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Quantifying dual recharge mechanisms in deep unsaturated zone of Chinese Loess Plateau using stable isotopes

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ABSTRACT

Apportioning recharge to piston flow and preferential flow is critical for groundwater resource management and hydrologic connectivity evaluation, but the occurrence of the two flows in thick unsaturated zones is still poorly understood. The main objective of this study was to quantify the relative contributions of these two flows in the deep unsaturated zone of the Loess Plateau, China (CLP). The Bayesian mixing model (MixSIAR) was integrated with the line conditioned excess (lc-excess) of stable isotopes (δ^2H and δ^{18}O) in precipitation, soil water (8–25 m), and groundwater to determine the contributions. Piston flow contributed 62 ± 8% to the total recharge while preferential flow did only 38 ± 8%, indicating piston flow was the dominant groundwater recharge mechanism. On comparison with results from similar study sites in the CLP, the primary groundwater recharge mechanism differed among sites and may depend on spatial scales or geomorphology. The information is critical for understanding hydrological process and evaluating groundwater quantity and quality in the highlands of the CLP and other regions with similar hydrogeological conditions.

1. Introduction

Groundwater is critical for water and food security, human and environmental health protection, and ecosystem functioning (Gleeson et al., 2016). However, the ever-increasing population has exerted a tremendous pressure on this scarce natural resource and severely depleted and contaminated it across the world (de Vries and Simmers, 2002). As continuous withdrawal depletes groundwater faster than replenishment, it is critical to understand and quantify the recharge mechanisms for proper and sustainable management of this natural resource. This is especially true in arid and semi-arid regions where the groundwater recharge rate is slow due to the limited precipitation and higher evapotranspiration (Scanlon et al., 2006).

Hydrologists have long assumed that water movement in soil is dominated by piston flow (translative flow, soil matrix flow) where new water entering the soil mixes completely with the existing or resident water and moves deeper into the profile and ultimately reaching groundwater and stream (Mcdonnell, 2014). Another type of water movement is preferential flow, which is described as the water movement by preferred pathways at an accelerated pace through a fraction of active pores bypassing a small portion of the soil matrix (Lin, 2010). Increasing evidences from field observations (Beven and Germann, 1982; Beven and Germann, 2013; Brooks et al., 2010; Clothier et al., 2007; Evaristo et al., 2015; Nimmo, 2012) and theories (Lin, 2010) have indicated that preferential flow is a universal phenomenon in hydrology. Thus, the duality of flow is likely ubiquitous in any soil hydrological system. Moreover, the water moving via piston flow is the connected water and its fraction in the total flux determines the hydrological connectivity; therefore, increased hydrological connectivity may result from a large percentage of piston flow over preferential flow (Good et al., 2015). Overall, quantifying relative contributions of these two flows and identifying the dominant one is critical for assessing hydrological connectivity and groundwater quantity and quality, but still a challenging scientific issue.

A large body of literature have identified and quantified dual flow in soils with high heterogeneity (e.g. sand, fractured granites, and karst geomorphology) using various methods (Manna et al., 2017; Sharma and Hughes, 1985; Sukhija et al., 2003). For example, Sharma and Hughes (1985) first used dual components chloride mass balance in a sandy aquifer from the southwestern coast of Australia, and reported

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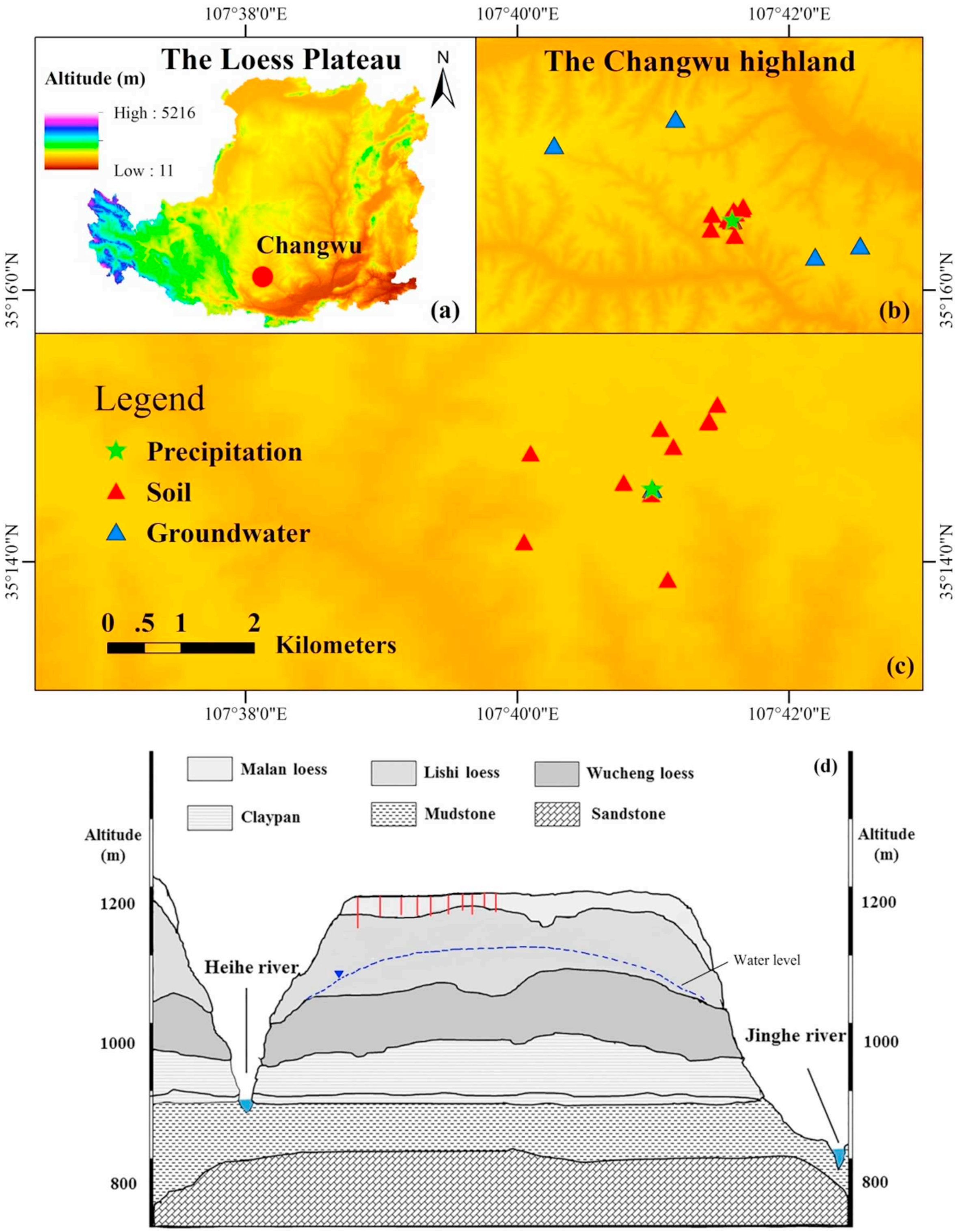

