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# Applicability of Difference in Oxygen-18 and Deuterium of Water Sources and Isotopic Hydrograph Separation in a Bamboo Catchment during Different Rainfall Types

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**Abstract:** Typhoon storm and plum rain are two typical rainfall types in the lower regions of the Yangtze River Basin, which frequently cause flood disasters in China. New information in stable water isotopes offers the opportunity to advance understanding of runoff mechanisms and water source dynamics in response to these two typical rainfall types. We intensively monitored two representative rainfall events in a small bamboo forestry watershed in 2016. Results showed that precipitation isotopic variations during the event were generally larger than those of other monitored compartments (including throughfall, surface overland water, groundwater and river water) and also larger for the plum rain than for the typhoon event ($\delta^{18}O$ varied in 5.2‰ and 3.7‰, respectively). Importantly, the differences of isotopic temporal variation between rainfall and throughfall showed significant impacts on the two-component hydrograph separation for both rainfall types (e.g., if not considered, the pre-event water fractions were 26.6% and 15.3% higher for the typhoon and plum rain events, respectively). Furthermore, we evaluated the role of soil water on the three-component isotopic hydrograph separation model; results revealed that soil water accounted for 10.9% and 28.3% of the total discharge in typhoon and plum rain events, respectively. This underpins the important role of soil water dynamics during the rainy season in this humid region.

**Keywords:** $\delta^{18}O$ and $\delta D$ isotopes; temporal variations during event; plum rain; typhoon; isotopic hydrograph separation; water sources

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## 1. Introduction

The lower region of the Yangtze River Basin, as one of the most active economic regions of China, has experienced dramatic development over recent decades; meanwhile, high risks of social-economic damages have been raised by flood-induced disasters, which might occur regularly due to the regional climate pattern (i.e., heavy rainfall events occurring during the plum rain and typhoon seasons) [1]. For precise flood predictions and optimal water resource management, it is of highly importance to advance understanding of the regional hydrological cycle and catchment mechanistic responses to the typical plum rain and typhoon events. Stable isotopes of water ($\delta D$ and $\delta^{18}O$), as an ideal tracer, have been increasingly introduced in hydrology research to inform hydrological processes and their responses to environmental changes [2]. Among others, stable water isotopes and their temporal variations have been widely integrated into discharge hydrograph separation,

revealing event-scale water source portioning and its dynamics. However, it is much less intensively studied in the lower region of the Yangtze River Basin with regular plum rain and typhoon events.

Isotopic hydrograph separation (IHS) makes use of the stable isotopes of water (δD and $\delta^{18}O$) as conservative tracers to trace the movement of water through catchments and to estimate how much all water sources contribute to streamflow. Existing IHS studies highlighted the importance of the old water (pre-event water) in the runoff generation [3,4], which contrasts to the traditional perception that "rainfall was considered to dominate storm hydrographs". Many hydrologists have focused on five assumptions in the traditional two-component hydrograph separation [5–7] to improve the understanding of different dominant runoff components in different watersheds.

One assumption is that the isotopic composition of rain or snowmelt is constant, or its spatiotemporal variation can be accounted for [8]. Many studies have found that the isotopic composition of rainfall exhibited significant change during an event and evaluated how the isotopic temporal variation influences the results of IHS [5,9].

Additionally, isotopic composition of direct rainfall from the open sky is different from that of throughfall due to many potential reasons, for example, their different formation processes [10,11]. Some studies have found that, compared with direct rainfall, throughfall is enriched in $^{18}O$ and D [12], while others have observed completely opposite results [10]. Spatiotemporal variability of throughfall isotopic composition at both the plot and catchment scale has been reported by previous studies [13], but few have explored impacts of the difference between rainfall and throughfall isotopic variations on hydrograph separation to date. This is of particular importance for forestry catchments, where canopy interception might significantly impact catchment responses to rainfall.

Another assumption is that the contribution of soil water can be ignored, or its isotopic composition is the same as that of groundwater. In the development of three-component models in hydrograph separation, interflow from soil water is assumed to be playing a key role in runoff partitioning and in regulating seasonal variations of stream water isotopes [14]. Soil water contributions are not negligible based on investigations of hydrochemical parameters in different watersheds, and the mechanism of interflow generation is examined in experimental watersheds [15,16]. However, the temporal variability of soil water isotopic composition during rainfall events has not been intensively focused on [17], nor its influence on the results of three-component isotopic hydrograph separation.

Typhoon is a mature tropical cyclone that develops between 180° and 100° E in the Northern Hemisphere typically between July and September; plum rain is known as an annual meteorological cycle of precipitation in the East Asia monsoon region and occurs during May to July in the lower region of the Yangtze River Basin [18]. Plum rain and typhoon events result from two different climatic systems, thus exhibiting differences in runoff partitioning, as well as isotope signatures in different runoff components. Currently, transport trajectories and water sources of plum rain and typhoon have been studied in many basins [19,20], while detailed isotopic comparisons and their applications in hydrograph separation remain rare.

