



# Identifying the water source for subsurface flow with deuterium and oxygen-18 isotopes of soil water collected from tension lysimeters and cores



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## SUMMARY

The conventional identification of soil water with pre-event water limits deep insights into the involvement of stationary and mobile soil water in subsurface hydrological processes. In three tilled sloping field plots at a hilly area of southwestern China dominated by Entisols, soil water collected with a suction lysimeter was distinguished from the total soil water through an analysis of the stable isotopes deuterium and oxygen-18. Differences in the depth profile of soil water before and after storm events were observed and used to examine how rainwater mixes with soil water and to identify the source contribution of different fractions of soil water in subsurface flow generation. Only water in the 0–10 cm soil layer was significantly affected by evaporation and infiltration. Water in the top 5 cm layer of the soil exhibited the lowest residence time because a storm can replace a substantial proportion of the pre-event water. Soil water at the 10–20 cm depth showed the longest residence time, as indicated by its high proportion of pre-event water. The isotopic signatures demonstrated that piston flow and preferential flow coexisted in this soil. High antecedent soil water content and high rain intensity favor the formation of piston flow. The water collected with the suction lysimeter represented the mobile fraction of the pre-event water in the soil, which effectively participates in the generation of subsurface flow. Newly infiltrated rainwater did not well mix with stationary pre-event water in the soil. The use of recent rainfall to represent mobile soil water may provide a practical solution for overcoming the negative effect of the spatial heterogeneity of the isotopic composition of soil water on hydrograph separation results. Bulk soil water and lysimeter water showed significant differences in isotopic composition under low soil water content or in the top soil layer. Stable isotopes in bulk and lysimeter soil water should be monitored synchronously to reveal the sources and pathways of soil water and their contributions to the generation of subsurface flow in the vadose zone.

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## 1. Introduction

Accurate identification of different water sources is essential for the understanding of water and pollutant pathways, especially for underground hydrology studies (Renshaw et al., 2003). Based on the time source, water can be separated into event (new) water and pre-event (old) water. Event water is the water from current rain or snowmelt that enters the hillslope or catchment during a rain event. Pre-event water refers to the water already stored in the soil or catchment prior to the onset of the rain event. Usually, pre-event water is termed soil water in the unsaturated zone and

groundwater in the saturated zone. Event water was originally considered the principal source of runoff on hillslopes or in channels (Horton, 1933). However, accumulating evidence from isotopic studies in hydrology has shown that the storm hydrograph is dominated by pre-event water (Hooper et al., 1990; Laudon and Slaymaker, 1997; Shanley et al., 2002). Even the hydrograph of steep catchments are dominated by pre-event water, as shown by isotopic studies (Sklash et al., 1986). McDonnell (1990) built on previous work to provide a plausible explanation for the contribution of pre-event water to flow, but much controversy and doubt remain (Buttle and Sami, 1992). For example, how event water mixes with pre-event water in the generation of subsurface flow is still poorly understood.

The use of isotopes such as deuterium and oxygen-18 allows the researcher to gain deep insights into subsurface hydrologic processes. The concept of event water and pre-event water is

Abbreviations: EMA, end-member mixing analysis; IR, isotopic ratio; SW, soil water; SWC, soil water content; BSW, bulk soil water; LSW, lysimeter soil water.

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helpful to clarify how water from different sources mixes in the soil and contributes to the generation of subsurface flow. Using the oxygen-18 isotope, Gazis and Feng (2004) found that piston-type flow in the shallow soil was accompanied by significant mixing with stationary soil water (SW). In contrast, traditional methods, such as soil water content (SWC) measurement, could not identify that phenomenon accurately.

Recent studies have shown that the isotopic ratios (IRs) of bulk soil water (BSW, matric-bound stationary water and mobile water) and lysimeter soil water (LSW, mobile water) are isotopically different (Landon et al., 1999; Figuéroa-Johnson et al., 2007; Brooks et al., 2010). However, both BSW and LSW have been adopted in the literature as a measure of the pre-event water present in the soil prior to a storm. For example, several studies used the measured IR of SW extracted by low-tension suction lysimeters to calculate the proportion of SW in the hydrograph (Dewalle et al., 1988; Lee and Krothe, 2001; Vogel et al., 2010). Using the same method, McGuire et al. (2002) and Lee et al. (2007) estimated the mean residence time of water in the soil. However, other researchers measured the IRs of the total SW extracted by the cryogenic vacuum distillation method as a measure of pre-event water for water source separation (Ladouche et al., 2001; Zhang et al., 2011). To date, little evidence has been reported to identify the value or values that represent pre-event water in terms of its involvement in subsurface flow generation.

