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Multiple tracers reveal different groundwater recharge mechanisms in deep loess deposits

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ABSTRACT

Interpretation of groundwater recharge mechanisms is problematic because of the muted instantaneous response of subsurface water to rainfall and limited recharge rates, particularly in semi-arid environments with deep loess deposits. Here we identify the possible groundwater recharge mechanisms in 200-m thick loess deposits with unsaturated zone thickness of over 40 m. We collected soil samples up to 15 m deep under four land use types (one grassland and three apple orchards with stand ages 15, 24 and 30 years old), and used three-year precipitation and groundwater samples to determine the contents of stable water isotopes, chloride, and tritium. Our overarching goal is to determine the relative importance of piston and preferential flow in groundwater recharge using multiple tracers and quantify the effects of land use change on groundwater recharge. We find that while both piston and preferential flows are important in groundwater recharge, the unsaturated and saturated zones have yet to come to hydraulic equilibrium. This suggests different groundwater recharge mechanisms: tracers in the unsaturated zone suggest piston flow, while the detectable tritium in the saturated zone implies preferential flow. Recharge rates in the unsaturated zones range between 23 and 82 mm year^{-1}, accounting for 4%–14% of mean annual precipitation, and increasing with depth presumably because of land use and/or climatic conditions. Total recharge rate in the saturated zone is 112.6 ± 44.1 mm year^{-1}, accounting for 19 ± 9% of mean annual precipitation. Overall, our study finds that piston flow contributes more to total recharge (53%–69%) than does preferential flow. Nevertheless, piston flow may become less important because of land use change (farmland to apple orchard conversion). Our findings have implications for the need to strike a delicate balance between the economic gains from afforestation and the possible risks to groundwater supply sustainability.

1. Introduction

Understanding groundwater recharge mechanisms is important for sustainable management of groundwater resources (Gleeson et al., 2016; Jasechko et al., 2017). Achieving this ultimate goal, however, is a challenge especially in regions with thick unsaturated zones because of large potential water storage, a function of soil thickness and porosity (Camacho Suarez et al., 2015; Li et al., 2017b). The large potential water storage in these environments effectively acts as a buffer, dampening the dynamic response of groundwater to rainfall, hence, contributing to uncertainties in water balance calculations (Li et al., 2017d; McDonnell and Beven, 2014). Further, if these regions have arid climate, the resultant limited recharge rates would complicate the recharge mechanism interpretation (Allison et al., 1994; Scanlon et al., 2002). These uncertainties propagate to groundwater recharge estimation, which in turn is essential for, among others, characterizing land use change (LUC) impacts (Scanlon et al., 2007a). While LUC-related soil water deficit may be estimated around rooting zones, it is unclear to what extent LUC influences recharge rates in deep soils. It is therefore important to investigate recharge mechanisms and their relations, if any, to LUC in these regions.

Tracer-based methods are useful for recharge mechanism studies in regions with thick unsaturated zones because they "follow" the water parcel itself (McGuire and McDonnell, 2015). They are suitable for recharge estimation in arid regions because they can directly quantify small amounts of recharge (Allison et al., 1994; Scanlon et al., 2002). However, the reliability of tracer-based recharge estimation depends on tracer selection, model assumptions, and local recharge mechanism (Allison et al., 1994; Crosbie et al., 2018; Gee and Hillel, 1988; Kurylyk and MacQuarrie, 2013; Li and Si, 2018; Scanlon, 2000; Scanlon et al.,

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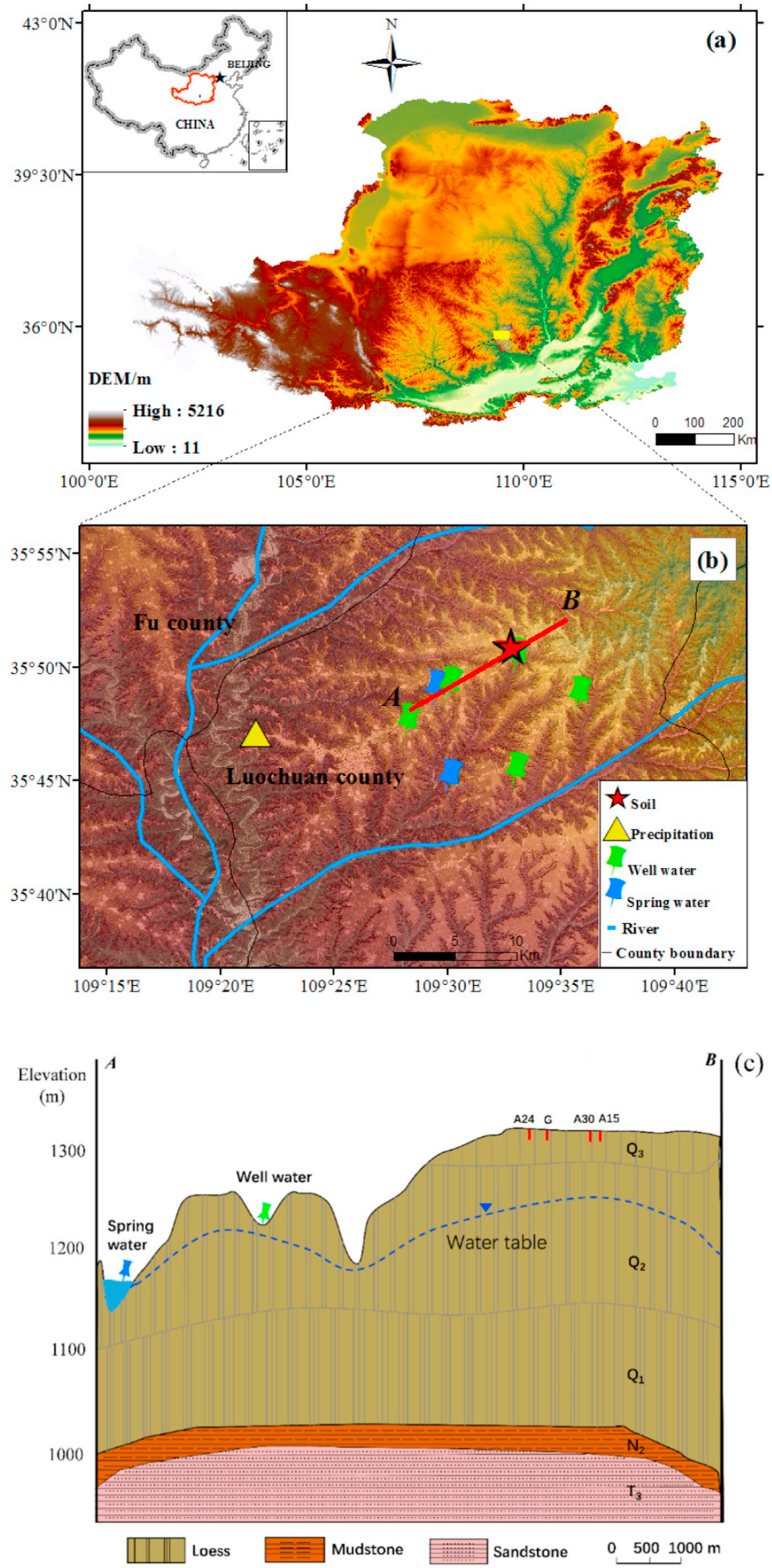

