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Stable isotopes applied as water tracers for infiltration experiment

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Abstract The δD and δ^{18}O vertical profiles of soil water were measured prior to and after a rainfall event. Mechanisms of soil water movement were deciphered by comparing the soil water isotope profiles with the isotopic composition of precipitation. The results show that evaporation at the upper depth led to enrichment of the heavy isotopes. Compared to the loess profile, the shallow soil water of sand profile is relatively enriched in D and ^{18}O due to macro-pore and low water-holding capacity. The precipitation is infiltrated into soil in piston mode, accompanied with significant mixing of older soil water. The preferential fluid flow in loess was observed at depths of 0–20 cm, caused by cracks in the depths. The hydrogen and oxygen isotopic compositions in outflow are close to the precipitation, which shows a mixing of the precipitation and old soil water, and indicates that the isotopic composition of outflow water is mainly controlled by that of the precipitation. The δD and δ^{18}O in outflow decreased with time until stable δ values of outflow are close to those of the precipitation.

Key words Precipitation infiltration, Stable deuterium and oxygen isotopes, Soil water, Piston flow

1 Introduction

Water cycle consists of rainfall, surface water, soil water and groundwater. Soil water as a dissolved nutrient of plants and a reservoir plays an important role in the hydrologic cycle. Understanding water movement in the unsaturated zone is essential for describing transport of chemical weathering products, carbon and other nutrient cycle, which are strongly tied to hydrological conditions[1–2].

Stable isotopes as tracers in hydrology are ubiquitous and natural environment components without artificial substances. In particular, ^{18}O and D are ideal tracers of water flow to reveal information about infiltration, evaporation and percolation, which can hardly be achieved by other techniques[3]. Then, knowledge of factors affecting isotopic compositions of soil water is a must for above applications.

Rainfall infiltration, which is an important and basic process[4], affects the isotopic compositions of soil water, and rainfall infiltration and subsequent downward percolation are complicated by soil texture, initial soil water contents, and topographic characteristics[5]. According to a previous scenario, which is described as piston flow, the water from precipitation forces the older soil water to flow downward. Carey *et al.*[6] and Jessica *et al.*[7] compared stable isotopic composition of soil water with precipitation to study the recharge rates. Van Omment *et al.*[8] and Vincent *et al.*[9] did soil column experiments and clarified that the preferential fluid flow was mainly caused by cracks in dried soils, decayed plant roots, earthworm burrows, and rocks. However, many studies found no evidence of isotope fractionation during soil water movement [10–12].

In China, the precipitation infiltration studies using isotopic tracing methods was mainly focused on the isotopic characteristics of soil water at different depths[13–15], but relationships among the isotopic compositions of precipitation, soil water and groundwater are less reported. This study is aimed at examining changes in isotopic compositions of soil

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water after a rainfall event, and exploring mechanisms of soil water movement by comparing the isotopic compositions of precipitation with soil water in sand and loess. After a rainfall event, δD and δ^{18}O of soil water were enriched slightly compared to the rainwater. The rainwater infiltration into the soil was accompanied with a significant mixing of older soil water, and preferential fluid flows were observed in the loess. The δD and δ^{18}O values of the outflow are close to the precipitation, which is a key factor controlling the outflow.

2 Experimental

2.1 Materials and Methods

All experiments were performed with sand and loess, and their grading curves are shown in Fig.1. Four polyvinyl chloride columns of Φ11 cm × 75 cm were packed with air-dried sand or loess, and were assigned into sand (S1 and S2) and loess (T1 and T2) columns. The sand and loess in lifts of approximately 5 cm were added each time, without compacting. The average dry densities of sand and loess columns were 1.58 and 1.32 g/cm^3, respectively. The base (about 5 cm) of columns was packed with gravel to allow free drainage of water. Each column was run through by about 2.5 times pore volumes of tap water, allowed to drain for 24h, and the column bottoms were sealed for water evaporation of 8 months. The control soil samples (Profile **a** and **b**) were collected from S1 and T1 at depths of 5, 10, 15, 20, 30, 40, 50 and 60 cm.