Fig. 1. Geographic locations of the Changwu highland (a), the sampling sites for precipitation, soil and groundwater (b–c) and the hydrogeological information about the study area (d, modified from Huang et al. (2018)). In the (d), the red vertical solid lines represent the soil cores. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

the existence of dual flow with the contribution as high as up to 50% of the total annual recharge from preferential flow. Sukhija et al. (2003) have quantified these flows at three sites in India with different geological settings and reported the dominance of piston flow in alluvial (100%) and sandstones (68%) aquifers, while the dominance of preferential flow in granites aquifer (75%). Manna et al. (2017) have also quantified the contribution of piston flow (80%) and preferential flow (20%) in an upland exposed sandstone unsaturated zone from southern California. In these studies, the dual flows were easily identifiable and verifiable due to the highly heterogeneous nature and/or shallow depth of soil, while very few studies have tested these in systems with relatively homogeneous and thick unsaturated zone.

The Loess Plateau, China (CLP), situated in the upper and middle reaches of the Yellow River, has a total area of approximately

640,000 km^2, and is predominantly covered by loess deposits ranging from 30 to 80 m in thickness (Wang et al., 2010b; Zhu et al., 1983). The loess represents a terrestrial clastic sediment, composed predominantly of silt-size particles (relatively coarse and homogeneous), which is formed essentially by the accumulation of wind-blown dust (Pye, 1995). The sustainability of the livelihood and the agricultural production of this region depends on the shallow groundwater, which has already experienced a rapid decline within last few decades (Currell et al., 2012; Gao et al., 2015; Huang and Pang, 2013; Wang et al., 2010a). Additionally, the accumulation of solutes such as fertilizer nitrate within the upper unsaturated zone were widely noticed in the CLP (Huang et al., 2013; Huang et al., 2016), deteriorating the quality of water if solutes reaching groundwater. Therefore, it is urgent to determinate the groundwater recharge mechanism or the dominant mechanism (contributions of these two flows) for groundwater quantity and quality management. Numerous studies have confirmed the existence of piston flow and quantified it at different sites within the CLP using methods including chloride mass balance, water isotopes (e.g. tritium concentration), unsaturated zone modeling (Deng et al., 2015; Huang and Gallichand, 2006; Huang and Pang, 2011; Huang et al., 2013; Huang et al., 2017a; Huang et al., 2017b; Huang et al., 2016; Li et al., 2016; Lin and Wei, 2006). For example, some studies have analyzed pore water tritium within the deep soil profiles and concluded dominant piston flow from the clearly defined peaks that corresponded to the high concentration tritium of 1963 precipitation (Li et al., 2016; Lin and Wei, 2006; Zhang et al., 1990; Zhang et al., 2017). Zheng et al. (2017) simulated rainfall infiltration using Brilliant Blue dye and concluded the presence of piston flow. Recently, Huang et al. (2017a) also confirmed the dominance of piston flow after combining signatures of multiple tracers from thick unsaturated zone and saturated zone in Zhengning highland. They also concluded that the time required for annual precipitation to reach the water table is about 160–400 years and the shallow groundwater has relatively old ^{14}C age.

Conversely, the dominance of preferential flow was concluded from the monitoring of soil water and groundwater table in a highland area (Luochuan) on the CLP (Wang, 1982; Yan, 1986). Moreover, Tan et al. (2016) used stable isotopes and showed the preferential flow in the western CLP, occurring likely though sinkholes, slip surface or landslide surface. In general, these studies have qualitatively characterized the preferential flow, but the contribution towards the total recharge was rarely quantified. In a recent study, Li et al. (2017) concluded the dominance of preferential flow (87% of the total groundwater recharge) at a watershed (Heihe) within the CLP using the chloride mass balance approach. However, due to the variability of the landscapes within the CLP, the relative importance of dual flow behavior in the thick homogenous soils remains controversial.

Therefore, the overall objectives of this study were (1) to identify recharge mechanisms based on soil water, precipitation and groundwater isotopes; and (2) to quantify the relative contribution of each recharge mechanism to groundwater using the state of the art Bayesian framework. Particularly, this study identified the dominant processes and their relative contributions in total groundwater recharge using the water isotopic mixing ratio in the precipitation, soil water and groundwater. Loess covers 10% of the world terrestrial land surface (Lin and Wei, 2006; Liu, 1985) and the results may help improving our knowledge of water cycle in the thick and relative homogeneous unsaturated loessial soil and other similar systems on Earth.