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Fig. 1. (a) Location of the study area, (b) sampling sites of precipitation, soil and groundwater and (c) hydrogeological profile. T_3, sandstone; N_2, Neogene mudstone beds; Q_1, Wucheng Loess (lower Pleistocene); Q_2, Lishi Loess (middle Pleistocene); Q_3, Malan Loess (upper Pleistocene).

2002). Conservative hydrochemical indices (e.g. chloride), stable and radioactive water isotopes (^{18}O, ^{2}H and ^{3}H) are widely used; however, their applicability varies with tracer properties. For example, ^{3}H is more suitable than ^{18}O or ^{2}H for systems with transit times over four years because the temporal variability in stable isotopes is generally small, and therefore less useful, beyond a few years (Stewart et al., 2010). Meanwhile, chloride may result in larger infiltration rates than tritium (Gvirtzman and Gorelick, 1991), while stable isotopes may indicate different surface-ground water relationships when compared with insights from hydrochemistry methods (Li et al., 2017c). These tracers provide non-unique but possibly complementary utility for recharge studies. To account for as much sources of uncertainty as possible, there is a need to employ multiple tracers and test a variety of recharge models henceforth.

The Loess Plateau is located in the middle reaches of the Yellow River, northern China with an area of 6.4×10^6 km^2, spanning 33°43′–41°16′N latitude and 100°54′–114°33′E longitude (Fig. 1). Covered by loess up to 350 m (Xiong et al., 2014; Zhu et al., 2018), the Loess Plateau is characterized by a thick vadose zone and large potential water storage (Huang et al., 2013; Li et al., 2017b; Lin and Wei, 2006; Tan et al., 2016). Results from earlier studies on groundwater recharge mechanisms in this region, mostly using stable isotopes, were inconclusive. Some suggested that groundwater is recharged by a combination of piston and preferential flows of varying relative contributions (Li et al., 2017b; Lin and Wei, 2006; Tan et al., 2017; Xiang et al., 2019), while others concluded that only piston flow is at play (Huang et al., 2013; Huang et al., 2017). The controvertible state-of-knowledge regarding groundwater recharge mechanisms in this region may simply reflect the limitations of stable isotopes alone as a tracer. Tritium may obviate many of these limitations by providing constrained estimates of water age, particularly for modern waters that are less than 50 years old (Allison and Hughes, 1978; Gleeson et al., 2016; Le Gal La Salle et al., 2001). The tritium-based methods are not as widely applicable as they used to in most regions of the world; however, recent studies suggested that these methods are still applicable in the Loess Plateau (Li et al., 2019; Li and Si, 2018; Lin and Wei, 2006; Zhang et al., 2018). And while earlier studies using tritium focused on either soil water movement in the unsaturated zones (Li et al., 2018; Zhang et al., 2018) or groundwater age determination in the saturated zones (Huang et al., 2013; Huang et al., 2017), integrated unsaturated-saturated zone investigations have been rare. This knowledge gap presents an opportunity for research, especially against the backdrop of possible effects of LUC on groundwater resources sustainability.

Indeed, land use at the Loess Plateau has changed in the last 50 years, particularly since the implementation of the Grain for Green Project in 1999 (Li et al., 2017a; Peng and Li, 2018). Farmlands were converted into grasslands and forestlands (e.g. apple orchards) to minimize soil erosion and stimulate economic growth. While it is known that LUC results in significant soil water depletion, runoff and river sediment reduction (Feng et al., 2016; Huang et al., 2018; Wang et al., 2016; Wang et al., 2011), it is unclear as to what extent LUC may or may not affect groundwater recharge.

This study seeks to advance our knowledge of the relationship between LUC and groundwater recharge. We achieve this overarching objective by leveraging the unique physiographic and LUC features of the Loess Plateau – large potential water storage because of thick loess deposits and changing land cover. We identify soil and groundwater recharge mechanisms by using multiple tracers, spanning both unsaturated and saturated zones. Specifically, we ask the following questions: (i) What is the recharge mechanism in the loess deposits? (ii) How does LUC influence groundwater recharge? (iii) What are the implications, if any, to groundwater resources sustainability?

2. Methods and materials

2.1. Study area

The study area is on the Luochuan loess tableland in the Loess Plateau (Fig. 1a). The tableland site is isolated from neighboring regions by gullies deeply cut under the bedrock, the elevation of which is higher than the riverbed. This topography suggests that the main route for groundwater recharge is via vertical infiltration from precipitation and neither via lateral flow nor streamflow. A detailed description of site hydrogeology is shown in Fig. 1c and described in Huang et al. (2016). Depth to water table is between 40 and 60 m, with aquifer thickness of about 100 m (Fig. 1c). Long-term (1956–2017) mean annual precipitation (P) is 608 mm, with 60% distributed as rainstorms between July and September. Mean annual potential evapotranspiration (PET) is 1009 mm, and dryness index (PET/P) is 1.7. Apple orchard farming at Luochuan County dates back 70 years, spanning an area of over 300 km^2, which is ~80% of arable lands. Over 95% of incomes of local people come from apples (Shi et al., 2013).