The S2 and T2 columns were run through by tap water (700 mm for S2, and 1000 mm for T2) with –56‰ of δD and –7.8‰ of δ^{18}O, and the outflow water in 20- mL interval was used for isotopic analysis. Without outflow, soil samples (Profile **a** and **b**) at the same depths as above were collected after rainfall event. Soil water was extracted from soil samples by vacuum distillation for isotopic analysis.

2.2 Measurements of isotopic compositions

The δD and δ^{18}O were measured by MAT253 mass spectrometer using the H_2O equilibration technique. The analytical precisions of δ^{18}O and δD measurements were 0.1 and 2‰, respectively. The results were reported as per mil differences (δ) relative to standard values of V-smow,

$$\delta^{18}O/\delta D = [(R_{\text{sample}}-R_{\text{smow}})/R_{\text{smow}}]\times 1000$$

where R stands for the ratio of D/H or $^{18}O/^{16}O$. The experimental results are given in Tables 1 and 2.

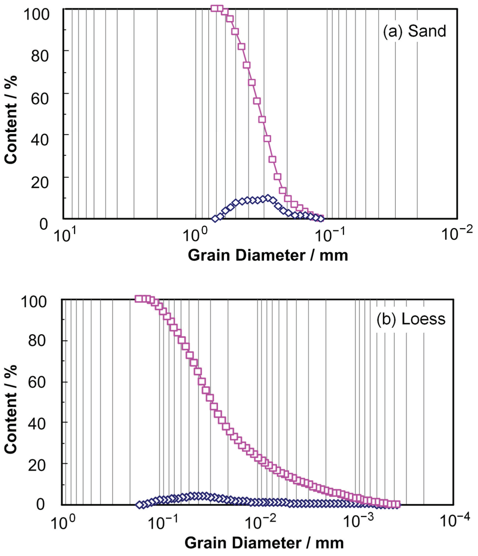

Fig.1 Gradation curve of pure sand and loess.

Table 1 Isotopic compositions of soil water.

Depth	Profile a			Profile b			Profile c			Profile d		
/ cm	Water / %	δD / ‰	δ^{18}O / ‰	Water / %	δD / ‰	δ^{18}O / ‰	Water / %	δD / ‰	δ^{18}O / ‰	Water / %	δD / ‰	δ^{18}O / ‰
0	10.01	–45	–5.2	0.40	–20	–0.8	27.81	–56	–7.4	3.46	–54	–6.8
5	12.43	–44	–5.0	1.62	–30	–1.6	27.81	–57	–7.8	4.14	–56	–7.1
10	14.04	–48	–6.4	2.41	–38	–3.0	27.19	–55	–7.4	4.04	–57	–8.0
15	14.92	–47	–6.5	1.72	–38	–2.9	27.60	–54	–7.2	4.72	–58	–8.1
20	15.31	–47	–6.4	2.56	–41	–4.9	29.16	–57	–7.5	5.11	–56	–7.4
30	16.24	–56	–7.5	4.20	–53	–6.3	30.61	–53	–7.2	5.53	–55	–7.2
40	16.87	–54	–6.3	4.68	–53	–7.1	33.50	–55	–6.5	11.83	–54	–6.9
50	16.94	–51	–6.5	10.48	–54	–6.8	30.12	–52	–7.3	20.71	–53	–6.7
60	17.13	–50	–6.6	16.67	–54	–7.3	31.32	–50	–7.0	24.46	–53	–6.9

* (a) Initial loess profile, (b) Initial sand profile, (c) Loess profile after a rainfall event, and (d) Sand profile after a rainfall event.

Table 2 Isotopic compositions of outflow water.