Overall, the isotopic composition of rainfall, throughfall and soil water might exhibit significant temporal variations during the different plum rain and typhoon rainfall events in the lower Yangtze River regions; and considering such variations in hydrograph separation may offer new insights into the regional hydrological processes and their responses to typical rainfall types. The objectives of this study are: (1) to examine the temporal variations and characteristics of isotopic composition in different water sources during different rainfall types at event scale and to illuminate the reasons for the differences; (2) to calculate the hydrograph separation based on isotopes in precipitation and throughfall, and compare the differences of using constant isotopic values and temporal variations during the event; (3) to evaluate the soil water contribution of the two different rainfall types using a three-component hydrograph separation model; and (4) to explore the runoff generation mechanism between rainfall types based on the isotopic signatures and hydrograph sepa-

ration results. To achieve these objectives, we intensively monitored one typical typhoon and plum rain event in 2016 and analyzed the temporal variations of isotopic compositions in rainfall, throughfall, surface water, soil water, groundwater and river water. Hourly sampling scheme was conducted in the Hemuqiao watershed, a small bamboo watershed in the lower region of the Yangtze River Basin. Together, the stable isotope data obtained from all key hydrological compartments established the data foundation for the detailed hydrograph separation and process understanding in the study watershed.

## 2. Materials and Methods

### *2.1. Study Area*

The Hemuqiao catchment is a 1.35 km$^2$ bamboo headwater catchment (Figure 1: 30°34′05″~30°34′55″ N; 119°47′05″~119°48′20″ E), located in in the lower regions of the Yangtze River Basin. This wet subtropical monsoon climate region is one of the highest rainfall zones in China. The region has high mean annual rainfall of 1580 mm/y. Most of the rainfall occurs during the monsoon from June to September. The watershed is located in a mountain area, and the elevation varies from 150 to 600 m above sea level. The mean annual air temperature is 14.0 °C, and monthly mean minimum and maximum temperatures are 1.3 °C and 25.0 °C, respectively. Annual average water surface evaporation is 805 mm. Land cover is dominated by bamboo. The study area exhibits multiple soil types, including red soil, yellow soil, lithologic soil, fluvo-aquic soil and paddy soil. The typhoon event on the 14 September and the plum rain event on the 24 June, which occurred in 2016, were selected for study.

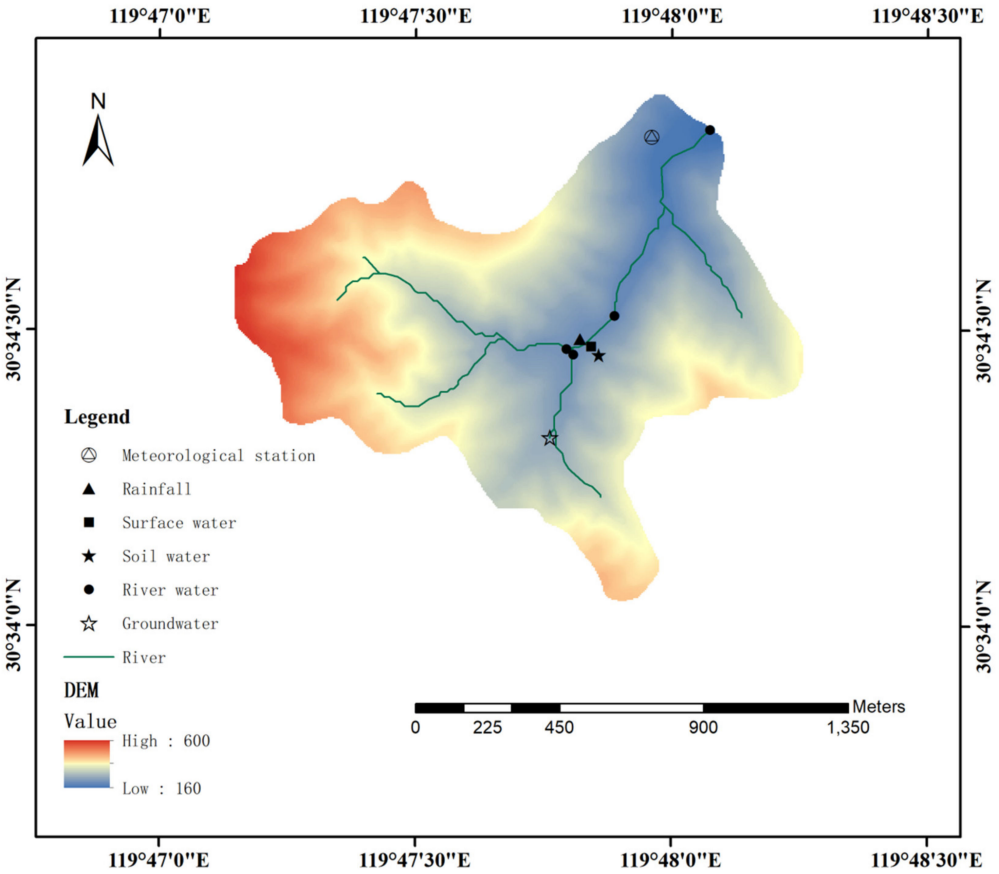



**Figure 1.** Location of the Hemuqiao catchment and sampling sites for different water sources.

### *2.2. Methods*

#### 2.2.1. Sampling Method during the Rainfall Events

Rainfall, throughfall and meteorological data were monitored at Hemuqiao meteorological station during the two selected storms (Figure 1). Precipitation was monitored using a rain gauge with a tipping bucket. A water level sensor was deployed above a rectangular thin-wall weir at the outlet of the catchment to record the water level dynamics. Discharge was calculated using the rating formula of a right triangle thin-wall weir from the water level data.