In addition, the strong spatial heterogeneity of IRs in pre-event water posed a difficult problem for hydrologists and limited deep insights into hydrological processes (Kendall and McDonnell, 1998). The heterogeneity of IRs has been reported most frequently for the SW in different soil layers. In particular, due to evaporation, the top soil layer always has higher amounts of relatively heavy isotopes than the deeper layers. Brooks et al. (2010) found that the variation in the isotopic depth profile of SW in a single plot was much greater than the corresponding spatial variation at any depth across the catchment in which the plot was located. Genereux and Hooper (1998) noted that subsurface sampling is necessary to evaluate the isotopic homogeneity of pre-event water. A hydrograph separation should be based on spatially distributed sampling of pre-event water, avoiding the logistical pitfalls of point measurements on hillslopes. Finding a simple way to measure the IR of pre-event water in the subsurface as an alternative to laborious spatially distributed sampling is valuable for isotopic hydrology studies.

Seasonal floods and droughts frequently occur in the hilly area of southwestern China. Purple soil, an Entisol in USDA Taxonomy, is widely distributed in this region. This soil is characterized by high productivity, low water retention ability and a high nitrogen loss rate (Zhu et al., 2009). However, the subsurface hydrological processes in the soil remain poorly understood. Consequently, the objectives of this study were to (1) explore the changes in the isotopic depth profile of soil water before and after a rainfall event on a hillslope; (2) determine which measure of pre-event water (BSW or LSW) can better represent the water source for subsurface flow; and (3) clarify the isotopic difference between BSW and LSW for different soil water contents and different depths.

## 2. Materials and methods

### 2.1. Study site

The experimental hillslope site investigated in this study is situated in a small agricultural catchment in hilly central Sichuan, southwestern China (31°16'N, 105°28'E) (Fig. 1a). The catchment is dominated by a loamy soil, locally termed purple soil (an Entisol in USDA Taxonomy), and is representative of the Upper Yangtze River Reaches region. The study soil is a loamy soil with average textural data of 27.1% sand, 51.6% silt and 22.3% clay. Hillslope

hydrology conditions in this region significantly affect the water quality of the Yangtze River. This influence has become particularly important following the construction of the Three Gorges Dam (Zhu et al., 2009). The region has a moderate subtropical monsoon climate with an annual mean temperature of 17.3 °C and a mean annual precipitation of 826 mm (both mean values based on the years 1981–2006). During the same period, 85.2% of annual precipitation occurred in the summer and autumn. The hillslope of this region was characterized by a developed subsurface flow with a runoff coefficient of 24% on average from 2003 to 2006. Subsurface flow had been identified to be the main pathway for nitrogen transport which accounted for over 88% of the total loss amount from runoff.

The subsurface hillslope discharge and soil water were monitored in three sloping experimental plots during three storms. The schematic setup of the plots is shown in Fig. 1b. The length, width and depth of the plots were 8 m, 4 m and 0.4 m, respectively. The average slope in this region is approximately 6%. The field plots were planted by maize (*Zea mays* L.) with a density of 40,000 plants ha<sup>-1</sup>. Our preliminary experiment has proven that the maize did not cause significant isotopic fractionation.

### 2.2. Sampling and field measurements

The amount of rain was measured with a tipping bucket rain gauge with 0.1-mm resolution. Rainwater was collected with a 20-cm diameter glass funnel connected to a high-density polyethylene bottle. The subsurface flow discharge was measured continuously with a tipping bucket connected to an HOB0 event data logger (Onset Computer Corp., MA, USA). Beginning with the initiation of flow, water samples of the subsurface hillslope flow were taken at 0.5-h intervals until the flow ended. A mixture of the effluent water in a large bucket was sampled, and the resulting IR values represent time-averaged concentrations.

Two methods were used to extract SW: cryogenic vacuum distillation (West et al., 2006) and a low-tension lysimeter. Soil samples were collected from 5 depths (0–5, 5–10, 10–20, 20–30 and 30–40 cm) with a hand soil auger and were collected in glass vials for subsequent cryogenic vacuum distillation. In addition, the volumetric SWC was measured by TDR trase system (Soil moisture Equipment Corp., CA, USA) at the same soil layer. At the same soil sampling depth, low-tension (a maximum tension of 80 kPa was applied) porous-cup lysimeters (Soil moisture Equipment Corp., CA, USA) were installed to collect SW. During the first (22 July 2012) and third (30 August 2012) storms, these two methods were used synchronously to extract SW at an interval of approximately 2–3 h. Because the second storm occurred after a long period of drought, no LSW samples were extracted before and during the storm due to the low SWC. As mobile SW is defined in terms of the most recent precipitation (Landon et al., 1999), the IR of the rainwater on 19 August 2012, representing a total rainfall of 3.2 mm (the sole rainfall event during approximately 1 month before the second storm) occurring immediately prior to this storm (the second storm occurred on 20 August 2012, with 55.9 mm of rainfall) was used as the LSW value and was evaluated to determine whether this value appeared reasonable for the purposes of this research.

### 2.3. Isotopic data

Rainwater, SW and subsurface flow samples were collected in glass vials with cap inserts and sealed with parafilm to prevent evaporation. Water samples were analyzed for  $\delta D$  and  $\delta^{18}O$  with a laser absorption water–vapor isotope analyzer (Picarro-i2120, CA, USA). All  $\delta D$  and  $\delta^{18}O$  values are expressed relative to VSMOW in ‰ (permil):

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