Sand	Sample No	1	2	3	4	5	6	7	8	9	10	11	12	13						
	δ^{18}O / ‰	−7.9	−8.9	−7.2	−7.1	−8.3	−6.6	−8.1	−7.5	−5.7	−7.6	−7.6	−6.8	−6.9						
	Volume / mL	25	25	25	25	25	25	25	25	25	25	25	25	18						
Loess	Sample No	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32
	δ^{18}O / ‰	−7.3	−7.0	−6.8	−7.0	−6.6	−7.3	−8.6	−7.6	−7.7	−7.4	−7.4	−7.5	−7.6	−7.3	−7.6	−6.9	−7.2	−7.4	−7.3
	Volume / mL	24	25	25	25	8.4	8.4	8.4	8.4	8.4	8.4	8.4	8.4	8.4	8.4	8.4	25	25	25	5

3 Results and Discussion

3.1 Isotopic characteristics of soil profiles

It is known that heavy isotopes can be enriched in free water due to evaporation fractionation[16]. Sun *et al.*[17] reported that soil water followed the same fractionation mode as free water. As shown in Fig.2, the vertical isotopic profiles of S1 and T1 indicate increased δD and δ^{18}O of soil water in the upper 30 cm. The enrichments can be a result of multiple processes, including evaporation, mixing of new and old water and change in isotopic compositions of precipitation[8], but S1 and T1 did not have rainfall events during the 8-months evaporation. Then, the enrichment in the top 30 cm of soil was due to evaporation.

From Fig.2(a), the δD and δ^{18}O in the soil water at depths of 0–10 cm of S2 after the rainfall event were slightly more than those in rainwater, and were a bit less than those of S1 at the same depths. The S2 profile was sampled on the next day after the rainfall event, and the δD and δ^{18}O differences between the soil water and rainwater could not be caused by evaporation. Further evidence is given by the T2 profile, in which the surface soil water has δD and δ^{18}O values similar to those of rainwater. Therefore, the difference is the result of mixing of the rainwater and the soil water existed prior to the rainfall event, which is called ‘old soil water’. The similar δD and δ^{18}O values of soil water in the S2 profile and the rainwater might be due to piston effect at depths of 10–15 cm, i.e. the mobile soil water displaces the old soil water and pushes it downward.

However, the situation with loess seemed different. With a piston flow, the rainwater in loess would have replenished the old soil water, and the δD and δ^{18}O in the soil water after the rainfall event would have been similar to those of the rainwater, but these could not be observed in T2 profile (Fig.2b). The δD and δ^{18}O values of the soil water at depths 5–20 cm (except at depths of 10–15 cm) is of about the same, and are similar to those of the rainwater. Because the crack growth depth is about 20 cm, this can be a trend of preferential fluid flow along the depth of 5 to 20 cm in T2 profile. The cracks provide a preferential pathway of precipitation infiltration from soil surface to deep depths.

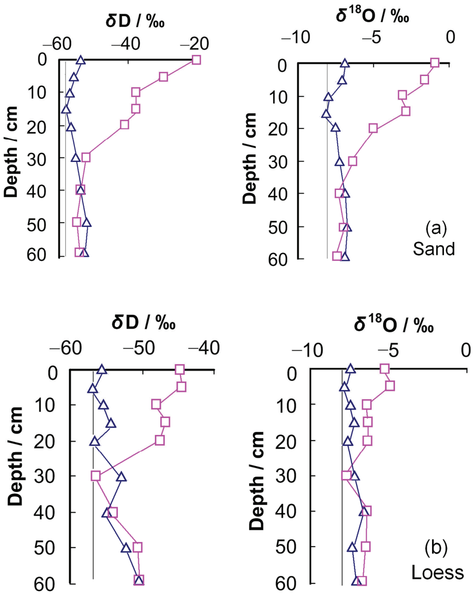

Fig.2 δ profiles *vs* depth of sand and loess before (□, S1 and T1) and after (Δ, S2 and T2) a rainfall event. The vertical line indicates the isotopic compositions of precipitation.