The sampling locations are shown in Figure 1. Rainfall, throughfall, surface water and soil water sampling sites were located in the middle of the catchment. Groundwater samples were collected from the piezometer site. Stream water was sampled at the outlet of the catchment. Bulk rainwater and throughfall were collected with a 50 cm-diameter stainless steel barrel. In addition, incremental rainfall and throughfall sampling were conducted simultaneously using modified versions of a Kennedy [21] sequential rainfall sampler. At the same time, hourly sampling for rainfall was conducted using a 20 cm-diameter stainless steel barrel. Surface water samples were collected using collectors emplaced in the gully and soil water samples were collected from a soil sampler installed 50 cm beneath the surface. The temporal resolution of samples of rainfall, surface water, soil water and stream water was hourly, from 8:00 to 20:00. Groundwater was sampled every two hours during the typhoon event and was set to have the same value for the baseflow during the plum rain event.

Vacuum distillation was used to extract soil water. The collected samples were analyzed for isotopic compositions ($\delta^{18}O$ and $\delta D$) using mass spectrometry (Thermo Fisher MAT 253) and EC (electronic conductivity) in the State Key Laboratory of Hydrology-Water Resources and Hydraulic Engineering, Hohai University. The isotopic compositions were reported in δ-notation as per mil (‰) relative to the standard isotope ratio of Vienna Standard Mean Ocean Water (V-SMOW). The accuracy of measurements is ±0.2‰ for oxygen-18 and ±1‰ for deuterium.

#### 2.2.2. Hydrograph Separation Method

The two-component isotopic hydrograph separation model is based on the water and isotopic concentration balance equations, as follows:

$$Q_t = Q_o + Q_n \tag{1}$$

$$\delta_t Q_t = \delta_o Q_o + \delta_n Q_n \tag{2}$$

where Q and δ denote the discharge and the isotopic values of different water components, respectively. Subscripts t, o and *n* refer to the total stream water, old water (pre-event water, contributing from groundwater and soil water that are stored in watershed prior to event) and new water (event water, contributing from rainfall), respectively. The relative contributions of the old water and new water can be calculated as:

$$\frac{Q_o}{Q_t} = \frac{\delta_t - \delta_n}{\delta_o - \delta_n}, \ \frac{Q_n}{Q_t} = \frac{\delta_t - \delta_o}{\delta_n - \delta_o} \tag{3}$$

Considering temporal variation of rainfall (throughfall) isotopic composition during the event, an arithmetic mean isotopic value (δ) for the isotopic value can be computed as:

$$\delta = \sum_{i=1}^{n} \frac{\delta_i}{n} \tag{4}$$

where $\delta_i$ denotes the isotopic value of the *i*th samples, and n denotes the total number of samples throughout the rainfall event.

Additionally, the isotopic hydrograph separation was also conducted using the three-component model, which explicitly separates the pre-event water sources into soil water

and groundwater. Equation (5) is used to determine the relative contributions of rainfall, soil water and groundwater:

$$\begin{bmatrix} Q_r/Q_t \\ Q_i/Q_t \\ Q_g/Q_t \end{bmatrix} = \begin{bmatrix} 1 & 1 & 1 \\ \delta^{18}O_r & \delta^{18}O_i & \delta^{18}O_g \\ EC_r & EC_i & EC_g \end{bmatrix}^{-1} \begin{bmatrix} 1 \\ \delta^{18}O_t \\ EC_t \end{bmatrix} \quad (5)$$

where $\delta^{18}O$ is the oxygen-18 value and EC is electronic conductivity. Subscripts t, r, i and g refer to stream water, rainfall, soil water and groundwater, respectively. If surface overland water is sampled and its isotopic signature is considered to be different from rainfall water, then r refers to surface water.

## 3. Results and Discussion

### *3.1. Characteristics of Isotopic Value and EC in Waters during Different Rainfall Type*

#### 3.1.1. Rainfall and Throughfall

Characteristics of stable isotope values and EC in the water reservoirs during different storm events at the Hemuqiao catchment are given in Table 1. In this study, rainfall and throughfall isotopic composition both exhibited significant temporal variations and the variation range of rainfall was greater than that of throughfall (for typhoon, $\delta^{18}O$: 3.7‰ Vs 0.6‰, δD: 33.4‰Vs 4.9‰; for plum rain, $\delta^{18}O$: 5.2‰ Vs 1.8‰, δD: 37.7‰ Vs 17.9‰).

**Table 1.** Characteristics of stable water isotopes and EC in each hydrological compartment during the typhoon and plum rain events at the Hemuqiao catchment.

| Water Source | Rainfall Type | $\delta^{18}O$(‰) | | | δD(‰) | | | EC(μS/cm) | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | Min | Max | Mean | Min | Max | Mean | Min | Max | Mean |
| Rainfall | Typhoon | −9.1 | −5.4 | −7.7 | −64.3 | −30.9 | −54.8 | 20.2 | 58.6 | 43.7 |
| | Plum rain | −13.5 | −8.3 | −11.1 | −95.3 | −57.6 | −74.2 | 30.4 | 70.2 | 54.1 |
| Throughfall | Typhoon | −8.7 | −8.1 | −8.3 | −64.3 | −59.4 | −62.0 | 47.4 | 50.9 | 49.2 |
| | Plum rain | −13.0 | −11.2 | −11.9 | −89.9 | −72.0 | −79.3 | 42.4 | 69.2 | 54.7 |
| Surface water | Typhoon | −8.7 | −7.0 | −8.0 | −57.5 | −48.5 | −53.8 | 70.4 | 98.4 | 84.7 |
| | Plum rain | −7.2 | −4.2 | −6.3 | −42.9 | −34.4 | −40.2 | 33.1 | 57.9 | 44.2 |
| Soil water | Typhoon | −13.7 | −5.7 | −8.3 | −106.1 | −54.0 | −71.4 | 57.6 | 79.3 | 67.6 |
| | Plum rain | −11.4 | −1.0 | −7.2 | −89.2 | −46.1 | −67.3 | 43.1 | 88.1 | 67.1 |
| Groundwater | Typhoon | −8.2 | −7.1 | −7.6 | −55.8 | −47.2 | −51.7 | 141.7 | 179.9 | 157.3 |
| | Plum rain | −4.5 | −4.5 | −4.5 | −36.9 | −36.9 | −36.9 | 43.8 | 43.8 | 43.8 |
| Stream water | Typhoon | −8.7 | −6.9 | −7.9 | −56.6 | −49.9 | −53.6 | 89.6 | 136.7 | 105.2 |
| | Plum rain | −7.5 | −4.6 | −6.7 | −44.7 | −36.9 | −41.5 | 46.9 | 78.8 | 60.2 |

