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<td>Chen, Xi; State Key Laboratory of Hydrology-Water Resources and Hydraulic Engineering, Hohai University Zhang, Zhicai; Hohai University, State Key Laboratory of Hydrology-Water Resources and Hydraulic Engineering Soulsby, Chris; University of Aberdeen, School of Geosciences Cheng, Qin-Bo; Hohai University, College of Hydrology and Water Resources Binley, Andrew; Lancaster University, Lancaster Environment Centre Jiang, Rui; Hohai University, State Key Laboratory of Hydrology-Water Resources and Hydraulic Engineering Tao, Min; Hohai University, State Key Laboratory of Hydrology-Water Resources and Hydraulic Engineering</td>
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Characterizing the heterogeneity of karst critical zone and its hydrological function: an integrated approach

Xi Chen¹²*, Zhicai Zhang²³, Chris Soulsby³, Qinbo Cheng²⁴, Andrew Binley⁴, Rui Jiang², Min Tao²

¹: Institute of Surface-Earth System Science, Tianjin University, Tianjin China
²: State Key Laboratory of Hydrology-Water Resources and Hydraulic Engineering, Hohai University, Nanjing 210098, China
³: School of Geosciences, University of Aberdeen, Aberdeen AB24 3UF, United Kingdom
⁴: Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, United Kingdom

*Corresponding author E-mail: xichen@hhu.edu.cn

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Abstract
Spatial heterogeneity in the subsurface of karst environments is high, as evidenced by the multi-phase porosity of carbonate rocks and complex landform features that result in marked variability of hydrological processes in space and time. This includes complex exchange of various flows (e.g. fast conduit flows and slow fracture flows) in different locations. Here, we integrate various “state-of-the-art” methods to understand the structure and function of this poorly-constrained critical zone environment. Geophysical, hydrometric and tracer tools are used to characterize the hydrological functions of the cockpit karst critical zone in the small catchment of Chenqi, Guizhou province, China. Geophysical surveys, using electrical resistivity tomography (ERT), inferred the spatial heterogeneity of permeability in the epikarst and underlying aquifer. Water tables in depression wells in valley bottom areas, as well as discharge from springs on steeper hillslopes and at the catchment outlet, showed different hydrodynamic responses to storm event rainwater recharge and hillslope flows. Tracer studies using water temperatures and stable water isotopes (δD and δ¹⁸O) could be used alongside insights into aquifer permeability from ERT surveys to explain site- and depth-dependent variability in the groundwater response in terms of the degree to which “new” water from storm rainfall recharges and mixes with “old” pre-event water in karst aquifers. This integrated approach reveals spatial structure in the karst critical zone and provides a conceptual framework of hydrological functions across spatial and temporal scales.
Key words: Cockpit karst; critical zone; hydrological functions; geophysical survey; tracers; stable isotopes.
1 Introduction

Karst covers ~10% of the continents and about one quarter of the global population is completely or partially dependent on drinking water from karst aquifers (Ford and Williams, 2013). The southwest China karst region is one of the largest continuous karst areas, covering ~540×10^3 km^2 over eight provinces. The karst terrain displays a geomorphic transition as the topography gradually descends by 2,000 metres over 700 kilometres from the western Yunnan-Guizhou Plateau to the eastern Guangxi Basin. Cockpit karst is a specific geomorphology found in some tropical areas underlain by limestone formations. Conical hills and star shaped valleys are characteristic of such karst landscapes. Due to the distinct nature of karst geology and geomorphology in the humid tropics and subtropics, spatial heterogeneity in the subsurface is high, evidenced by specific landforms features (e.g. heavily fractured outcrops, sinkholes etc.) and complex subterranean conduit networks. This leads to highly dynamic variability of hydrological processes in space and time. This heterogeneity in karstic environments and their rapidly evolving nature makes them extremely vulnerable to natural and anthropogenic hazards. Hillslope springs and groundwater in valley bottom depressions in cockpit karst areas are the main water resources for local agriculture, industry and domestic use. However, the high hydrological variability results in vulnerability to the frequent occurrences of floods and droughts. Consequently, understanding the karst critical zone, its structures and hydrological functions in the southwest karst region of China is essential to mitigate natural disasters and adapt appropriate management strategies for sustainable water resource utilization.
Hydrological responses to rainfall in cockpit karst areas can be conceptualised as in Fig 1. The hydrological processes include rainfall infiltration in the soil and epikarst, water storage and flow in the transmissive zone, and hydrological connectivity between hillslope and depression units. To consider the effects of marked heterogeneity in the profile of the karst critical zone on hydrological function, the profile can be sub-divided into soil, epikarst, unsaturated and phreatic zones (Perrin et al., 2003). On the hillslope, since the permeability usually reduces with the depth (Williams, 1983) or less permeability layers underlie the epikarst zone, infiltrated water usually flows out as a spring. The phreatic zone is regarded as a transmissive zone through its well-developed conduit network (e.g. Emblanch et al., 2003; Perrin et al., 2003). Nevertheless, it is extremely heterogeneous in space. For example, the solutinal conduits in karst aquifers connect with intergranular pores and fractures (often termed as matrix porosity), showing dual or even triple porosity zones (Worthington et al., 2017). Thus, karst aquifers are often conceptualized as dual porosity systems as residence times in the matrix are several orders of magnitude longer than those in the conduits (Goldscheider and Drew, 2007). However, even for the matrix porosity at a specific site, fracture apertures can widely vary, e.g. 0.01–0.1 mm (Long et al. 1982; Hyman et al. 2015); and for larger conduits, the density and connectivity with the surrounding fractures can vary greatly due to heterogeneity in the stratigraphy and geologic structure. Consequently, the characterization of the functioning of such complex systems remains a great challenge.