The δD and δ^{18}O of old soil water at different depths after precipitation can be estimated. Assuming that the old soil water is immobile and the δ difference ($\Delta\delta$) of water content between S1 and S2 (or T1 and T2) at any depth is due to the rainwater recharging, the δD and δ^{18}O values of soil water in S2 and T2 profiles were estimated by mass balance. The results are given in Table 3, where one finds a trend that $\Delta\delta = \delta_{\text{calculated}} - \delta_{\text{measured}} > 0$ in soil water at 0–20 cm of T2 and 0–30 cm of S2, and $\Delta\delta < 0$ in larger depths of the profiles. Because D and ^{18}O are more depleted in the rainwater than in the old soil water, the $\Delta\delta > 0$ results may be caused by the large proportion of old soil water used in the calculation, that is, during the infiltration process the old soil water was mobile and part of it could be pushed downward. The mixture of old soil water in upper part of the columns and precipitation downward percolating is thought to cause $\Delta\delta<0$ in the lower part of the columns.

3.2 Effect of soil textures on isotopic composition distribution of soil water

The S1 and T1 columns shared an identical climate, but their upper parts (30 cm) evaporated differently. The δD and δ^{18}O values in soil water of S1 are apparently greater than that of T1 (Fig.3). The differences are beyond the uncertainty of δD and δ^{18}O measurements, and exist systematically at evaporation depths. However, the difference of soil water δD and δ^{18}O between S2 and T2 is not systematic.

Soil water evaporation is affected mainly by meteorological conditions and soil texture [18]. Because of the same meteorological conditions for S1 and T1, the different δD and δ^{18}O values are caused by the soil texture. Sand grains are of mush larger sizes than loess, hence the poor water-holding ability, which can also be seen in water content of S2 and T2 profiles. According to the Rayleigh model, the smaller the proportion of remaining water is, the heavier the isotopic composition of the remaining water is[17]. Consequently, the upper part soil water in S1 has more D and ^{18}O than T2 at the same depths. The differences in lower part soil water between S1 and T1 profiles may be caused by the proportion of immobile and mobile soil water.

The δD and δ^{18}O values of T2 and S2 after rainfall event did not differ systematically along the depths. T2 and S2 had different soil texture but received identical isotopic precipitation, then the soil water δ at depths is decided by the amount of rain water arrived at groundwater level.

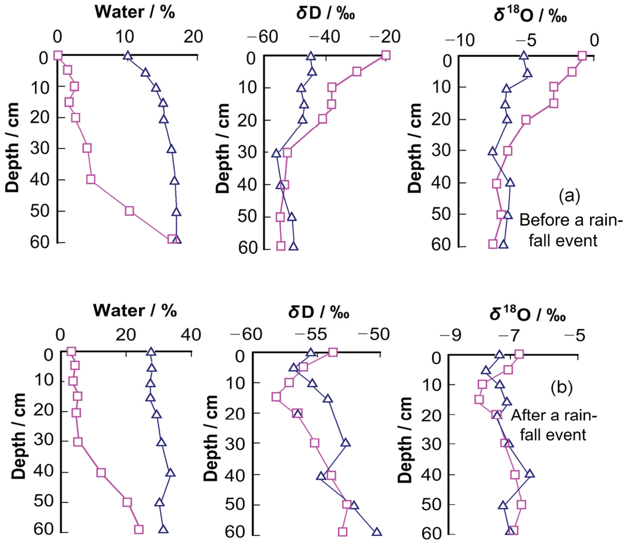

Fig.3 Vertical δD, δ^{18}O and water content profiles of sand and loess before (□: S1and S2, S1 and T1) and after (Δ: T1 and T2, S2 and T2) rainfall event.

Table 3 The measured and calculated δD and δ^{18}O values of soil water in S2 and T2 profiles. ($\Delta\delta=\delta_{\text{Calculated}}-\delta_{\text{Measured}}$).