2.2. Sample collection and analysis

We selected four sites with different land use types: one grassland and three apple orchards. All four sites were converted from farmlands and were less than 100 m apart (Fig. 1b), sharing similar climatic conditions, soil properties and geology. The grassland was farmland but has been abandoned for 25 years. The current vegetation is dominated by local shallow-rooted grasses. Trees at the apple orchard sites were 15, 24 and 30 years old, respectively. All four sites have characteristically flat surfaces, minimal runoff, and without irrigation. We collected soil samples using a hollow-stem hand auger at a 20-cm interval in 2015. Because young apple trees have been found to only slightly affect soil moisture (Huang et al., 2018), the 15-year-old apple orchard was drilled only down to 10 m. Soils at the other three sites were drilled down to 15 m, considered deep enough to represent the rooting system, which in turn is deduced from the soil water profiles discussed in later sections. Each soil sample was divided into two subsamples. One subsample was used to determine soil water content via oven-drying method. We then used the dried soil samples to determine chloride and nitrate concentrations. The other subsample was used to extract soil water via cryogenic vacuum distillation method for determination of stable isotopic composition and tritium content. Detailed description of methods can be found in an earlier study (Li et al., 2017b).

Because soil water contents in deeper layers do not change appreciably over time (Huang et al., 2018; Li et al., 2017b; Zhang et al., 2018), we decided to obtain soil profiles once. However, we collected precipitation samples daily at one site and groundwater samples once a month from three wells for the period 2015–2017 (Fig. 1b). Total annual precipitation amounts were 488, 510 and 660 mm, respectively, with different frequency and intensity of wet events. We determined the stable isotopic composition and chloride concentrations of all precipitation and groundwater samples. We determined the tritium contents of groundwater samples from five wells and two springs to quantify the percentage of modern water with the methods presented in the following section.

Stable water isotopic composition was determined using an LGR LIWA V2 isotopic liquid water analyzer. Chloride concentrations were analyzed via ion chromatography (DIONEX ICS-1100, Thermal Fisher Scientific). Tritium was measured by an ultra-low-level scintillation counter (Quantulus 1220), with a detection limit of 2 tritium unit (TU). Soil water tritium contents were directly determined without enrichment since these were high enough, but electrolytic enrichment was

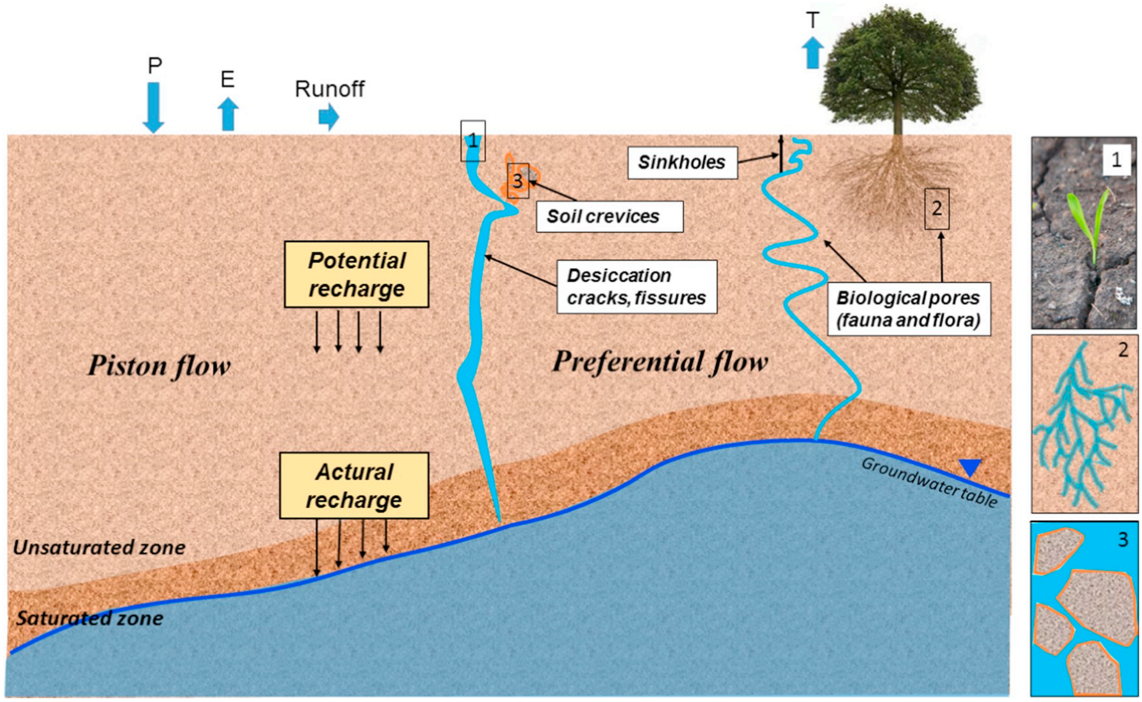

Fig. 2. Conceptual model for the two-component recharge processes. Piston flow occurs uniformly in soils, while preferential flow only occurs in some restricted areas such as cracks, fissures, biological pores and soil crevices. Only infiltration reaching the water table is actual recharge, while water that may or may not reach water table is potential recharge. P, E, T represents precipitation, evaporation and plant transpiration, respectively. 1 represents preferential flow from desiccation cracks and fissures, 2 represents preferential flow from biological pores, and 3 stands for preferential flow from soil crevices.

carried out for groundwater samples because of low tritium contents.

2.3. Estimating recharge

Qualitative and quantitative analysis were respectively carried out to investigate recharge mechanisms. Qualitative analysis was conducted by comparing tracer concentrations in precipitation, soil water and groundwater. The quantification of recharge rates was carried out for processes in different subsurface layers or in different forms shown in Fig. 2.