Hydrometric observations and tracer investigations are traditional and effective approaches to inferring flow processes within karst systems (Fig 1). Water table response is
particularly sensitive to changes in hydrometeorological forcing because of the direct connectivity of the water table with the ground surface in surficial and shallow aquifers (Winter, 1999; Healy and Cook, 2002; Sophocleous, 2002; Lee et al., 2006). The water-table rise and fall during a specific storm period can be translated into a corresponding amount of groundwater recharge and discharge (Sophocleous, 1991; Xie and Yuan, 2010; Fan et al, 2014; Biswas et al, 2017). In karst areas, abundant springs on hillslopes and at catchment outlets reflect regional flow accumulation along conduits and underground channels. Spring hydrographs can be used to characterize karst systems (Kovacs and Perrochet, 2008) and better understand system behavior during floods (Winston and Criss, 2004) or droughts (Fiorillo, 2009).

As water flowing through soils or fractures carries sensible heat that can affect subsurface water temperatures on mixing (Anderson, 2005; Rau et al., 2014), distributed temperature measurements can provide excellent indications of flow and connectivity. Using water temperature as a tracer can thus help assess groundwater flow, groundwater-surface water exchanges (Becker et al., 2004; Niswonger et al., 2005; Lowry et al., 2007; Irvine et al, 2015), flow paths, mixing effects and residence times (Sun et al., 2016). This is especially useful in karst aquifers as fast flows overcome limitation of heat tracing at lower velocities (Rau et al., 2010, Rau et al., 2014). As rainwater may has different temperatures to groundwater, the arrival of new recharge at a karst spring is evidenced by changing water temperature (Ford and Williams, 2013). Subsurface temperature variability can thus be used
to assess the degree of connectivity and relative importance of mixing (Hartmann et al., 2014; Mudarra et al., 2014).

Isotope studies have also substantially increased our understanding of detailed hydrological flow paths and mixing processes in the critical zone (Sprenger et al., 2017, 2018). Stable isotopes of water (δ[^18]O and δ[^2]H) have also been widely used to provide insight into the functioning of karst systems (Barberá and Andreo, 2012; Mudarra and Andreo, 2011), the mixing of water from different sources (Aquilina et al., 2006; Plummer et al., 1998), and the their residence times (Batiot et al., 2003; Long and Putnam, 2004). Due to the high degree of variability of karst systems, continuous monitoring or event-based high-frequency sampling is essential, but logistically challenging (Goldscheider and Drew, 2007).