Depth /cm	T2						S2					
	δD / ‰			δ^{18}O / ‰			δD/ ‰			δ^{18}O/ ‰		
	Measured	Calculated	$\Delta\delta$	Measured	Calculated	$\Delta\delta$	Measured	Calculated	$\Delta\delta$	Measured	Calculated	$\Delta\delta$
0	–56	–52	4	–7.4	–6.8	0.6	–54	–52	2	–6.8	–7.0	–0.2
5	–57	–51	6	–7.8	–6.5	1.3	–56	–46	10	–7.1	–5.4	1.7
10	–55	–52	3	–7.4	–7.1	0.3	–57	–45	12	–8.0	–4.9	3.1
15	–54	–51	3	–7.2	–7.1	0.1	–58	–50	8	–8.1	–6.0	2.1
20	–57	–52	5	–7.5	–7.0	0.5	–56	–49	1	–7.4	–6.4	1.0
30	–53	–56	–3	–7.2	–7.7	–0.5	–55	–53	2	–7.2	–6.7	0.5
40	–55	–55	0	–6.5	–7.1	–0.6	–54	–55	–1	–6.9	–7.5	–0.6
50	–52	–53	–1	–7.3	–7.0	0.3	–53	–55	–2	–6.7	–7.3	–0.6
60	–50	–53	–3	–7.0	–7.2	–0.2	–53	–55	–2	–6.9	–7.5	–0.6

3.3 Isotopic compositions of outflow water

The δ^{18}O values of outflow from S2 and T2 columns ranged from –8.9‰ to –5.7‰ and –.5 to –7.7‰, respectively, while the δ^{18}O of soil water in S1 and T1 profiles was –7.3 to –0.8‰ and –7.5 to –5.1‰, respectively (Fig.4). The similar δ^{18}O values of S1 and S2 are due to the high permeability of sand. However, loess is of low permeability, there were enough time for the isotopic compositions in the soil water to undergo a change, hence the increase ^{18}O in the first outflow campared to that of precipitation.

Information about soil water movement can be obtained by comparing the isotopic composition between initial soil water and outflow[19–21]. In a piston flow mechanism of precipitation infiltration, the outflow would be consistent with the isotopic variation of initial soil profile, but this consistency cannot be observed in Fig.4. Then the soil water movement in the column cannot be in the piston model.

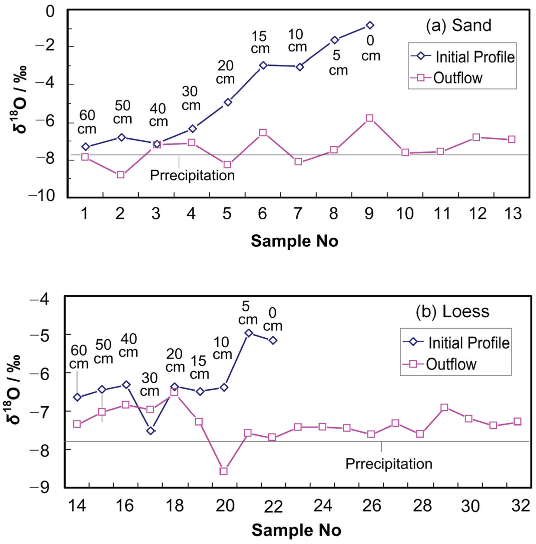

Fig.4 $\delta^{18}O$ of initial soil water and outflow water collected in 20 mL interval. Soil water No.1 is from 60-cm depth in S1, and No.9 is from surface. Similarly, No.14 soil water is from 60-cm depth in T1 and No.22 is from surface.

The relationship of δD and $\delta^{18}O$ in S1 profile was plotted in Fig.5(a), with data of the precipitation, soil water in S2 profile and the outflow water. The isotopic characteristics of initial soil water are overlapped by those of precipitation. Fig.5(b) shows relationship of the δD and $\delta^{18}O$ of the soil water in the loess profiles, indicating that the outflow water was the mixing between initial soil water and precipitation. In T2 profile, the δD and $\delta^{18}O$ of soil water in upper part of the column are similar to those of precipitation, and the values in lower part were similar to those in initial soil water. The vertical isotope distributions indicate that precipitation affected more on the upper part soil water than the lower part. The soil water at depth of 20 cm is closer to precipitation than at depths of 10–15 cm, which is a further evidence of the preferential fluid flow at depths of 0–20 cm.