Based on data from different subsurface layers, recharge can be classified as potential and actual recharge (Dyck et al., 2003; Scanlon et al., 2002). Potential recharge, estimated from the unsaturated-zone data, is infiltration water that may or may not reach the water table. Actual recharge, usually estimated from groundwater data, refers to water that reaches the water table. Depending on water flow paths, recharge may pertain to piston flow and/or preferential flow (Beven and Germann, 1982; Sharma and Hughes, 1985). Piston flow refers to soil water movement whereby new infiltration water pushes the old resident soil water downward relatively uniformly within the soil matrix. The sequential propagation of 'old' resident water relative to 'new' incoming water suggests that water parcels are stacked, vertically, from the soil surface through to deeper layers with increasing ages. On the other hand, preferential flow may occur if rainwater effectively bypasses the soil matrix and directly reaches the water table via preferred pathways. The high flow velocities in preferential flow pathways suggest that preferential flow waters are 'younger' than soil matrix waters (Evaristo et al., 2019).

Our study sampled water from both unsaturated and saturated zones, estimating potential and actual recharge, respectively. These two components of our recharge model are underpinned by the presumed steady state in soil profiles, suggesting piston flow, and detectable tritium in groundwater, suggesting preferential flow.

2.3.1. Potential recharge

We used the peak depth and mass balance methods, under the assumption that soil water movement was in the form of piston flow (Allison and Hughes, 1978). If the history of tracer application is known, then the infiltration rates can be estimated by dividing moving distance with elapsed years, approximating recharge rates as the product of infiltration rates and volumetric soil water content:

$$\overline{R}_d = \frac{\int_0^{z_p} \theta(z) d_z}{\Delta t} \approx \overline{\theta}\frac{z_p}{\Delta t} \tag{1}$$

where $\overline{R}_d$ represents mean potential recharge rates in the form of piston flow, mm; z_p is the moving distance of the tracer, m; Δt is the elapsed years of tracer application; $\theta(z)$ and $\overline{\theta}$ is volumetric water content at depth z or the average value for the profile of interest, $cm^3\,cm^{-3}$.

Because the predominant source of water in our study area was via vertical infiltration from precipitation, we calculated the long-term recharge rates using the chloride mass balance (CMB) method.

$$\overline{R}_d = (\overline{P} * \overline{Cl}_p)/\overline{Cl}_{sw} \tag{2}$$

where $\overline{P}$, $\overline{Cl}_p$ and $\overline{Cl}_{sw}$ stand for mean annual precipitation (mm), mean chloride concentration in precipitation and soil water below root zones ($mg\,L^{-1}$), respectively.

2.3.2. Actual recharge

The chloride and tritium mass balance methods were both employed to calculate actual recharge (Allison and Hughes, 1978; Gleeson et al., 2016). CMB is similar to Eq. (2) except for the substitution of $\overline{Cl}_{sw}$ with mean chloride concentration in groundwater $\overline{Cl}_{gw}$. $\overline{R}$ refers to the mean total recharge, and it may come from different components.

$$\overline{R} = (\overline{P} * \overline{Cl}_p)/\overline{Cl}_{gw} \tag{3}$$

The tritium mass balance method estimates the mass proportion ($R_{modern,3H}$) of modern groundwater, defined as the ratio of m_{modern} to

m_{sample} in the groundwater (Gleeson et al., 2016).

$$R_{modern,3H} = \frac{m_{modern}}{m_{sample}} = \frac{{}^{3}H_{sample} - {}^{3}H_{old}}{{}^{3}H_{modern} - {}^{3}H_{old}} \tag{4}$$

where m_{modern}/m_{sample} is the percentage of groundwater less than 50 years within the water sample; $^3H_{sample}$ represents ^3H contents of groundwater sample, and $^3H_{old}$ and $^3H_{modern}$ represent ^3H values of groundwater that was recharged from pre-bomb (old) water and post-bomb (modern) water, respectively. $^3H_{sample}$ is directly determined from groundwater samples. $^3H_{old}$ was set to 10 TU according to Gleeson et al. (2016). $^3H_{modern}$ was estimated from reconstructed precipitation tritium contents via trend surface analysis method as recommended in an earlier study.

2.3.3. Two-component recharge estimation

If piston and preferential flows are considered as possible recharge mechanisms, then the two-component mass balance method can be used to separate the contributions of each component (Sharma and Hughes, 1985; Wood, 1999).

$$\overline{R} \bullet \overline{C}_{gw} = \overline{R}_d \bullet \overline{C}_{sw} + \overline{R}_{pf} \bullet \overline{C}_p \tag{5}$$

where $\overline{R}_{pf}$ is the average preferential flow, mm year^{-1}. $\overline{C}_{gw}$, $\overline{C}_{sw}$ *and* $\overline{C}_p$ are tracer concentrations (e.g. chloride or isotope) in groundwater, soil water and precipitation, respectively. The mean tracer concentration in preferential flow is represented by that of precipitation.

2.3.4. Estimating atmospheric chloride inputs

Long-term atmospheric chloride flux is essential for the CMB method in recharge estimation. The longest record to date, beginning in 2001, is from a site in the Acid Deposition Monitoring Network in East Asia, approximately 100 km east of our study area. Our previous study has evaluated and estimated the precipitation chloride concentration as 1.0 ± 0.4 mg L^{-1} (Li et al., 2017b). Nevertheless, this precipitation chloride dataset is too short compared with the soil water chloride records for millions of years in the thick loess.

To extend the short-term precipitation chloride observation or to evaluate the accuracy of precipitation chloride of 1.0 ± 0.4 mg L^{-1}, we inversely calculated the mean precipitation chloride for the past 52 years by using soil water chloride records. Assuming soil water moves in the form of piston flow, the atmospheric inputs of chloride and tritium would be stored in soils from lower to upper layers year by year. As soil water tritium profile corresponds to the time series of precipitation tritium contents, the peak value of soil water tritium contents should represent the peak of nuclear weapons testing activities in 1963 (Gleeson et al., 2016; Li et al., 2019; Li and Si, 2018; Scott and Richard, 2015). Consequently, the profile from the soil surface to peak depth stores tritium inputs from 1963 to the sampling year. Specific to this study, this profile is 0–8 m for the past 52 years (soil samples collected in 2015).