2. Materials and methods

2.1. Study site

A field experiment was conducted at Changwu highland (35.28° N, 107.88° E) within the southern part of the CLP (Fig. 1a). The landscape of the study area is a typical topographic-ecological units in the CLP, which consists of two geomorphic subunits: highland and gully. The Changwu highland is flat with the mean altitude of 1220 m above sea level. The soil at the study area belongs to the Castanozems (or Heilutu series, named by Zhu et al., 1983), which is mainly silty clay loam, with the silt content > 50% (Cheng et al., 2014). The climate is temperate continental monsoon with the mean annual precipitation of 578 mm (1957–2015), with 55% falling from July to September. The mean annual temperature is 9.5 °C.

The mudstone and sandstone bedrocks were covered by three loess layers, including Wucheng Loess, Lishi Loess, and Malan Loess (Fig. 1d). The top soil is Malan Loess, with a thickness of about 15 m. The Wucheng Loess has a thickness of 22–78 m and is the aquitard due to it relatively low permeability. As gullies are deeply cut into the bedrock and the rivers located in gullies are much lower than water table (Fig. 1d), the groundwater system is isolated from surface water, thus, precipitation is the only water source for recharge. The depth to the water table ranges from 30 m at the center to 100 m at the edge of the highland. Groundwater flows from the center to the edge at a very slowly velocity (Huang et al., 2017a) and discharges to streams as spring and ultimately to rivers. Thus, groundwater is a major source to sustain river and critical for river-dependent irrigation agriculture in gullies, and is main fresh water source for local residents on the highland.

2.2. Sample collection and laboratory analysis

A total of 200 daily precipitation samples were collected from the rain gauge at Changwu Agro-Ecological Experiment Station (Fig. 1b–c) during 2005, 2010, and between 2012 and 2015. Each sample was stored in 150 mL polyethylene plastic bottle and refrigerated at 4 °C for further analysis.

Ten deep soil cores were obtained along the center to edge of the Changwu highland during 2015–2016 (Fig. 1c–d). Among those, three cores were taken within farmlands and the remainder were taken within apple orchards of different stand ages; 6, 9, 11, 15, 17, 18 and 24 years. At each site, a hollow-stem auger was used to obtain soil cores (about 800 cm^3) to a depth between 10 and 25 m (varied with locations) at a sampling interval of 0.2 m. Each soil sample was further split into two parts after mixing completely; one part of about 40 g was used to measure soil water content by oven drying method and the other part was stored in a 250 mL polyethylene plastic bottle sealed with parafilm for extraction of soil water using cryogenic vacuum distillation. The rate of water extraction ranged from 98% to 102%. The extracted water was then stored in a 10 mL glass bottle and refrigerated at 4 °C before stable isotope analysis.

Groundwater samples were collected from five sites (Fig. 1b–c) in the Changwu highland during 2015–2016. One site was very close to precipitation and soil sites while the others were far apart. In these sites, one was spring and the others were from wells, which are used as drinking water source. The spring samples were directly collected at the springhead. At well sites, the groundwater was pumped out for few minutes before sample collection. At each site, groundwater was sampled once per month for a total of 43 times.

Stable isotopic compositions of precipitation (n = 200), soil water (n = 212) and groundwater (n = 43) were analyzed using LGR LIWA V2 isotopic liquid water analyzer (Los Gatos Research Inc., San Jose, CA, USA) at the water isotope analysis laboratory, Northwest A&F University, China. The precision of the instrument is 1.0‰ for δ^2H and 0.2‰ for $\delta^{18}O$. Nitrate concentration of groundwater was analyzed by ion chromatography (ICS-1000, Thermal Fisher Scientific, USA) at the same laboratory. Two groundwater samples (600 mL) were sampled: one from a deep well (100 m below the ground) and the other from a shallow well (32 m below the ground). Tritium concentration was determined by an ultra-low-level scintillation counter (Quantulus 1220, PerkinElmer, Singapore).

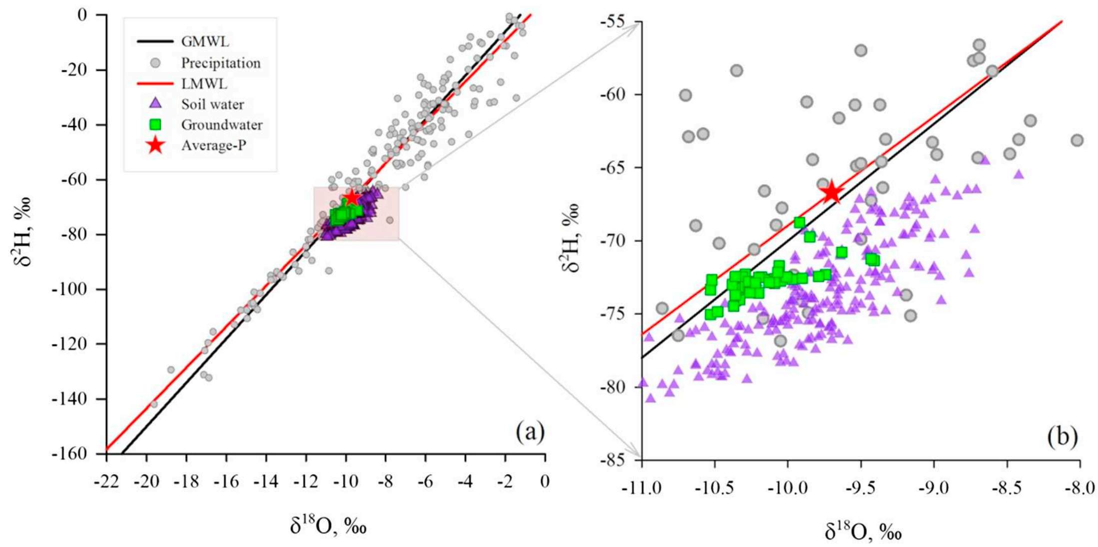

Fig. 2. Dual-isotope plots precipitation, deep soil water (8–25 m), and groundwater. In the legend, the GMWL and LMWL respectively represent the global meteoric water line ($\delta^2H = 8\delta^{18}O + 10$) and the local meteoric water line ($\delta^2H = 7.45\delta^{18}O + 5.52$, $R^2 = 0.93$, n = 200). The red star is the volume-weighted average values of precipitation. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