Hydrometric monitoring and tracer studies provide direct observations of hydro-dynamics which can indirectly hypothesize karst critical zone structures. However, explaining hydrological variability and identifying controls rely on detail information on karst hydrogeological properties in space. For example, Lee and Krothe (2001) used dissolved inorganic carbon (DIC) and δ[^13]DIC as tracers for conceptualizing hydrological functions using a four-component mixing model (rain, soil water, epikarstic water and diffuse phreatic water) for a karstic flow system. Similarly, Jiang et al. (2008) inferred runoff response thresholds in the epikarst by monitoring the spring’s discharge, pH, electrical conductivity, temperature and rainfall. Increasingly, however, geophysical techniques are being used to provide more direct evidence about the geological setting of a karst system. Survey techniques, such as ground penetrating radar (GRP) and electrical resistivity tomography.
(ERT), provide an efficient means of describing the karst structure, mapping karst conduits
(Carrière et al., 2013; Martínez-Moreno et al., 2014) and defining the soil-rock interface
(Chalikakis et al., 2011). Such geological evidence is crucial for delineating hydraulic
structures, such as complex subsurface drainage networks in the karst which can link local
fissure and fracture networks with the dominant large conduit systems (Chalikakis et al, 2011;
Hartmann et al., 2014; Binley et al., 2015). These insights can be particularly powerful if
geophysical approaches are integrated with hydrometric and tracer observations and at the
same study site (e.g. Soulsby et al., 2016).

Different methodologies have advantages and limitations (see Goldscheider and Drew,
2007) so isolated measurements and observations are inadequate for characterizing a karst
system. Although many researchers apply combinations of such methods for assessing the
hydrology karst areas, they seldom integrate insights from multi-scale distributed geophysical
surveys, hydrometric observations and tracer investigations. Consequently, in the southwest
karst region of China, we lack a conceptual framework that characterizes spatial heterogeneity
hydrological functioning at multiple scales and/or in different geographic regions.

The overall aim of this study was to integrate these approaches to visualize the complex
heterogeneity in karst critical zone and its effect on hydrological functions. By so doing, the
study will also help identify dominant processes to incorporate in the structure of hydrological
models. We based the investigation in the long-term study site of Chenqi in the cockpit karst
region of Guizhou province, southwest China. Our objectives were to: (1) undertake
geophysical surveys using electrical resistivity tomography to characterize the structure of the
karst critical zone and identify variations in the permeability which define fast and slow flow domains in karst aquifers. (2) Monitor groundwater levels in wells and discharge from hillslope springs and the catchment outlet to characterize the spatial and temporal variations of hydrodynamic response to rainfall events. (3) Use stable isotopes of water ($\delta^{18}$O and $\delta^2$H) qualitatively understand how “new” event water from rainwater and “old” pre-event water from low permeability aquifers recharges mix and contribute to catchment discharge, particularly during heavy rainfall in the vertical profiles. Improved conceptualization of a dual flow system that accounts for local aquifer heterogeneity influences on the hydrological connection between fast and slow reservoirs is proposed.

2 Study area, data and methods

2.1 Study catchment

The study was focused in the small (1.2km$^2$) Chenqi catchment, located in the Puding Karst Ecohydrological Observation Station, Guizhou Province, southwest China (Fig. 2). It is a typical cockpit karst landscape, with surrounding conical hills separated by star shaped valleys. Surface elevation ranges from 1320m at the catchment outlet to a maximum of 1500 m. Geological strata in the basin include dolostone, thick and thin limestone, marl and Quaternary soil (profiles of A-A’ and B-B’ in Fig 2). Limestone formations form the higher elevation areas with 150-200 m thickness, which lie above an impervious marlite formation.

The Quaternary soils are irregularly developed on carbonate rocks and unevenly distributed, with outcrops of carbonate rocks covering 10-30% of the catchment. Dominant vegetation
ranges from deciduous broad-leaved forest on the upper and middle parts of the steep
hillslopes and corn and rice paddy at the low of the gentle hillslopes and depression.

The catchment is located in a region with a subtropical wet monsoon climate with mean
annual temperature of 20.1°C, highest in July and lowest in January. Annual mean
precipitation is 1140 mm, almost all falling in a distinct wet season from May to September
and a dry season from October to April next year. Average monthly humidity is high, ranging
from 74% to 78%.

2.2 ERT survey in the valley depression

Electrical resistivity tomography (ERT) was used to survey the geological properties
along five transects crossing the depression (Fig 3). The surveyed profiles were located
adjacent to four observation wells (see below) and three sinkholes to identify the geological
controls on groundwater dynamics. The surveys were carried out in spring 2017 using a
Syscal Pro (Iris Instruments, France) resistivity meter with electrode spacing between 2 and
5m. We used a dipole-dipole electrode configuration (see for example, Binley, 2015) with
dipole spacings of one, two and three times the electrode separation and up to 20 levels. The
dipole-dipole configuration was selected to allow sensitivity of lateral variation in resistivity.