Simultaneously, the time t required to accumulate chloride in the 0–8 m profile can be calculated by dividing the cumulative total mass of chloride by the chloride input (Scanlon et al., 2007b).

$$t = \int_0^z \theta Cl_{sw} dz / (\overline{P} \times \overline{Cl}_p) \tag{6}$$

where t is 52 years, and the other variables except for $\overline{Cl}_p$ is known. Therefore, the chloride input or precipitation chloride concentration can be inversely calculated. As this method assumes chloride mass balance between atmospheric inputs and soil profiles, we chose the soil profile under the grassland site because of the absence of any fertilizer application at this site that would have otherwise violated the chloride method assumption.

Using the above method, precipitation chloride concentration for the past 52 years was determined as 1.3 mg L^{-1}, well within the uncertainty bounds of 1.0 ± 0.4 mg L^{-1} (mean ± standard deviation) estimated in our earlier study (Li et al., 2017b). We therefore used 1.0 ± 0.4 mg L^{-1} as the precipitation chloride concentration since it presents the uncertainty interval.

2.4. Quantifying land use change impacts on recharge

LUC impacts on recharge were quantified by using a newly developed water-chloride mass balance method. The traditional CMB (Eq. (2)) is only applicable to soil profiles not subject to root water uptake. The presence of roots requires soil profiles that are deeper than what would apply in a traditional CMB, and a correspondingly deeper profile when considering the presence of deep-rooted vegetation. To extend the applicability of this method, Zhang et al. (2018) estimated the recharge under deep-rooted plants by subtracting the recharge under shallow-rooted plants by the mean annual soil water deficit.

$$\overline{R}_{dr} = \overline{R}_{sr} - \Delta S / \Delta t \tag{7}$$

where $\overline{R}_{dr}$ and $\overline{R}_{sr}$ respectively stands for recharge under deep- and shallow-rooted plants, ΔS represents deficit of soil water storage that represents the differences in soil water storage between shallow- and deep-rooted plants, Δt represents the conversion time from shallow- to deep-rooted plants. In this study, shallow- and deep-rooted plants respectively refer to grassland and apple trees. Δt refers to the age of apple trees.

3. Results

3.1. Soil profiles

Soil water content varies with depth under each land use type (Fig. 3a), gradually increasing between 0 and 3 m, presumably because of evaporation and root water uptake near the surface, before stabilizing below 3 m. Soil water contents below 3 m decrease with increasing ages of apple tree stands, highlighting the impacts of root water uptake on soil water. The water contents abruptly increase below 8 m and 10 m under 30- and 24-year-old apple tree, respectively, suggesting older apple trees influence water content up to 8–10 m deep in the profiles. Under grassland, the water contents in 3–10 m are almost always greater than field capacity, promoting downward soil water movement and the likelihood of mixing between new and old water. Oxygen isotopic composition of soil water reflects similar patterns as soil water content (Fig. 3b), showing greater variability in shallow than in deep layers. Compared with the other sites, the isotope profile under grassland is more variable in terms of oxygen isotopic compositions.

The chloride, nitrate and tritium profiles reflect similar vertical patterns, i.e. piston-type flow with peak values at different depths (Fig. 3c–e). The peak depths are 3.4–3.8 m for both chloride and nitrate, and 8 m for tritium. The profiles and peak depths indicate the history of tracer application. Specifically, the peak depths of chloride profiles indicate the application of KCl fertilizer in 1993–1995, and those of nitrate profiles suggest the nitrate application starting in the 1980s. The tritium profile shows the signal of nuclear bomb testing between the 1950s and 1963 (Scott and Richard, 2015). The peak depths of different tracers can be used to interpret soil water movement and groundwater recharge.

3.2. Groundwater dynamics

Intra- and inter-annual variability of precipitation is evident for the period 2015–2017 (Fig. 4a). Within each year, precipitation mainly falls in July to September as rainstorms; however, the frequency and intensity of storms vary across the period illustrated. Precipitation oxygen isotopic composition and chloride concentration vary with rainfall events (Fig. 4b). But the change magnitudes of isotope and chloride concentration in groundwater are much smaller than those in precipitation, and the times with peak values are different between precipitation and groundwater (Fig. 4c), which is probably the