2.3. Analysis methods

2.3.1. Identifying dual flows

The dual flows included piston flow (soil water) and preferential flow (precipitation). In this study, we used dual-isotope approach (Evaristo et al., 2015), the line-condition excess (lc-excess), to identify the dual flows. The lc-excess has advantages over simpler and direct analysis of δ^2H and δ^{18}O to better distinguish the isotopic difference among precipitation, soil water and groundwater (Sprenger et al., 2017). The values of lc-excess were calculated following:

$$lc - excess = (\delta^2 H - a\ \delta^{18}O - b)/S \quad (1)$$

$$S = \sqrt{s_H^2 + s_O^2} \quad (2)$$

where a and b were the slope and the intercept of the local meteoric water line (LMWL), respectively. s_H and s_O were the standard errors for ^{2}H and ^{18}O for δ^2H and for δ^{18}O measurements, respectively. For LGR LIWA V2 isotopic liquid water, s_H = 1‰ and s_O = 0.2‰.

Stable isotopes of soil water are highly variable with time and depth due to precipitation infiltration, soil evaporation and land use type; thus, determination of the steady depth (critical depth) for soil water isotope is critical to piston flow. First, the long-term average infiltration depth in farmland is limited to 3 m depth (Chen et al., 2008) and an maximum infiltration depth is 5 m in an extremely wetter year (Liu et al., 2010). Second, Zhang et al. (2017) reported the 1963-tritium peak within the soil profile at the depth of 7.2 m indicating the depth of water movement in the past 50 years. As the apple trees have a stand age < 24 years, the water that recharged after farmlands converted to apple orchards has not reached 7.2 m or below, thus a depth of 8 m was considered stable to further exclude the impacts of land cover change on stable isotopes. Therefore, the stable isotopes of 8–25 m were used to compare with precipitation and groundwater for identifying groundwater recharge mechanism in this study.

2.3.2. Quantifying contributions of dual flows

The MixSIAR statistical package (Stock and Semmens, 2013), a general Bayesian framework, was used to quantify the contributions of different flows to the total groundwater recharge. The Bayesian mixing models have advantages over simple linear mixing models in estimating probability distributions of source contributions, have user-friendly interfaces, and can incorporate complexities such as variability in isotope signatures, discrimination factors, hierarchical variance structure, covariates, and concentration dependence (Phillips et al., 2014). It has been widely used in sourcing plant water, pollutants, and soil carbon and in food-web studies (Phillips et al., 2014; Rothfuss and Javaux, 2017).

In this study, only two end members were considered: piston flow and preferential flow (detail described in Section 3.1). In the framework, stable isotopes in soil water (average and standard error) and precipitation (volume weighted average and standard error) were used to represent the isotopic features of piston flow and preferential flow, respectively. Groundwater water isotopes of different samples (n = 43) were considered independent consumers (input) as they were collected from different sites. The error structure was set to default, “Resid*Process” (Stock and Semmens, 2016) and the Markov Chain Monte Carlo length was set to “very long” for improved prediction of the probability distributions of sources (e.g. soil water and precipitation). As the stable isotopes of recharging water do not change through both piston and preferential flows (assumption described earlier), the discrimination factor was set to zero. Because there was no prior information, the “Uninformative”/Generalist was selected in this framework. After running the MixSIAR, a check diagnostic was conducted according to Stock and Semmens (2013) for confirming the validity of the output (contributions). Finally, the mean values of the runs were considered as the most likely proportions of contributions and the standard deviation was considered as the uncertainty.

3. Results

3.1. Stable isotopes in precipitation, soil water, and groundwater

There was large variation in precipitation stable isotopes (Fig. 2a), ranging from −19.6‰ to −1.1‰ for δ^{18}O and from −142.0‰ to 1.1‰ for δ^2H, with the average of −9.7‰ and −66.7‰ for δ^{18}O and δ^2H, respectively (Table 1). In contrast, the stable isotope values of soil water and groundwater exhibited small variation (Fig. 2a). The δ^{18}O values ranged from −11.0‰ to −8.4‰ and the δ^2H values ranged from −80.8‰ to −64.6‰ for soil water; for groundwater, the δ^{18}O values ranged from −10.5‰ to −9.4‰ and the δ^2H values ranged from −75.0‰ to −68.7‰ (Table 1).

The Local Meteoric Water Line (LMWL) had a slope of 7.45 and an intercept of 5.52 (Fig. 2a–b). These were smaller than the slope (8) and the intercept (10) of the Global Meteoric Water Line (GMWL, developed by Craig (1961)), indicating some evaporation occurred during rainfall.

Table 1
The minimum, maximum, average and standard error of stable isotopes in precipitation, deep soil water (8–25 m), and groundwater.

Water types		Precipitation[a]	Soil water	Groundwater
No. of samples		200	212	43
$\delta^{18}O$ (‰)	Min	−19.6	−11.0	−10.5
	Max	−1.1	−8.4	−9.4
	Average	−9.7	−9.8	−10.1
	SE	0.31	0.03	0.04
$\delta^{2}H$ (‰)	Min	−142.0	−80.8	−75.0
	Max	1.1	−64.6	−68.7
	Average	−66.7	−73.5	−72.7
	SE	4.71	0.25	0.18
lc-excess (‰)	Min	−24.6	−12.7	−6.6
	Max	−18.8	−0.6	0.2
	Average	0	−6.1	−2.6
	SE	0.54	0.15	0.22

[a] The average and standard error (SE) are volume-weighted values, respectively.

Soil water and groundwater fell on the right-hand side of the LMWL and varied along with a trend line respectively (Fig. 2b), suggesting that they both originated from local precipitation. The lc-excess values of deep soil water and groundwater had an average of −6.1 and −2.6, respectively (Table 1; Fig. 3). Student *t*-test showed that the groundwater had a significantly smaller average lc-excess value ($P < 0.05$) than the soil water. This further indicated that the groundwater experienced less evaporation than soil water.