The short-term rainfall isotopic fluctuation during a rainfall event can be strongly influenced by factors such as moisture source, rainfall intensity, wind velocity and cloud structure [22–25]. As a result of their different formation processes, the variation range of rainfall δD and $\delta^{18}O$ can be greater than that of throughfall (Table 1). Typically, in forest catchments, throughfall occurs only when cumulative precipitation exceeds canopy storage capacity. This means the intercepted rainfall water will be fully mixed before dropping; meanwhile, the occurrence time of throughfall is later than that of rainfall, resulting in a more damped pattern in the throughfall isotopic composition.

Due to the finer sampling resolution, the temporal variations of rainfall isotopes during the event were nicely captured by our monitoring. In the typhoon event, rainfall $\delta^{18}O$ values exhibited the pattern of a first enrichment and were followed by a depletion, during the heavy rainfall period (highlighted in Figure 2a), with increasing rainfall rates, the $\delta^{18}O$ values increased from −7.28‰ to −5.58‰ and subsequently decreased to −8.43‰ as the rainfall rates decreased. In contrast, for the plum rain event, rainfall $\delta^{18}O$ value

decreased from −10.47‰ to −13.52‰ and reached the minimum value of the entire event. During the intensive rainfall period (highlighted in Figure 3a), the $\delta^{18}O$ values increased to −8.25‰, reaching the maximum value. Generally, a plum rain event has more abundant total precipitation, a longer duration, significant isotopic fractionation and presentation of amount effect; all together typically resulting in more depleted isotopic values in rainfall. This is in line with our measurements (Table 1), that the averaged isotopic value of rainfall in typhoons ($\delta^{18}O$: −7.7‰; δD: −54.8‰) was higher than that in plum rain ($\delta^{18}O$: −11.1‰; δD: −74.2‰). In these humid regions with high relative humidity, evaporation and small temperature variation in the process of rainfall, with a decrease in temperature and evaporation of raindrops, air saturation is easy to compensate; however, with prolonged rainfall, no evaporation and mixing with surrounding moisture occurred and resulted in a depleted value of rainfall.

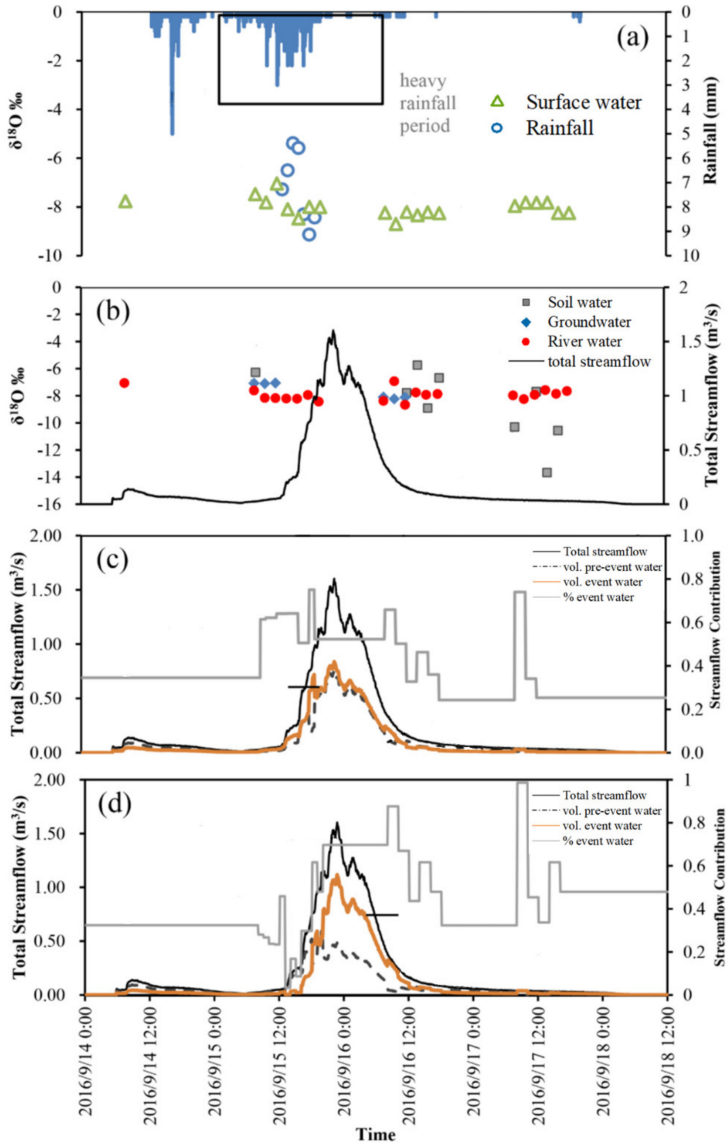



**Figure 2.** (**a**) Rainfall and surface water $\delta^{18}O$ composition during the typhoon storm and rainfall amount. (**b**) $\delta^{18}O$ values for soil water, groundwater and streamflow. (**c**) Discharge and results of the two-component mixing model, using $\delta^{18}O$ for pre-event and event waters as a constant. (**d**) Discharge and results of the two-component mixing model, using temporal variation in δ18O in rainfall.