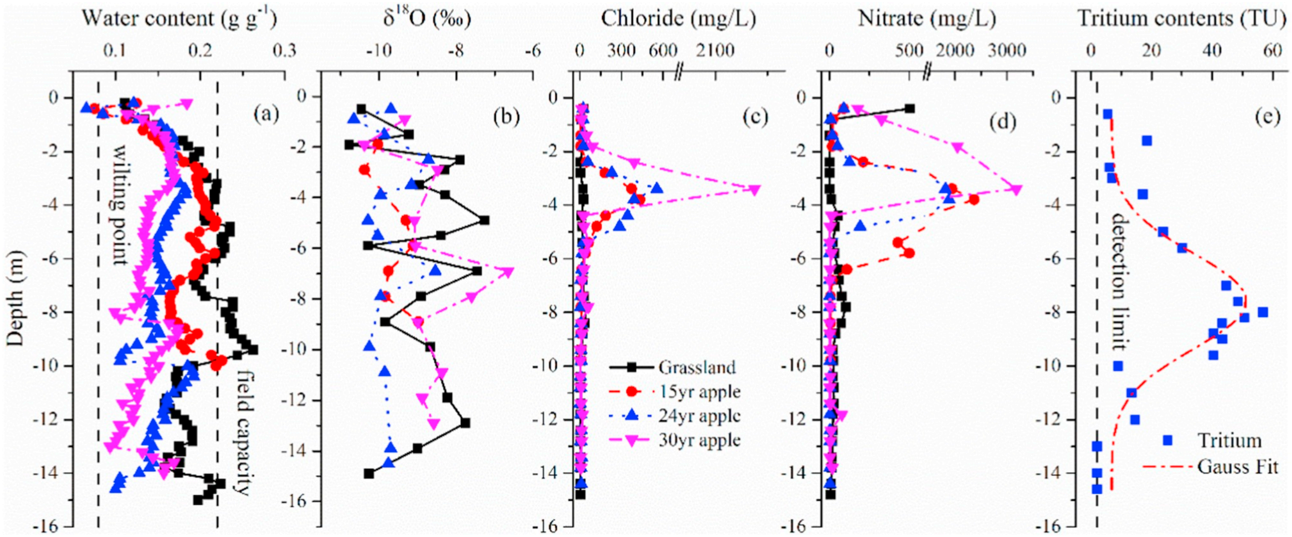

Fig. 3. Soil profiles of (a) water content, (b) oxygen isotope, (c) chloride, (d) nitrate and (e) tritium in soil water. Figures a–d share the same legend presented in c.

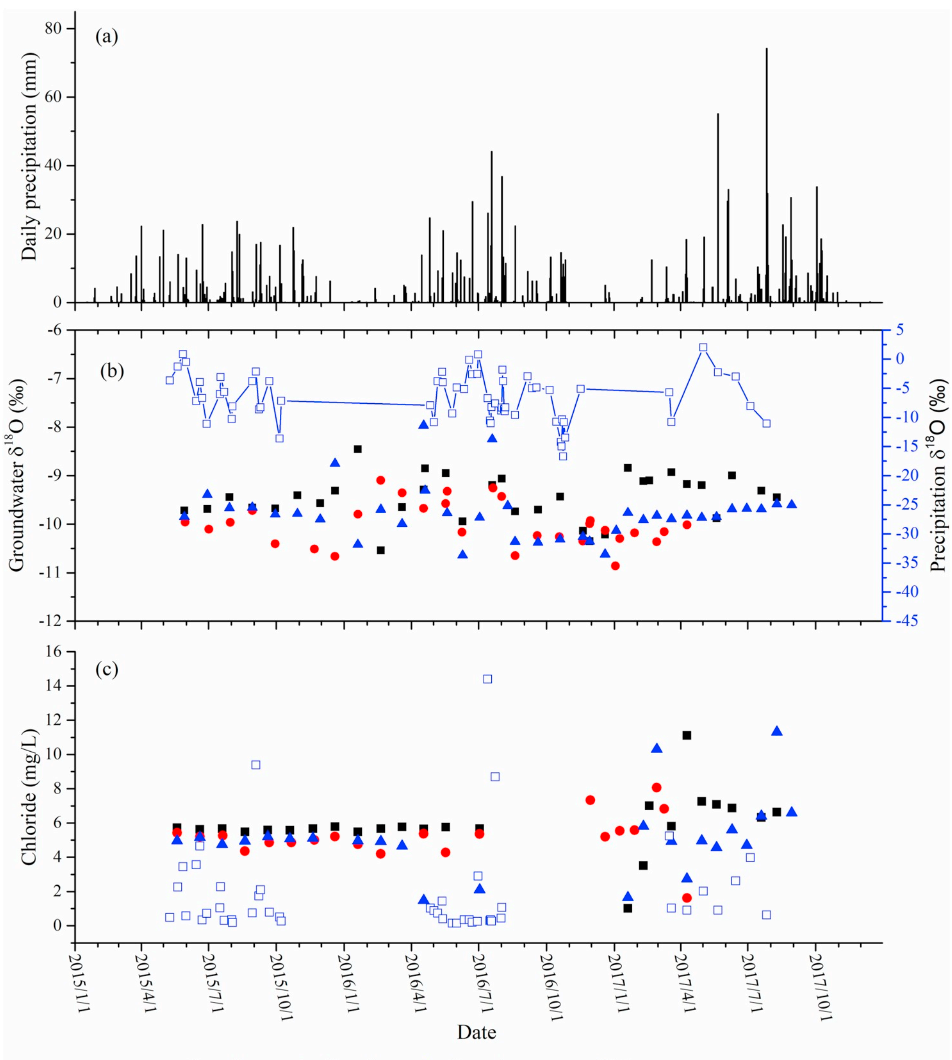

Fig. 4. (a) Daily precipitation amount, (b) precipitation and groundwater oxygen isotope and (c) chloride concentration for the period 2015–2017. Figures (b) and (c) share the same legend.

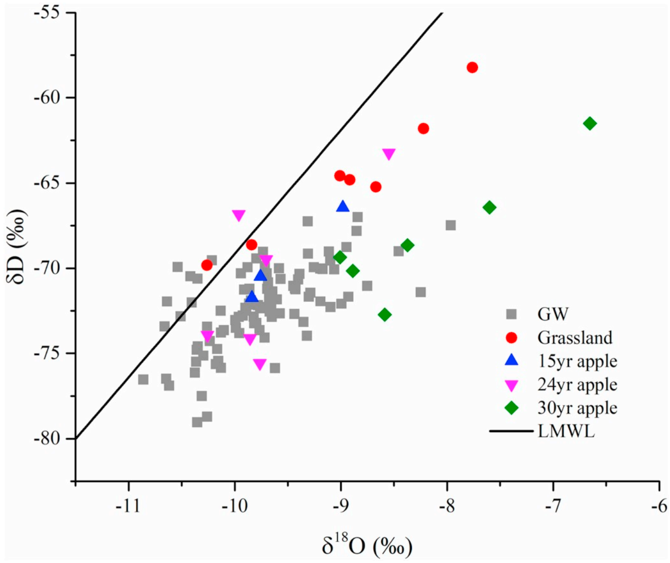

Fig. 5. Dual isotope comparison of groundwater (GW) and soil water at layers below 6 m (Grassland, 15 yr apple, 24 yr apple and 30 yr apple). LMWL is the local meteoric water line ($\delta D = 7.25\delta^{18}O + 3.35$, $R^2 = 0.93$).

combined results of the buffering effects of thick unsaturated zones and small amount of recharge (Izbicki et al., 2000; Mayers et al., 2005; Phillips, 1994; Smith et al., 1970).