The groundwater had average $\delta^{18}O$ and $\delta^{2}H$ values of −10.1‰ and −72.7‰, respectively (Table 1). They were significantly lower than those of the long-term monitored values ($\delta^{18}O$ of −9.7‰ and $\delta^{2}H$ of −66.7‰) in precipitation at Changwu station ($P < 0.05$). This phenomenon can be attributed to the selective infiltration of precipitation from isotopically depleted seasonal precipitation or rainfall events (Jasechko et al., 2014).

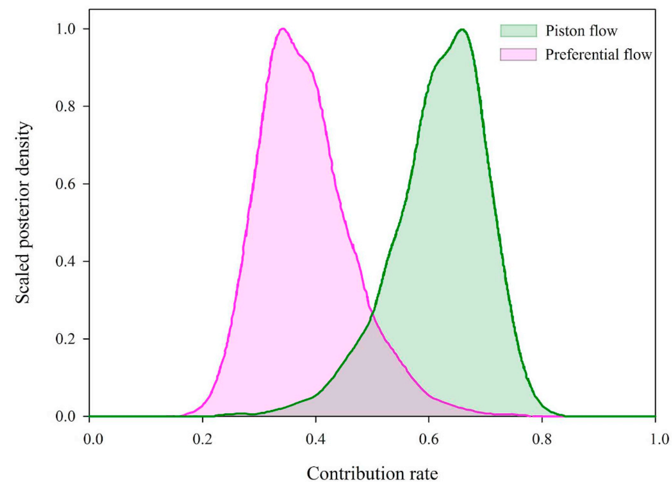

Fig. 4. Isotope-based estimation of relative contribution rate for piston and preferential flows using Bayesian mixing model (MixSIAR).

3.2. Contributions of piston and preferential flow to groundwater recharge

The relative contribution rates of piston and preferential flows were quantified using the Bayesian stable isotope mixing model (Fig. 4). Overall, piston flow contributed 62% ± 8% (average ± SD) to the total recharge and the preferential flow was 38% ± 8%. The difference between contributions from piston and preferential flow is statistically significant ($P < 0.05$), further confirming the dominance of piston flow to recharge. The narrow peaks, smooth curves, and relatively small standard deviation (8%) indicated that our isotopic data and methods are appropriate for apportioning the recharge components between the piston and preferential flows.

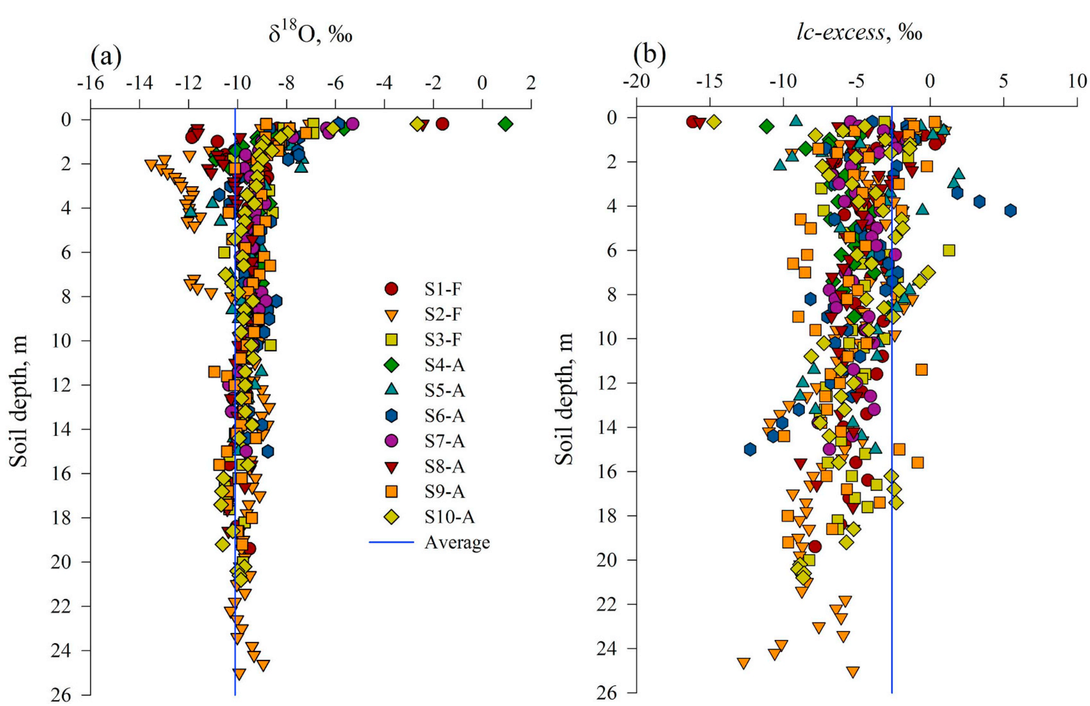

Fig. 3. Vertical distribution of $\delta^{18}O$ (a) and lc-excess (b) of soil water. In the legend, S1 to S10 represent the sampling sites. The land use is marked following the sampling site with F for farmland and A for apple orchard. The vertical blue line represents the average values of groundwater ($\delta^{18}O$ = −10.1‰ and lc-excess = −2.6‰). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4. Discussion