### 3.1.2. Surface Water

From Table 1 and Figures 2a and 3a, it can be seen that although the surface water isotopic composition variation was smaller than that of rainfall (for typhoon, $\delta^{18}O$: 1.7‰ Vs 3.7‰, $\delta D$: 9‰ Vs 33.4‰; for plum rain, $\delta^{18}O$: 3.0‰ Vs 5.2‰, $\delta D$: 8.5‰ Vs 37.7‰), it still exhibited significant temporal variation (for typhoon, $\delta^{18}O$: −8.7‰ to −7.0‰, $\delta D$: −64.3‰ to −59.4‰; for plum rain, $\delta^{18}O$: −7.2‰ to −4.2‰, $\delta D$: −42.9‰ to −34.4‰). The changes in surface water isotopic values might be caused by the dynamic mixing of precipitation during the event and the water stored before the event [26,27]. The significant temporal variability of surface water isotope values emphasizes the importance of measuring the $\delta^{18}O$ and $\delta D$ of sequential surface water samples, rather than using a single fixed value for hydrograph separation [5,12,26,28–31].

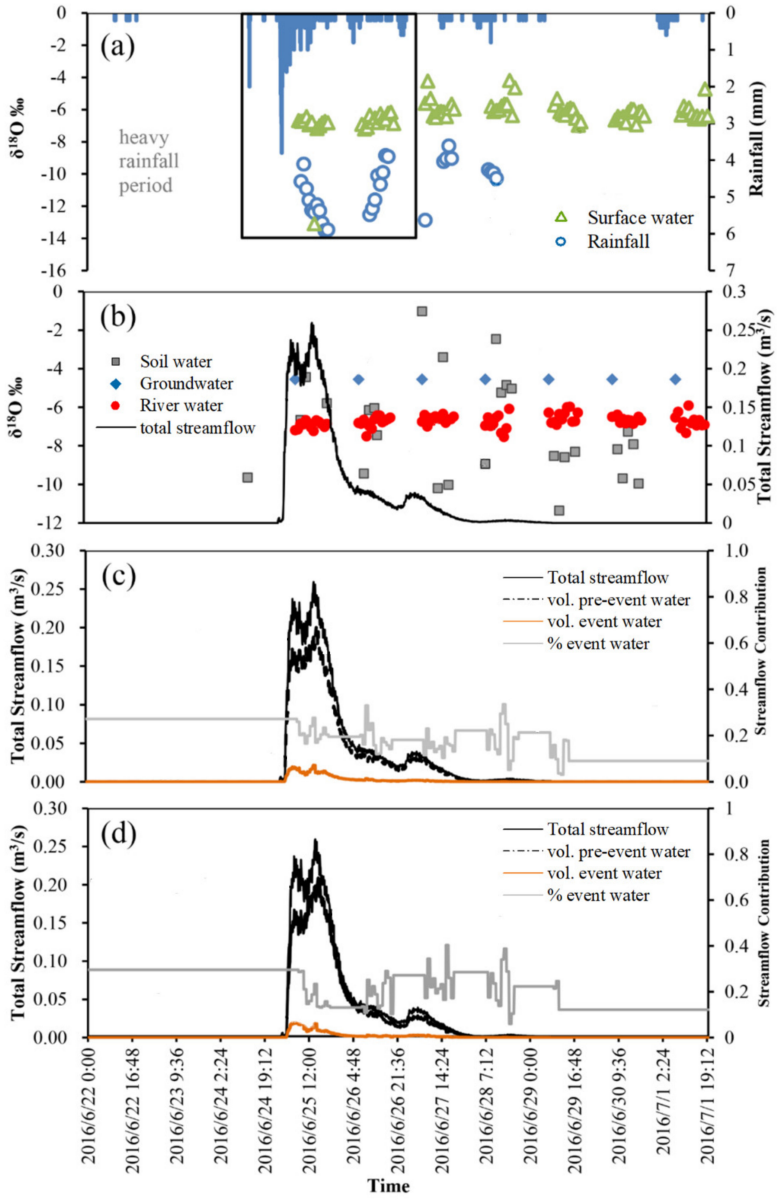



**Figure 3.** (**a**) Rainfall and surface water $\delta^{18}O$ composition during the plum rainstorm and rainfall amount. (**b**) $\delta^{18}O$ values for soil water, groundwater and streamflow. (**c**) Discharge and results of the two-component mixing model, using $\delta^{18}O$ for pre-event and event waters as a constant. (**d**) Discharge and results of the two-component mixing model, using temporal variation in $\delta^{18}O$ in rainfall.

### 3.1.3. Soil Water

The soil water isotopic value exhibited significant temporal variation during two type rainfall events (for typhoon, $\delta^{18}O$: −13.7‰ to −5.7‰, δD: −106.1‰ to −54.0‰; for plum rain, $\delta^{18}O$: −11.4‰ to −1.0‰, δD: −89.2‰ to −46.1‰; Table 1 and Figures 2b and 3b). The significant variation in soil water isotopic value reinforces the importance of soil water sampling, which will improve the accuracy of isotopic hydrograph separation. The variation range of soil water $\delta^{18}O$ values was greater than that of rainfall (for typhoon, 8‰ Vs. 3.7‰; for plum rain, 10.4‰ Vs. 5.2‰). This indicates that soil water can be replenished not only by event precipitation but also by other water sources, such as groundwater or river water due to the rising groundwater and stream water level.