Inversion of the ERT data was carried out using the code R2
(http://www.es.lancs.ac.uk/people/amb/Freeware/R2/R2.htm). The resulting composite ERT
image is shown in Fig 3. The resistivity ranges from ~15 Ωm to ~8000 Ωm. Based on ERT
surveys carried out adjacent to outcrops in neighbouring areas of the catchment we interpret
the ERT results in Fig 3 as: (i) an upper layer consisting of moist soils or extensively
fractured rock (resistivity <100 Ωm, blue in Fig. 3); (ii) carbonate rock with a high secondary porosity (and hence permeability) (100 Ωm < resistivity < 1000 Ωm, green/yellow in Fig. 3); (iii) an underlying carbonate rock with low secondary porosity and hence relatively low permeability (resistivity >1000 Ωm, red in Fig. 3).

The ERT image shows that the depression aquifer is highly heterogeneous in both the horizontal direction and vertical direction. Based on our interpretation of the ERT image: well W1 is surrounded by the less permeable aquifer (red colour) and its upper layer of carbonate rock appears particularly impermeable; wells W3 and W4 are located in the higher permeability area (blue colour); well W5 is located in the less permeable aquifer and its top soils (blue colour) and the carbonate rock in the upper layer (yellow colour) is relatively more permeable than that in the deep layer (red colour). The relatively permeable upper layer at W5 appears much thinner than those at other wells.

In a previous study, a GPR MALA Professional Explorer (ProEx) System was used for investigation of the fracture zone thickness on the hillslope. The epikarst thickness on the hillslope was identified to be in a range of 7.6~12.56 m. Along the hillslope, the thickness and the epikarst zone at the lower areas is deeper than in the upper areas (Zhang et al. 2013).

2.3 Hydrometric observations

In the Chenqi catchment, groundwater levels in the valley depression were routinely monitored at the four wells (W1, W3, W4 and W5) with depth to ground surface of 35, 23, 13 and 16 m, respectively. The well screening was installed over the whole depth for each of the wells to reflect local flow exchanges at various depths. Flows discharging from a hillslope
spring (HS) at the foot of the eastern steep hillslope and leaving the catchment outlet were measured by v-notch weirs (Fig. 2). The water level and temperature at each well, the hillslope spring and the catchment were automatically recorded by HOBO U20 water level logger (Onset Corporation, USA) with a time interval of 15 minutes. Additionally, an automatic weather station was set on the upper hillslope to record precipitation, air temperature, and air humidity and pressure.

Data collection ran from 28 July 2016 to 30 October 2017. Hourly variations of rainfall, discharge from the hillslope spring and catchment outlet, as well as water levels in the four wells are shown in Fig 4, and their corresponding temperature are shown in Fig 5. Statistical characteristics of water levels, flow discharges and temperatures are summarized in Table 1 and Table 2.

2.4 Stable isotope analysis

For isotope analysis, the hillslope spring (HS), groundwater from outlet and rainfall were sampled at daily intervals during the wet season from June to August 2017. Additionally, they were intensively sampled during eight rainfall events in the wet season using an autosampler set to hourly intervals. Depression groundwater was sampled from the four wells during four rainfall events within the study period; in each event, water samples were collected before, during and after rainfall. At each well, water was sampled from multiple depths with a depth-specific sampler to give a profile of the isotopic composition of the groundwater column. All water samples (1695 replicates) were collected by 5 ml glass vials. The stable isotopic composition of $\delta^2$H ($\delta$D) and $\delta^{18}$O ratios were determined using the MAT 253 laser
isotope analyser (the instrument precision ±0.5‰ for δ²H and ±0.1‰ for δ¹⁸O). Isotope ratios
are reported in the d-notation using the Vienna Standard Mean Ocean Water standards
(Coplen, 1994). The analyzed results of the isotopic values are shown in Figs 6-8.

3 Results

3.1 Hydroclimatic and groundwater variability in the depression

Generally, the hydrometric observations reveal sharp rises and falls in water level at the
four wells in response to rainfall events (Fig. 4). However, the magnitude of the water table
response and the rate of the water table recessions exhibit differences at the four wells, which
indicates spatial differences in the groundwater response to recharge and attenuation of the
hydrograph. The magnitude of the temporal dynamic is larger and more attenuated at W3 and
W4 in the low electrical resistivity (interpreted higher permeability) areas, and abrupt at W1
and W5 where particularly heavy rainfall is needed to produce a large water table rise. These
differences are apparent in the coefficient of variation (CV) of water level data for W3 and
W4 (0.21 and 0.61, respectively), whilst the CV of W1 and W5 is only 0.03 and 0.07,
respectively (see Table 1).