4.1. Piston flow and preferential flow coexist within highlands of the Loess Plateau

On the highlands of the CLP, local precipitation is the only recharge source for shallow groundwater (Huang et al., 2017a; Li et al., 2017; Tan et al., 2016). If shallow groundwater was only recharged by piston flow from deep soil water percolation, its stable isotopes should be similar to that of deep soil water. However, our results showed that the stable isotopes of shallow groundwater differed from those of deep soil water, as showed by lc-excess (Table 1 and Figs. 2–3). Even though our groundwater sampling sites are seemingly far from the soil sampling sites (Fig. 1), there were only small variations (between sites) in isotopic compositions of deep soil water and groundwater (Table 1 and Fig. 3). This is also supported by Li et al. (2017) and Huang et al. (2017a) at landscape or watershed scales. Similarly, a deep soil core (98 m) at Changwu highland showed that the isotopic compositions of soil water are stationary below 8 m (unpublished data), suggesting little change in groundwater recharge (piston flow) during the last 1000 years or so. This indicated that shallow soil water samples between 8 and 25 m (Fig. 3) had similar isotopic signature as deep soil water (25 m below or close to water table) and as the deep soil water that enters groundwater. This is also supported by Cheng et al. (2014) and Tan et al. (2017). Overall, this robust evidence suggested that the sampling sites are representative and the methodology adopted in this study is valid.

In fact, the isotopic composition of shallow groundwater was significantly lower than those of the long-term monitored precipitation values ($\delta^{18}O = -9.7‰$, $\delta^{2}H = -66.7‰$) and was between that of deep soil water and that of precipitation (Fig. 2b). This phenomenon in arid and semi-arid regions was widely observed and could be attributed to three mechanisms (Leaney et al., 2003): (1) recharge received from nearby locations and moved laterally in the direction of the hydraulic gradient to its current location; (2) recharge from past, wetter climatic periods; and (3) the selective infiltration of precipitation from isotopically depleted seasonal precipitation or rainfall events. At our study site, as gullies deeply cut through the bedrock, highlands are isolated as islands (Fig. 1d). As such, the groundwater is deeply buried 30–100 m below and its hydraulic potential decreases from the center to the edge. Consequently, the river water located in gullies will not be able to move horizontally to recharge the groundwater below the highland. Further, we measured nitrate concentration of groundwater in all five sites and the tritium concentration of groundwater in two sites (2 of 5). The nitrate concentration of groundwater was higher than 3.0 mg L^{-1} and the average values range from 6.5 mg L^{-1} at Changwu site to 24.1 mg L^{-1} at Qianshui site (Fig. 5). Meanwhile, the groundwater has a lower but detectable tritium concentration: the deeper (100 m below the ground) one is 2.5 TU whereas the shallower (32 m below the ground) is 4.3 TU. These high nitrate and tritium concentrations in groundwater suggested that even though the ^{14}C age ranges from 136 to 23,412 years (Huang et al., 2017a), the groundwater should contain a portion of water that is younger (< 60 years). As nitrate concentration of soil water below 8 m was not detectable (Huang et al., 2017a; Huang et al., 2018), the younger water is likely a result of the fast recharge that bypasses deep soil matrix and ultimately recharges the shallow groundwater, which may be the other recharge mechanism for groundwater. The fast recharge could be possible from the presence of preferential flow pathway that allowed direct or selective infiltration of precipitation water to the groundwater, which was discussed in detail in Section 4.3. Therefore, the groundwater recharge mechanism in the Changwu highland may be a dual model: piston flow and preferential flow, which was consistent with previous isotope study by Cheng et al. (2014) and chloride study by Li et al. (2017).

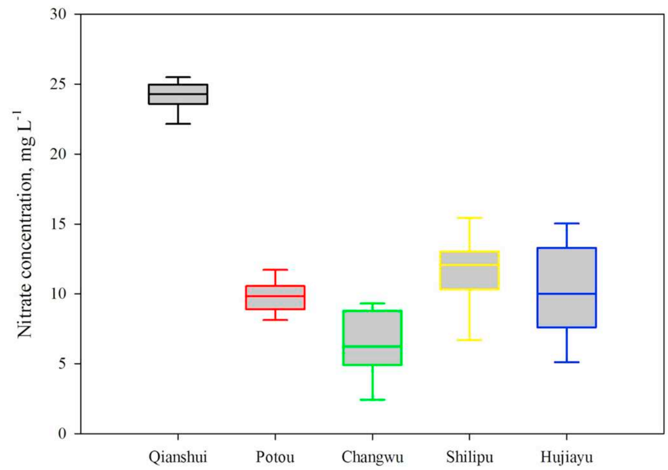

Fig. 5. Nitrate concentration of groundwater for five sites at Changwu highland.

4.2. Piston flow is the dominant groundwater recharge mechanism on the Loess Plateau

In the Changwu highland, the stable isotopes indicated that, in addition to piston flow, a portion of precipitation water may bypass soil water and recharge the groundwater directly through preferential flow pathway. Our results showed that the contribution of piston flow (62% ± 8%) to the total recharge was significantly (t-test, $P < 0.05$) larger than that of the preferential flow (38% ± 8%) (Fig. 3). This implied that piston flow is the dominant recharge mechanism. Our conclusion was consistent with previous studies conducted at other CLP sites. For example, many researchers investigated tritium, chloride, and nitrate in pore water at Changwu, Pingding, Guyuan, Xifeng, and Luochuan Counties (Huang and Pang, 2011; Huang et al., 2013; Huang et al., 2016; Huang et al., 2018; Li et al., 2018; Lin and Wei, 2006; Zhang et al., 1990; Zhang et al., 2017). These studies all suggested the dominance of piston flow over preferential flow in the unsaturated zone. Recently, Huang et al. (2017a) obtained similar conclusion using multiple tracers within the deep unsaturated zone and saturated zone in a nearby highland (18 km east of the study area).

As stated above, the piston flow is the dominant flow mechanism on the CLP. This may be because: First, the loess is a terrestrial clastic sediment, which is formed by accumulation of windblown dust (Pye, 1995), resulting in relatively homogeneous soil texture; Second, silt (0.002–0.05 mm), sand (0.05–2 mm) and then clay (< 0.002 mm) contents of loess are, 64–68%, 20–24%, and 12–13%, respectively (Zhao et al., 2016), suggesting loess is relatively coarse. The relatively coarse and homogeneous soil material is an ideal media for piston flow.