Figure 4 shows the correlations between $\delta^{18}O$ and δD of different water source compartments. The enriched soil water isotope values, i.e., located right-below the meteoric water line, indicate that soil evaporation (in addition to transpiration) has induced fractionation effects on water in the soil zone, and that rainfall are the greatest source of recharging soil water. Some of the soil water values are higher than precipitation, and a possible reason is that soil water in certain parts of the catchment is mixed with enriched pre-event water. A similar conclusion is found in Gazis's study that $\delta^{18}O$ average values in precipitation are −17.2‰, compared with an enriched average in soil water (−14.0‰) [32]. The above results of isotopic soil water revealed that there is a large isotopic fluctuation in soil water, which could be regarded as a mixture of precipitation and pre-event water.

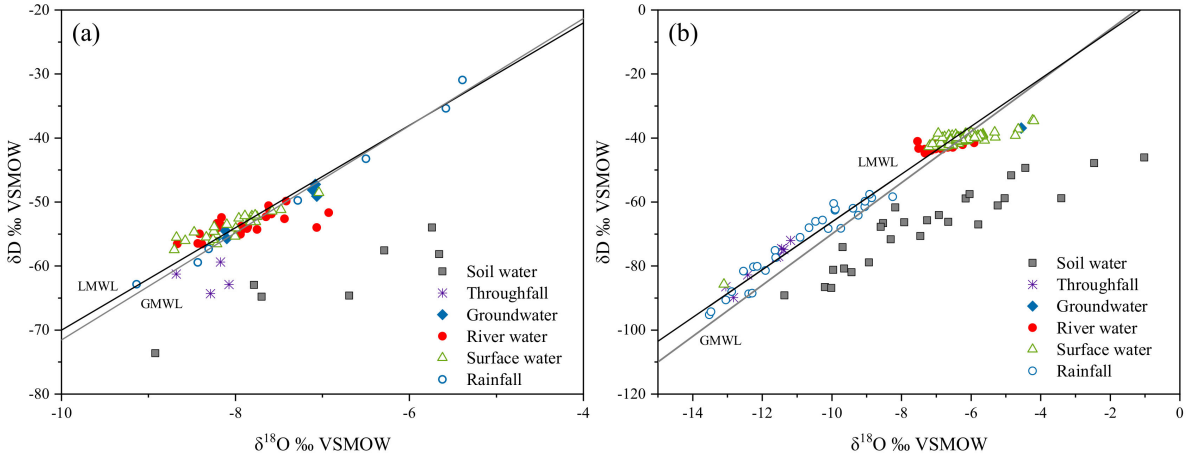



**Figure 4.** All $\delta^{18}O$ and δD data of rainfall, surface water, soil water, groundwater, and stream water relative to the local (LMWL) and global (GMWL) meteoric water lines. The LWML is calculated from rainfall collected in all rainfall events between 2015 and 2016 in the Hemuqiao catchment. (**a**) Typhoon event and (**b**) plum rain event.

### 3.1.4. Groundwater

There was consistent limited change in the groundwater isotope values throughout the typhoon event ($\delta^{18}O$: −8.2‰ to −7.1‰, δD: −55.8‰ to −47.2‰; Table 1 and Figure 2b). Isotope values of sampled groundwater prior to the typhoon event were −7.6‰ for $\delta^{18}O$ and −51.7‰ for δD, respectively. The $\delta^{18}O$ value in the groundwater samples had slightly smaller variation range (1.1‰) compared to that of rainfall (3.7‰), surface water (1.7‰) and soil water (8‰) samples. The relatively constant groundwater isotopic composition supported the assumption that using the pre-event baseflow isotopic value as the initial groundwater isotopic value for hydrograph separation in the Hemuqiao catchment [16].

#### 3.1.5. River Water

Compared with rainfall, the variation ranges of stream water isotopic composition were smaller (for typhoon, $\delta^{18}O$: 1.8‰ Vs. 3.7‰, $\delta D$: 6.7‰ Vs. 33.4‰; for plum rain, $\delta^{18}O$: 2.9‰ Vs. 5.2‰, $\delta D$: 7.8‰ Vs. 37.7‰); however, it still showed significant temporal fluctuations, suggesting that a constant isotopic value of stream water is unlikely sufficient for hydrograph separation.

In the typhoon event, the dynamic of stream water isotopic composition demonstrated a tendency of a slight decrease followed by an increase, which is opposite to the dynamics of the stream discharge (Figure 2b). At the beginning of the event, the discharge was $0.02 m^3/s$ and the average stream water $\delta^{18}O$ value was −7.06‰; at the peak time, the discharge increased to $1.60 m^3/s$, and the $\delta^{18}O$ value decreased to −8.43‰. After 11h of the peak discharge, the stream discharge decreased to baseflow level (ca. $0.24 m^3/s$), and the stream water $\delta^{18}O$ values returned to −6.93‰.

The evaporation of river water can also potentially influence the isotopic value of river water and then affect the component of discharge hydrograph. The river water points to the dual-isotope space being distributed around the LMWL (Figure 4), indicating that precipitation water is the primary recharge source to river water; however, the slope was less than LMWL, indicating that the source waters have also undergone intense dynamic fractionation.

