18}O$ values in the precipitation. It also indicated that the underground water was mainly replenished by the precipitation.

The underground water level was monitored in a large scale to illustrate more deeply the underground water recharging process. The results in Table 2 and Fig. 8 reveal that the water levels of the wells remarkably rose from 0.475 m to 6.013 m (corresponding to a precipitation of 33.8 mm) from 18:00 on September 17 to 6:00

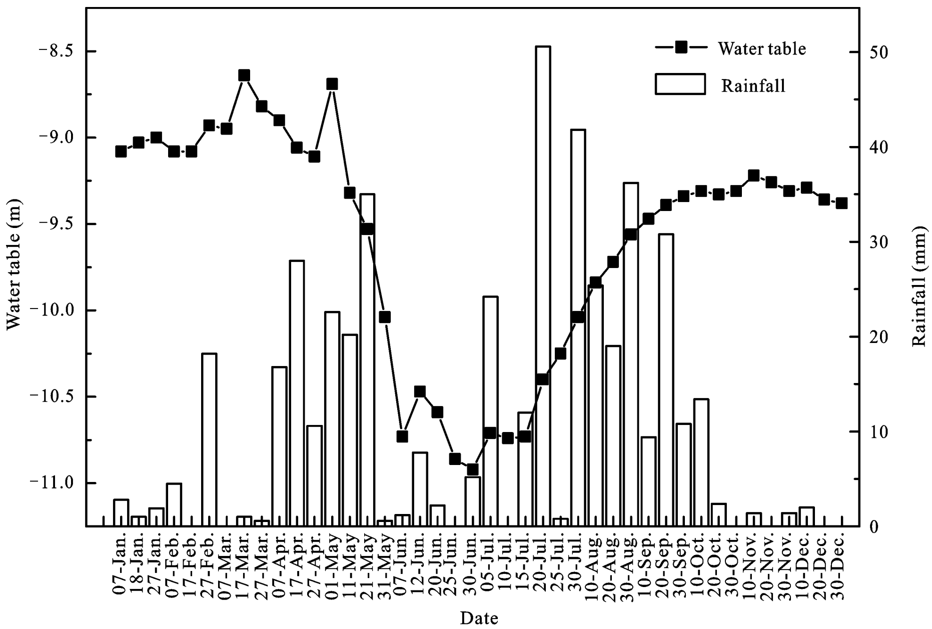

Fig. 6 Seasonal variability of rainfall and water table at meteorological station in study area in 2005

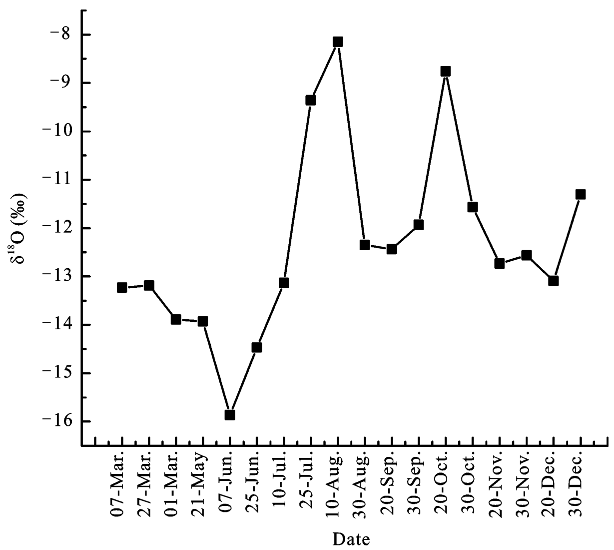

Fig. 7 Seasonal variability of $\delta^{18}O$ values in underground water at meteorological station in study area in 2005

on September 18. The sharp change in the underground water table within a short period basically revealed that the underground water in this area might not come from distant sources; thus, the direct replenishment by precipitation was quite high. The differences among the water levels in the wells are possibly due to local landforms. For example, water table of W3 increases rapidly because W3 is at the center of a farmland. This phenomenon may contribute to the 'funnel phenomenon' resulting from the excessive extraction of the underground water during irrigation periods. The water level was rapidly restored because of the large-volume replenishment from rainwater. Therefore, the rise of the underground water level could be attributed to the replenishment of underground water by the precipitation. In addition, the rising level of the Bielahong River caused by the lateral replenishment of underground water in this region.