4.3. Why there is preferential flow?

The loess is an ideal medium for piston flow and plenty of results had also verified the dominance of piston flow over preferential flow; however, our result showed that the preferential flow was a non-negligible recharge mechanism as it accounted for 38% of the total groundwater recharge. This induced a critical question, that is, why there is preferential flow? According to massive early efforts (Beven and Germann, 2013; Jarvis et al., 2016; Jarvis, 2007; Lin, 2010), the potential for preferential flow occurrence in field soil is likely universal, but its occurrence is highly dynamic in space and time. Wiekenkamp et al. (2016) have found that the generation of preferential flow is poorly correlated with spatial parameters, but strongly depends on precipitation amount, small-scale soil and biological features, and

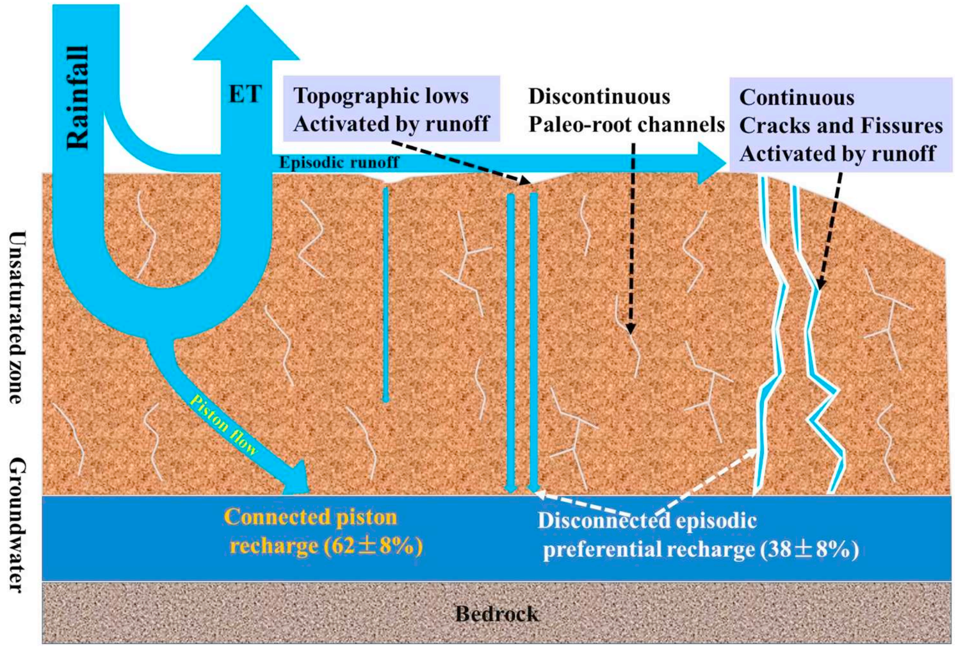

Fig. 6. A conceptual model for piston and preferential recharge on the highlands of Chinese Loess Plateau. Disconnected preferential recharge occurs episodically under localized topographic lows and in continuous cracks, while connected piston flow recharge occurs regularly in soil matrix.

localized processes. Therefore, the conditions for generating preferential flow can be grouped into two categories: (1) precipitation characteristics; and (2) preferential pathways.

On the Changwu highland, the climate is a temperate continental monsoon, and thus the most of precipitation events are light rainfall. However, intense rainfall events do occur in summer (July to September) that accounts for > 55% of the total annual precipitation (Liu et al., 2010). Because of the high infiltration capacity of loess (Duan et al., 2016; Li, 2001; Wu et al., 2017), a large proportion of small rainfall generally enters soil quickly, unlikely to generate surface runoff through saturation excess or infiltration excess. However, long-term observation (1957–2006) of rainfall events and surface runoff suggested that, only a few intense rainfall events (≥25 mm/day) are conducive to generating surface runoff (Chen et al., 2009). The extreme rainfall events and the resulting runoff create saturated soil surface and pooling of runoff water at topographic lows (Fig. 6), conditions that are conducive to preferential flow. Consequently, precipitation events ≥ 25 mm/day can be considered as the threshold to generating preferential flow in this study. In additional, the isotopic composition of precipitation showed an "amount effect" and also exhibited seasonal variation (Cheng et al., 2014). Thus, the intense rainfall (≥25 mm/day) have a weighted-average $\delta^{18}O$ and $\delta^{2}H$ values of −10.7‰ and −75.0‰, respectively. These isotopic signatures are more depleted than that of groundwater. Therefore, the relatively depleted isotopic composition of groundwater could be explained and the small ratio (38%) of preferential flow triggered by rarely intense rainfall was reasonable.

Preferential pathways can be grouped into two categories (Fig. 6): (1) focused infiltration through topographic lows such as depressions and sink holes; that is, the topographic lows serve as the pools for the runoff water that generated during rainstorm (Gates et al., 2011; Yan, 1986); and (2) preferential paths through macro-pores, cracks, and fissures. The cracks and fissures are highly visible at the edge of the highlands and inside of the soil (Yan, 1986). There are also abundant root channels within the deep soil profile (Fig. 6). These preferential pathways may differ in their respective connectivity: The cracks and fissures at the edge of the flat plateau are highly connected and if receiving runoff water and extreme rainfall, these cracks and fissures are hydrologically activated. However, the cracks and root channels within the soil are sparsely distributed and discontinuous, and therefore are rarely activated in the deep unsaturated zone (Fig. 6). This may explain why there are overwhelming evidence supporting predominant piston flow recharge and there is little preferential flow recharge at the point scale found on the Chinese loess plateau; but there is substantial preferential flow recharge at the landscape and watershed scales (Gates et al., 2011; Li et al., 2017).