3.3. Recharge components

Piston flow is apparent in all profiles (Fig. 3). Infiltration rates estimated by the peak depth method (Eq. (1)) are 15.5–19.0, 11.3–12.7 and 15.4 cm year^{-1} for chloride, nitrate and tritium, respectively. These infiltration rates suggest that it has taken rainfall 158–530 years to reach the water table at 40–60 m (residence time is calculated by dividing the depth to water table by infiltration rates). If groundwater was recharged exclusively via piston flow, it would not contain modern water less than 50 years old because modern water would not have reached the water table. However, three out of seven groundwater samples have low but detectable tritium content of 2.2, 2.9 and 3.3 TU. This suggests that the component of modern water may be small. Furthermore, the low infiltration rates via piston flow are not likely to result in dynamic changes in groundwater isotope and chloride concentration (Fig. 4). Therefore, piston flow and preferential flow both contribute to groundwater recharge. However, dual isotope comparison shows that most groundwater and soil water data fall below LMWL (Fig. 5, $\delta D = 7.25\delta^{18}O + 3.35$, $R^2 = 0.93$). This suggests that soil water subject to evaporation contributes more to groundwater and the contribution of piston flow might be larger than preferential flow.

3.4. Recharge rates

Potential recharge rates, estimated by the peak depth method (Eq. (1)), are 31.7–39.0, 23.3–26.0 and 31.6 mm year^{-1} for chloride, nitrate and tritium, respectively, accounting for 4–6% of mean annual precipitation. It should be noted that the recharge rates were respectively estimated for the past 20–22, 30 and 52 years according to the application history of chloride, nitrate and tritium. As such, these may be referred to as short-term recharge rates. Meanwhile, long-term potential recharge rates were estimated by Eq. (2). As CMB is applicable to profiles without impacts of root water uptake (Fig. 3a), the chloride concentration in soil water was calculated as the average below 10 m under all land use types. This corresponds to a long-term recharge rate of 82.1 ± 53.0 mm year^{-1}, accounting for 14 ± 9% of mean annual precipitation, which are much larger than the above short-term recharge rates.

Assuming a chloride or tritium mass balance between precipitation and groundwater, the actual recharge from precipitation and the percentages of modern water in groundwater can be estimated using Eqs. (3) and (4), respectively. Using the CMB method, the actual recharge is 112.6 ± 44.1 mm year^{-1}, accounting for 19 ± 9% of mean annual precipitation. Using the tritium mass balance method, modern water accounts for 11–18% of the total volume of groundwater.

As dual modes coexist for groundwater recharge, chloride and stable isotopes were respectively used as inputs to Eq. (5) to determine the contributions of these modes. Results from these two tracers suggest that the contributions of piston flow are greater than preferential flow. Specifically, the chloride and oxygen isotopes, respectively, indicated a 69 ± 50% and 53 ± 74% contributions of piston flow to groundwater.

3.5. Land use change impacts on recharge

The water-chloride mass balance method (Eq. (7)) was used to estimate the LUC impacts on recharge rates via piston flow. As the depth profiles were different (Fig. 3a), the water storage under grassland, 24- and 30-year-old apple trees was calculated for the profile of 0–14 m as 3568, 2761 and 2546 mm, respectively. Compared with grassland, the water storage deficit is respectively 808 and 1023 mm for 24- and 30-year-old apple trees, accounting for 23% and 29% of water storage under grassland. Consequently, they both decrease recharge rates by about 34 mm year^{-1} under 24- and 30-year-old apple trees. As the short-term piston flow was calculated as 23.3–39.0 mm year^{-1}, the old apple trees almost prevent deep drainage and thus greatly reduce groundwater recharge.

4. Discussion

4.1. How to interpret groundwater recharge mechanism considering uncertainties in the use of multiple tracers?

The estimated recharge rates vary with tracers and/or methods. Potential recharge rates from peak depth methods (Eq. (1)) span a range of 23.3–39.0 mm year^{-1} for the profile between 0 and 8 m. These values, however, are much lower than those from CMB for soil depths between 10 and 15 m (Eq. (2), 82.1 ± 53.0 mm year^{-1}). Furthermore, the actual recharge rates from the CMB method when applied to groundwater, i.e. the saturated zones below 40 m (Eq. (3)), are 112.6 ± 44.1 mm year^{-1}. Recharge rates become larger with depth, possibly because of the combined effects of climate and/or vegetation impacts and recharge mechanisms.

Potential recharge rates derived from the different methods appear to be controlled mainly by climate and/or vegetation in different periods. The recharge rates from peak depth methods are for different periods. Specifically, chloride and nitrate peaks represent the last 20–30 years while the tritium peaks represent last 50 years. Similarly, the potential recharge rates from CMB in layers below 10 m are for periods older than 50 years. Crop productivity and the areal extent of apple orchard development have increased during the last 30 years. This would have led to an increase in soil water consumption and a decrease in infiltration (Huang et al., 2018; Li et al., 2018; Zhang et al., 2018). Conversely, the potential recharge rates prior to 50 years ago were possibly controlled by climate because apple orchard development was not an important factor at the time. Hence, recharge rates were much larger than the recent 50 years.