4.3 Evaporation and water supply in Wolulan River

In the Wolulan River, $\delta^{18}O$ and δD values were distributed on the two sides of the LMWL (Fig. 3). It implied that the Wolulan River did not evaporate all the time; otherwise, the $\delta^{18}O$ and δD values in the Wolulan River would form an evaporation line below the LMWL. The distribution of $\delta^{18}O$ and δD values to LMWL indicated that the evaporation is almost equal to the replenishment of the river water. As shown in Fig. 4, from May, 2005 to October, 2005, the δD values in the Wolulan River reached the peak in August and September and reached the bottom in October. According to the Rayleigh equation (Lee *et al.*, 1999), when water evaporates, the remaining water body becomes enriched with heavier isotopes. Therefore, the values of $\delta^{18}O$ and δD in the remaining water would increase. The air temperature in the Wolulan River was high from July to September, and the amount of evaporation was higher than those in

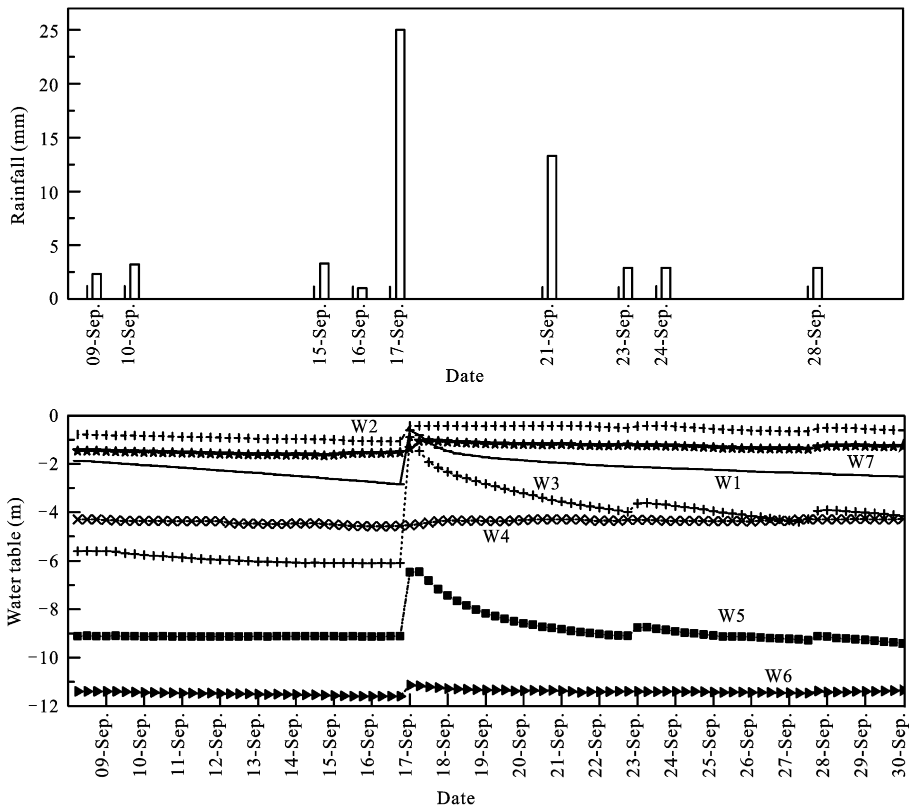

Fig. 8 Variability of rainfall and water table at Belahong River in September 2009

Table 2 Comparison of well-related characteristics in 2009

Well	Depth (m)	Beginning time of water table raising	End time of water table raising	Rise in water level (m)
W1	3	September 17th at 18:00	September 17th at 24:00	2.218
W2	5	September 17th at 24:00	September 18th at 06:00	0.635
W3	6	September 17th at 19:20	September 18th at 01:10	6.013
W4	6	September 17th at 18:00	September 18th at 12:00	0.136
W5	9	September 17th at 20:00	September 18th at 02:10	2.760
W6	12	September 17th at 19:20	September 17th at 23:50	0.475
W7	13	September 17th at 19:50	September 18th at 09:00	1.040

other months. As a result, heavy isotopes enriched the surface water, and the value of δD values increased. However, the rainfall in the Wolvlan River was the highest in July, resulting in an obviously decreased δD values. Therefore, the river was replenished by the rainfall in July, indicating that the Wolulan River underwent the most remarkable evaporation in August and September.

5 Conclusions

This study provides the LMWL in the Sanjiang Plain based on the changes in $\delta^{18}O$ and δD values in the underground water, surface water, and the precipitation in 2005 and 2009. The slope and intercept of the LMWL are both lower than those of GMWL and CMWL, indicating that the evaporation is higher than the global mean value as well as the value of China. The changes of $\delta^{18}O$ and δD values in the precipitation (i.e., increasing in summer and decreasing in winter) are mainly due to the temperature as well as the seasonal effects. The Wolulan River exhibits the most apparent evaporation in August and September. Thus, the evaporation and replenishment balance each other. Knowledge of the recharging process of the groundwater is important in assessing the regional water resources in the Sanjiang

Plain. The results show that the shallow groundwater, the surface water, and the precipitation in the Sanjiang Plain have a strong hydrologic link. Furthermore, the underground water is mainly replenished by the vertical infiltration of precipitation. The study area is located in the northeast of the Sanjiang Plain. More studies should be carried out in other areas because of the different geologic structure. The chemistry method also should be used to identify the transformation among waters in the Sanjiang Plain.

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