4.4. Difference in groundwater recharge mechanism on the Loess Plateau

The preferential flow accounted for 38% of the total groundwater recharge in the Changwu highland (Fig. 4). The contribution was substantially more than that at the point scales (although not quantified in field) as indicated by previous tritium studies (Li et al., 2016; Lin and Wei, 2006; Zhang et al., 1990; Zhang et al., 2017); Moreover, the contribution is much smaller than that at the watershed scale as quantified by Li et al. (2017), who concluded that the contribution of preferential flow as high as 87% of the total recharge at Heihe watershed (at a similar site with this study). These indicated that the contribution of preferential flow is likely a function of the spatial scales and the contribution increases with the increase of scale from point to landscape (current study) to watershed. This may be because the heterogeneity and similarity in hydrology manifests itself at multiple spatial scales (Famiglietti and Wood, 1994; McDonnell and Beven, 2014; Peters-Lidard et al., 2017). Nevertheless, the difference in contributions (87% in Li et al. (2017) vs 38% in our study) created an open question on the effect of scale on the dominant recharge mechanisms, which requires further investigation covering multiple spatial scales.

Additionally, studies conducted at hilly region (Gates et al., 2011) and high-mountains (Tan et al., 2016) within the CLP indicated that preferential flow might be the main groundwater recharge mechanism. Though the contribution of preferential flow was not quantified in these regions, the quick and significant groundwater response to the precipitation through water table fluctuation, stable isotopes, or high concentration of tritium, indicated the dominance of preferential flow. Thus, it implied that the dominant groundwater recharge mechanism may also change with the topographic variations within the CLP, which induces a critical consideration in modeling regional groundwater recharge processes.

Different from previous study, we used lc-excess to apportion the groundwater in this study. For a precipitation-derived groundwater system, the formation of groundwater is a mixture of two sources: soil water and precipitation. The difference between soil water and precipitation is that soil water is evaporation-modified precipitation. Therefore, to accurately isolate the effect of evaporation is the key to source the groundwater using water isotopes. As such, lc-excess is better to isolate the influence of evaporation than single water isotopes (Evaristo et al., 2015; Sprenger et al., 2017). Furthermore, we used the Bayesian framework (MixSIAR) in groundwater sourcing and took uncertainties of the precipitation, soil water and groundwater into consideration. Our study is the first attempt, to the best of our knowledge, to integrate lc-excess with a Bayesian framework in sourcing the groundwater.

4.5. Implications for groundwater management on the Loess Plateau

In the terrestrial water cycle system, piston flow is the connected water, but the preferential flow is the disconnected water (Good et al., 2015). Hence, the hydrologic connectivity is the proportion of piston flow to the total flow, because in a fully connected system the water movement is assumed to be solely by piston flow. Consequently, the contribution of piston flow determines the degree of hydrologic connectivity in the system. The high contribution of piston flow to the total water flow suggested that there was strong hydrologic connectivity between soil water and groundwater in the highlands of the CLP. Nevertheless, the identified hydrologic connectivity here had important implications to: (1) groundwater resources management and (2) groundwater quantity risk evaluation.

Shallow groundwater of the CLP is an important fresh water resource but it has suffered from over-exploitation during last few decades, which resulted in severe groundwater resources shortage (Gao et al., 2015). Numerous studies have evaluated the groundwater recharge rate from piston flow across the CLP (Deng et al., 2015; Gates et al., 2011; Huang and Gallichand, 2006; Huang and Pang, 2011; Huang et al., 2013; Huang et al., 2017a; Huang et al., 2017b; Huang et al., 2016; Li et al., 2016; Zhang et al., 2007), but few of these have considered about the presence of preferential flow, which resulted in incomplete estimation of the total groundwater sources. As the preferential flow accounted for 38% of the total recharge on highlands and might be affected by spatial scales and topographic variation, the hydrologic connectivity or groundwater recharge mechanism should be considered in thorough assessment of the total groundwater sources.

The solutes originated from over-fertilization over last three decades were accumulated within the upper unsaturated zone (0–6 m) on the CLP (Huang et al., 2013; Huang et al., 2016; Huang et al., 2018). The high concentration of solutes may ultimately contaminate groundwater as it can be flushed into groundwater by piston flow, leading to groundwater salinization and water quality deterioration. According to a fully connected system, Huang et al. (2013) estimated that after about 820 years, the groundwater solute (chloride) will double its current concentrations (from 7.7 to 15.4 mg L^{-1}). We showed that the system is well connected, but is not fully connected on the highlands of the CLP. Therefore, if piston flow is the main cause of the risk, the shallow groundwater biogeochemistry might be dominated by the unsaturated zone hydrological processes, and the above evaluation may over-estimate the potential risk of groundwater quality as it may be affected by the contributions from fast preferential flow. However, if preferential flow is the problem, at small area where preferential pathways exist, substantial amount of surface runoff water can be conducted rapidly to deep subsurface or groundwater. This will create groundwater recharge hotspots, and in some cases, the groundwater pollution hotpots if the runoff water is contaminated. Therefore, careful consideration of the hydrological connectivity or groundwater recharge mechanisms should be placed in effect when evaluating the risks of pollutant to groundwater quality on the CLP and other regions of the world.

5. Conclusion

Stable isotopes (δ^2H and $\delta^{18}O$) of precipitation, soil water, and groundwater were used to determine the groundwater recharge mechanism in the Changwu highland of the CLP. The stable isotopes of shallow groundwater fell within that of the mixing area between precipitation and soil water. This indicated that the groundwater recharge processes had a dual flow mode, piston flow and preferential flow. Based on a Bayesian framework (MixSIAR), the relative contribution of piston flow to the total recharge was estimated to be 62% ± 8%, while that of preferential flow was 38% ± 8%, suggesting the dominance of piston flow as the groundwater recharge mechanism. The high contribution of piston flow indicated the well hydrologic connectivity. These findings are critical for understanding hydrological process and evaluating groundwater quantity and quality in highlands of the CLP and other regions with similar hydrogeological conditions.

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