### *3.2. Isotopic Hydrograph Separation*

#### 3.2.1. Impacts of Temporal Variations of Rainfall Isotopic Composition

The results of the two-source isotopic hydrograph separation model in the typhoon event showed that during the first 10.75 h, the river water was dominated by pre-event water (Figure 2d). At the flood peak point, stream discharge was dominated by the event water (69.6%), which may be a result of saturated overland flow during the heavy rainfall period (Figure 2d). Meanwhile, there was still considerable pre-event water contributing to the peak stream water (accounting for 30.4%). In the flood recession period, the event water contribution decreased, and the pre-event water predominated again after 15.25 h of the peak discharge.

Considering temporal variations of rainfall isotopes during the typhoon event, we could observe a three-phase pattern during the rising stage of the hydrograph (Figure 2b,d): (1) the increase of flow is initially supported by pre-event water; (2) then, the event water contribution increased sharply associated with a decrease in stream isotopic values; and (3) finally, the stream water becomes predominated by the event water. Interestingly, such dynamic pattern of event water contribution was not observed when using constant $\delta^{18}O$ value. Moreover, the event water fractions were underestimated compared to the former case (i.e., the whole event average: 54.6% Vs. 58.3%; at the discharge peak time: 47.9% Vs. 69.6%; Figure 2c,d). Continuous monitoring of stable isotope $\delta D$ and $\delta^{18}O$ are therefore necessary for capturing the dynamic variations in the rising limb of the stormflow hydrograph. This result emphasizes the importance of the consideration of temporal variation and the use of finer temporal scale [33] in $\delta^{18}O$ in rainfall in two-component isotopic hydrograph separation.

In contrast, during the plum rain event, the stream water continued to be predominated by pre-event water (i.e., the whole event average: 77.8%; at the discharge peak time: 76.7%). Moreover, the differences of event water contribution between using the constant rainfall $\delta^{18}O$ value and considering its temporal variation during the event were not significant in plum rain events, as compared with typhoon events (Figures 2 and 3). This might have been resulted from the different formations of the two types of rainfall.

#### 3.2.2. Impacts of using Isotopes of Rainfall and Throughfall

The different formation processes between rainfall and throughfall influenced the results of isotopic hydrograph separation for different rainfall types (Table 2; Figure 5a,b).

**Table 2.** Results of isotopic separation model in the Hemuqiao watershed.

| Event Type | Pre-Event Water Fraction Based on Two-Source IHS Model | | | | Three-Component IHS Model | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Peak/% | | Average/% | | Peak Fraction/% | | | Average Fraction/% | | |
| | Precipitation | Throughfall | Precipitation | Throughfall | Surface Water | Soil Water | Groundwater | Surface Water | Soil Water | Groundwater |
| Typhoon | 33.4 | 17.6 | 45.4 | 18.8 | 62.0 | 14.0 | 24.0 | 43.8 | 10.9 | 45.4 |
| Plum rain | 82.5 | 68.7 | 84.0 | 68.7 | 42.8 | 20.5 | 36.8 | 43.8 | 28.3 | 28.4 |

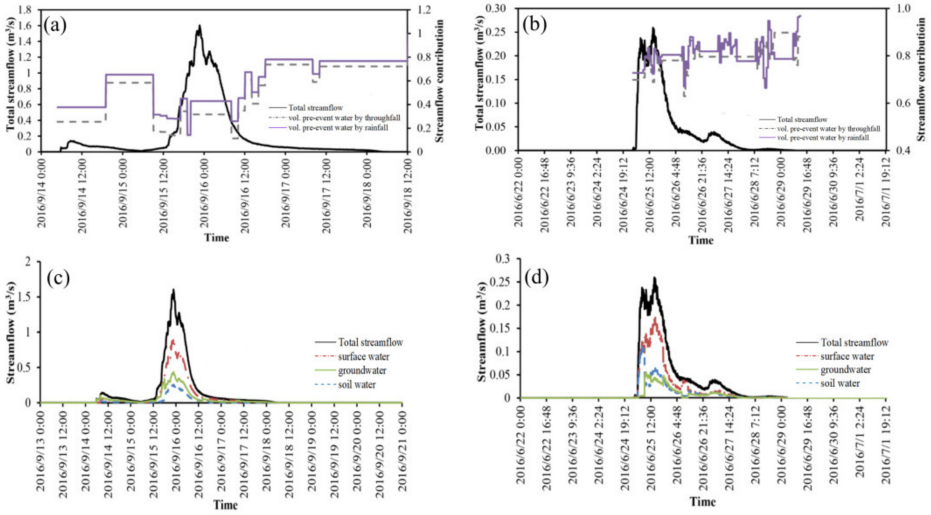



**Figure 5.** Results of isotopic hydrograph separation: (**a**) two-source model for typhoon, (**b**) two-source model for plum rain, (**c**) three-component model for typhoon, and (**d**) three-component model for plum rain.

It can be seen from Table 2 that the pre-event water fractions were lower using throughfall than using precipitation isotopes for both types of storm event; however, the differences were larger in the typhoon event than in the plum rain event (average: 26.6% Vs. 15.3%). We also found that the difference was generally lower for flood peaks than for the whole average values for both rainfall types (typhoon: 15.8% Vs. 26.6%; plum rain: 13.8% Vs. 15.3%).

The distributions of fractions of pre-event water during two storm events are presented in Figure 5a,b. It should be noted that the pre-event water percentages exhibited a large variation in the typhoon event, using either rainfall or throughfall. On average, the pre-event water percentage was 45.4% using rainfall and 18.8% using throughfall. The average pre-event water fraction of the plum rain event was significantly higher (84.0% for rainfall and 68.7% for throughfall) than that of the typhoon event (Table 2). This discrepancy is probably caused by the higher soil moisture content during the long plum rain season from June–July.