The actual recharge rates from CMB method (112.6 ± 44.1 mm year^{-1}) are even larger than the potential recharge estimated for deep soil layers (82.1 ± 53.0 mm year^{-1}). Assuming all potential recharge (i.e. piston flow) reached the groundwater, additional recharge could only come from preferential flow since there would have been no additional water from horizontal direction. This is

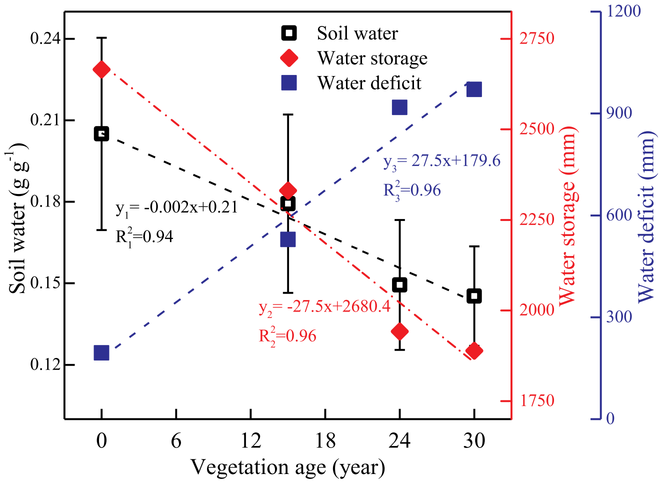

Fig. 6. Relationship between apple tree age and soil water content, water storage as well as water deficit for the profile of 0–10 m. The x-axis represents stand age of apple trees, and the points with ages of zero represent land use type as farmland. The y-axis respectively stands for soil water contents, soil water storage and soil water deficit (difference in soil water storage between farmland and apple orchard with different stand age of apple trees).

consistent with the conclusion that dual modes of mechanisms exist for groundwater recharge (Li et al., 2017b; Tan et al., 2017). Furthermore, the ratio of piston flow to actual recharge is about 73%, which is similar to the results of the two-component CMB method (69%, Eq. (5)). Therefore, the discrepancy in recharge rates between potential and actual recharge is mainly because of variable recharge mechanism indicated by different methods.

The unsaturated and saturated zones are not in hydraulic equilibrium. The unsaturated zones are dominated by piston flow, which can be clearly shown by the tritium profile in this study and some previous studies (Li et al., 2018; Li et al., 2019; Li and Si, 2018; Lin and Wei, 2006; Zhang et al., 2017). However, the saturated zones include both modern and premodern waters. The modern water is from preferential flow, an interpretation that is supported by the high groundwater tritium contents reported in previous studies (Chen et al., 2012; Li et al., 2017b; Lin and Wei, 2006; Song et al., 2017; Tan et al., 2017). However, preferential flow may only occur in restricted areas with vertical fissures, macropores or sinkholes (Beven and Germann, 2013; Gates et al., 2011; Li et al., 2017b; Li et al., 2017d; Lin and Wei, 2006; Nimmo, 2012; Tan et al., 2016).

4.2. How does land use change influence recharge?

Piston and preferential flow coexist for groundwater recharge in the study area. However, only piston flow can occur everywhere in the unsaturated zone because of the universal and uniform loess distribution. As such, LUC impacts on groundwater recharge depend on the interactions between plants and soil water (Huang et al., 2018; Li et al., 2018; Zhang et al., 2018). We calculated the mean soil water content, water storage and water deficit under different land use types, and further investigate the relationships between these indices and apple tree age.

After conversion from grassland to apple orchard, the mean soil water and water storage significantly decreases but soil water deficit significantly increases with age of apple trees. This is supported by the negative relationship between soil water content/storage and apple tree age (Fig. 6, R^2 = 0.94 and 0.96), as well as the positive correlation between soil water deficit and apple tree ages (Fig. 6, R^2 = 0.96). As such, vegetation is the key factor influencing soil water in this region. In the study region, soil desiccation and other soil physical phenomena can prevent rainfall infiltrating outright to deep layers, limited "new water" is not enough to support plant growth. Thus, the deep-rooted apple trees not only absorb new water inputs but also use old water up to several decades old (Zhang et al., 2017). This suggests that the impacts of 30-year-old apple tree on soil water can be detected for almost the entire profile up to 15 m (Fig. 3a). The impacts of apple trees on groundwater recharge were recently related to root density, and the older apple trees have larger root density and resultant smaller groundwater recharge (Li et al., 2018). Therefore, a conclusion can be made that apple trees use more water with increasing age.

4.3. Implications for water resources sustainability

Although piston and preferential flow coexist for groundwater recharge, preferential flow is not likely to be influenced by the vegetation change in the study area. Dye tracing experiment was once used to study the infiltration conditions under apple trees of 20 years old. Under intense rainfall of 50 mm, the maximum infiltration depth was 0.59 m with averaged infiltration depth of 0.53 m (Zheng et al., 2017). The results indicated that the occurrence of preferential flow was easily constrained; further, the water table is 40 m below the surface, it is thus difficult for trees to promote preferential flow by creating root holes.

However, piston flow is affected by soil water movement (Foley et al., 2005; Mahe et al., 2005; Nosetto et al., 2005), accounting for 53%–69% of the total recharge in this region. Thus, piston flow may be altered by LUC. That apple trees tend to consume more water with age suggests that progressively greater soil water deficit can only become increasingly likely (Fig. 6). LUC, specifically the conversion of farmlands to apple orchards, limits opportunities for groundwater recharge, only increasingly so as apple trees age. Currently, the apparent impacts are limited to the shallow layers, owing to low infiltration rates (11.3–19.0 cm year^{-1}). However, considering that the transit time from rainfall to groundwater is 158–530 years, the sustainability of groundwater resources may be at risk. Hence, striking a balance between economic activity (apple orchards) and groundwater supply sustainability is critical. We propose two solutions to this problem: (1) selective removal of apple trees using a target soil water content as input of the relationships developed in Fig. 6; (2) increase inter-tree planting distance to allow for more open canopy spaces and, thus, enhance infiltration.

5. Conclusion

Understanding groundwater recharge mechanisms in thick unsaturated zones with limited recharge rates can improve our understanding of hydrological processes. This study explored recharge mechanisms in thick loessial deposits with limited water resources using multiple tracers. Our results showed that the unsaturated and saturated zones have yet to reach hydraulic equilibrium. The unsaturated zones were dominated by piston flow, with increasing recharge rates with depths because of impacts of climate and land use change. The saturated zones were comprised of both modern and premodern waters, suggestive of both piston and preferential flows. Land use change from farmlands to apple orchards may largely reduce and even limit piston flow, posing a threat to groundwater resources sustainability.

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