Temporal variability in temperature clearly demonstrates that groundwater dynamics at
W3 and W4 are more directly influenced by hydroclimate variability compared with W1 and
W5 (Fig 5). The former wells show marked temperature excursions towards air temperatures
in response to rainfall events, whilst the latter wells exhibit highly attenuated temperatures
typical of deeper groundwater. As shown in Table 2, the CV of water temperature at W3 and
W4 is 0.026 and 0.036, while the CV of W1 and W5 is only 0.004 and 0.012, respectively. The sharp rise and drop of water temperature at W3 and W4 during rainfall is clearly consistent with the fast arrival of freshly infiltrated rainwater. In contrast, the limited seasonal fluctuation of water temperatures at W1 and W5 indicate little evidence of rainfall recharge consistent with the interpreted low permeability in the upper layers (high resistivity zone in Fig 3).

As shown in Fig. 6 and Table 3, temporal variations of rainfall $\delta D$ and $\delta^{18}O$ at Chenqi are marked, ranging from -120.2 to -17.9‰ for $\delta D$ and from -16.4 to 0‰ for $\delta^{18}O$. The $\delta D$ and $\delta^{18}O$ responses amongst the depression wells are attenuated or damped but they are markedly different in this restricted area due to spatial heterogeneity of the karst aquifer (Table 3 and Fig 6). In terms of the temporal changes in $\delta D$ and $\delta^{18}O$, values tended to become more negative in response to direct effects of rainwater recharge into the aquifer during storm events. Compared with isotope values at W1 and W5, groundwater at W3 and W4 is clearly receiving rainfall that has marked changes in isotope values evident in the variability in Fig 6 and the large range in Table 3. Groundwater at W5 is the most stable (Fig 6) and has the lowest range in Table 3 implying a limited influence of new recharge from rainfall. However, W1 is close to the catchment outlet and the high permeability of its lower aquifer inferred from the ERT survey may allow exchange with the subsurface flow leaving the catchment (Fig 3). The isotopic signatures of groundwater at W1 show intermediate isotopic values compared to the upper catchment (lower than the most stable water at W5 and
higher than those at W3 and W4 where the direct rainfall influence is evident, suggesting that groundwater at W1 likely reflects mixing of flow paths from the upper depression.

However, at the different stages of the storm event response (e.g. the sharp rises and falls in water level before and after rainfall), the degree of “new” event water recharge from rainfall and “old” water release from previous storage amongst the four wells differed according to the δD values from the depression aquifer (Fig 7). Generally, recharge of new rainfall occurs in the early recession phase of the groundwater response, and the release of previously stored (old) water occurs later in the recession. The impact of infiltrating new water in the early recession can be distinguished from changes of the δD values before rainfall and in the early recession in Fig 7; for rainfall events No. 1 and 4, as the rainwater δD values are much more negative than those of the groundwater. The subsequent decline of the δD values in these depression wells indicates the ingress of isotopically depleted (newer) rainwater. However, for rainfall event No 3, as the rainwater δD is more enriched than groundwater, the rise of the δD values at these depression wells also indicates the impact of more enriched rainwater recharge.

In the later recession, the effects of recent recharge generally decline. Hence, the δD values of groundwater increase again for rainfall events No 2 and 4. This would be consistent with the release of older water into aquifer. As shown in Fig 7, changes of the δD values in the three events are evident for wells W3, W4 and W5, but the changes become less distinguishable for well W1 close to the catchment outlet (Fig 7). This also indicate that
groundwater at the catchment outlet can be attributed by regional flow that endures a relatively high mixture of event new water with storage old water.

### 3.2 Hillslope-depression flow connectivity

Hillslope-depression flow connectivity can be identified from temporal variability in water table levels, temperatures and isotopic tracers at W4 located at the base of a steep hillslope and the hillslope spring (HS). Despite the marked response to rainfall events, the water level at W4 recesses much more slowly after rainfall ceases compared to the other wells (Fig 4). The slow recession in this interpreted high permeability area possibly arises from some additional water from upslope areas that continues to recharge the depression aquifer after rainfall ceases.

Water temperatures and isotopic values at W4 and the hillslope spring (HS) further provided evidence for hillslope contributions to the depression. Water temperatures at both sites show a similar pattern of the seasonal variations with air temperature and fluctuations with rainfall (see Fig 5), i.e. higher in the summer season and lower in the winter season. Water isotopic values at W4 are closest to those at the hillslope spring (HS), particularly, during the rainfall period (see the yellow for W4 and the green for HS in Fig 6 and mean values in Table 3).