Similar phenomena were observed for pre-event water percentages when the flood reached its peak. As the flood crested, the pre-event water percentages were 33.4% for rainfall and 17.6% for throughfall in the typhoon event, while in the plum rain event, they were 82.5% for rainfall and 68.7% for throughfall. In different stages of typhoon event hydrograph, the pre-event water accounted for higher percentages in the rising and recession limbs, while lower percentages in the flood peak, using rainfall or throughfall; the fractions of pre-event water varied more smoothly throughout the whole hydrograph of the plum rain event. Nevertheless, we can draw the conclusion that in a forested watershed, the isotopic values of throughfall and rainfall have strong impacts on hydrograph separation for both rainfall types.

3.2.3. The Contribution of Soil Water Based on the Three-Component Model

In the development of the three-component model for hydrograph separation, soil water plays an increasingly pivotal part in runoff components. Researchers have revealed the fundamental importance of soil water isotopic composition on soil water supply mechanisms, basin runoff formation, and hydrograph separation [34,35]. While the method has been extensively evaluated, the dynamics storage-flux mixing in the soil should be considered, especially when isotopic signals of soil water varied during the event as reported here. In our study, oxygen isotopes and electrical conductivity were used as hydrological tracers in the three-component hydrograph separation. The fractions of surface water, soil water and groundwater are shown in Table 2, and the separation results of the three-component model are presented in Figure 5c,d.

In the typhoon event, the three water component fractions changed prominently, which also reflected remarkable isotopic changes in the water sources. Source fractions in the plum rain event changed greatly in the earlier stage; however, this change tended to be steady, along with rainfall. Fractions of surface water and groundwater simultaneously changed in the opposite trend, while that of soil water fluctuated dynamically (Figure 5d). The average fractions of surface water and soil water were considerably lower in the typhoon event than in the plum rain event, and the fractions of groundwater were higher in the typhoon event (Table 2).

Water source fractions in different types of rainfall events had diverse trends from the beginning to the end of the event episode. From Figure 5c,d, due to the smaller rainfall amount at the beginning of the rainfall, contributions from surface water and soil water remained low, regardless of the type of rainfall and groundwater, giving priority to the discharge in the river. Along with the development of rainfall events, fractions of soil water and groundwater showed contrary trends. Soil water fractions increased first and then decreased until the flooding peaks. This demonstrates that the river was much less influenced by the soil water at the flood peak. In the late period of the event, the percentage of surface water was low and those of soil water and groundwater were high. The event-averaged percentages of surface water, soil water and groundwater were 43.8%, 10.9% and 45.4%, respectively, for the typhoon event and 62.0%, 14.0% and 24.0%, respectively, for the plum rain event. The results showed that soil water occupied a certain proportion in the water sources, indicating that soil water could not be neglected when separating hydrographs.

In the period of the flood peak, an increasing fraction of surface water was found, achieving 40~60%. This means soil water and groundwater together contributed approximately 40%. The two-component model underestimated the fraction of event water, when larger fractions of surface water were discharged during the flood. The maximum fraction of soil water usually occurred adjacent to the flood peak, instead of at the flood peak time, when the fractions of soil water were markedly lower, 14.0% for the typhoon and 20.5% for the plum rain.

## 4. Conclusions

The hourly isotopic data from different compartments of the catchment hydrological cycling were sampled during typical typhoon and plum rain in this study. The measurements indicated that $\delta^{18}O$ in precipitation and throughfall exhibited significant temporal variations, and the throughfall was more enriched in heavy isotopes than the direct rainfall. Isotope values in precipitation during the typhoon event had stronger variation compared with those during the plum rain event. Compared with rainfall, isotope concentrations in surface water were more damped. The groundwater $\delta D$ and $\delta^{18}O$ remained stable during the two-type storm events. $\delta D$ and $\delta^{18}O$ of soil water had significant variations, and the variation was more significant in the typhoon event than in the plum rain event.

The two-component isotopic hydrograph separation model results revealed that: (1) pre-event fractions were higher for the plum rain than for the typhoon event; and (2) fractions in the typhoon event exhibited remarkable variation compared with the moderate

variation in the plum rain. Furthermore, the pre-event water fraction was overestimated by 26.6% in typhoon storm and by 15.3% in plum rain when using rainfall isotopic value, compared to the results of using throughfall isotopic value in this watershed. This indicates that, in the forested watershed, the difference between rainfall and throughfall isotopic values should be considered in both rainfall types for hydrograph separation.

The results of the three-component isotopic hydrograph separation model in the Hemuqiao showed that fractions of surface water and groundwater had inverse dynamic patterns, and fractions of soil water varied dynamically in-between, accounting for 10.9% and 28.3% of the total stream flow in the typhoon and the plum rain events, respectively. The calculated three-source fractions in the typhoon event changed significantly during the course of rainfall, and the fractions in plum rain varied more strongly in the earlier stage of the event and tended to be steady in the following stage.

This paper comprehensively compared the differences between rainfall and throughfall and investigated the impacts of considering temporal variations of rainfall during the event on the isotopic hydrograph separation based on the two-component method. The importance of soil water isotopic values was further explored based on the three-component method. Based on our study, further studies focusing on the influence of both temporal and spatial variations of different water sources on isotopic hydrograph separation are of high interest. Furthermore, the coupling of isotope and hydrological models will improve our understanding of basin runoff generation and concentration and will provide advice for basin water resource planning and flood management.

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