The isotopic composition of groundwater recharged by precipitation is compatible with that of the annual weighted mean precipitation[22]. Still, the isotopic composition of the recharged water for individual rainfall event need be conducted. The dominant factor controlling outflow $\delta^{18}O$ can be determined by comparing the old soil water, precipitation with those of outflow. Fig.(6) shows that δD and $\delta^{18}O$ of outflow are close to precipitation, indicating that precipitation is the main recharging source of groundwater in individual rainfall event. In arid region, where evaporation is in greater levels, the soil water contains more D and ^{18}O than precipitation. Thus, the groundwater is more depleted in isotopic compositions than soil water due to the dilution effect of precipitation during infiltration processes.

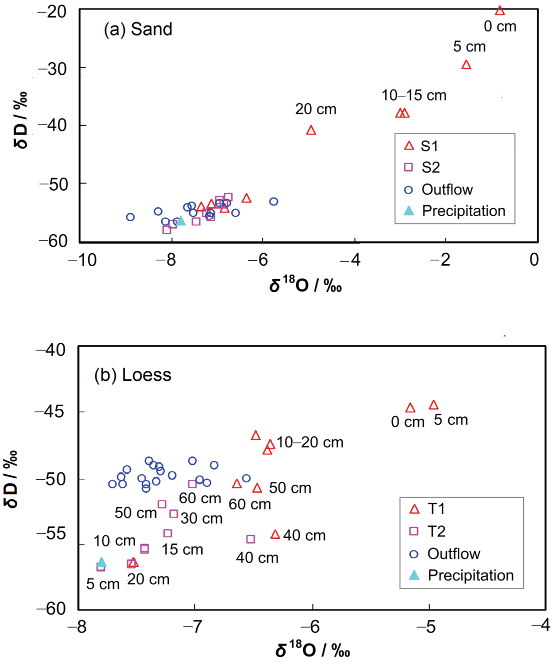

Fig.5 δD *vs* $\delta^{18}O$ relationship in precipitation, soil water and outflow water.

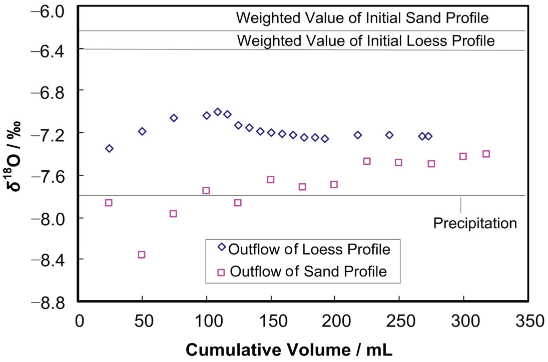

Fig.6 The $\delta^{18}O$ of outflow collected at the outlet of the column as a function of cumulative amount of the outflow. The weighted values of initial profiles were calculated using water content as weight.

4 Conclusions

The isotopic characteristics of δD and δ^{18}O in precipitation, soil water, and outflow suggest a significant mixing of precipitation with old soil water during rainfall infiltration, and preferential flow in loess profiles due to the crack presence. Consequently, mixing of precipitation with old soil water and preferential flow must be considered to implement precipitation infiltration simulation at a high certainty. And in arid region, it is necessary to study the movement mechanism before estimation of recharge rate using chloride mass balance method, which is assumed that the soil water movement is in the piston flow model.

Under the same climate conditions, isotopic composition of soil water in sand profile was more enriched than that of loess profile at soil surface due to lower water-holding capacity of sand. After a rainfall event, isotopic characteristics of precipitation are the main controlling factor for δ variation of soil water at depths under the condition of precipitation arriving at groundwater level.

The isotopic compositions of outflow, precipitation, and initial soil water show that precipitation is the key controlling factor of isotopic composition of outflow if precipitation can recharge the groundwater during a rainfall event. The first outflow sample of sand column is closer to precipitation than that of loess column due to the different velocity of precipitation infiltration.

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