Additionally, in the winter season, water temperature at W4 is more attenuated and lags behind that of the hillslope spring, which indicate that the hillslope flow is faster than the depression groundwater flow at W4. In the summer season, the response of water temperature to rainfall at W4 is more marked that those of the hillslope spring, which indicate that the depression groundwater flow at W4 could receive hillslope flow as well as rainfall recharge.
3.3 Effect of vertical heterogeneity on hydrological functions

The ERT survey demonstrates strong vertical heterogeneity of the karst aquifer at each site (Fig 3). The δD values at various depths to groundwater table (the uncertainty bar in Fig 7) illustrates the effect of vertical heterogeneity on hydrological functions. For the four wells, the range of the vertical variation in δD is generally much higher for W1 and W3 in the central and outlet depression, than the wells of W4 and W5 at the upper depression. This is consistent with groundwater in the upper depression areas mixing more effectively in a vertical direction than groundwater in the center and catchment outlet.

The isotopic values prior to rainfall and at the early and late stages of the recession for rainfall event 4 (12 to 14 August 2017) in Fig 8 further demonstrate the control of vertical heterogeneity in aquifer structure on hydrological function. The event has a total rainfall of 36.8 mm and the rainfall δD value ranging between -109.8 and -74.8‰ with a mean of -90.7‰, with the most negative values coinciding with peak rainfall. The event-scale changes in the vertical distribution of the δD values are different for the four wells.

(1) **W1:** there is little vertical variation of the δD values before rainfall (Fig 8a(i), which indicates strong pre-event vertical mixing. Early in the recession when groundwater levels rapidly decline, the mean δD value declined from -60.5 to -61.6‰ (Fig 8a(i)), indicating some new water influence. Moreover, the flow paths of the new water ingress can be identified from the vertical variation of the δD values (see 13/8 line in Fig 8a(ii)), i.e. at the depths of 7-8m and below 18m where the δD values rapidly decline. These depths correspond to low resistivity regions indicative of high permeable layers (green in Fig 8a(iii)). Since the
temperature variation in Fig 5 and ERT image in Fig 8a(iii) suggest that the lower layer at W1 receives little rainfall recharge, the decline of the δD values seems most likely explained by exchange of groundwater in the high permeability depths with the regional flow (affected by new rainfall). Between the two higher permeability layers, water in the less permeability layer (yellow in Fig 8a(iii)) changes only little, indicating that flow exchange between the permeable and less permeable layers is weak.

Later in the recession, the δD value increases to -60.8‰, which is close to the pre-rainfall value. At this time, vertical variations on 14 August (Fig 8a(ii)) demonstrate that the δD value in the lower permeable layer (below 18m) recovers close to pre-event conditions. Surprisingly, the δD values in the upper less permeable layer (the depth of 7.5 m) become much less negative. This suggests that there may be release or displacement of older water, possibly attributable to antecedent storage from the upper less permeable layer (Fig 8a(iii)).

(2) **W3**: temporal variation of δD values over the three periods (Fig.8b(i)) is similar but more marked than that at W1. Vertical variation at W3 (Fig 8b(ii)) is very different over the three periods. Before the rainfall on 12 August, the δD values generally are less negative throughout the water column. The more negative values in the upper profile may reflect more recent depleted rainfall and increasingly older water with depth. This seems consistent with the decreasing permeability with depth at W3 implied by the less resistive upper profile and more resistive deeper layers (Fig 8b(iii)).

In the early recession on 13 August, δD values (Fig 8b(ii)) become more negative and almost uniform in vertical distribution. This implies that new water from recharge of
rainwater rapidly mixes with older pre-event water in storage. In the later recession, groundwater isotope values recover indicating lessening influence of new water, though values do not recover to pre-rainfall levels. Meanwhile, groundwater in the upper aquifer (e.g. above 8m) still remains negative with continued new water influence. This suggests that the upper, more permeable (i.e. with lower resistivity) aquifer stores more new water during rainfall and releases this thereafter.

(3) **W4**: as shown in Fig.8c(i), the water level recession is slower than the other wells but the mean of the δD value declines greatly from -59‰ before rainfall to -67‰ in the early recession and then recovers to -65.9‰ in the later recession. The marked decline of δD is again consistent with substantial influence of event water ingress. Prior to rainfall when groundwater level is low (>4m in depth), the vertical distribution of the δD is nearly uniform (Fig 8c(ii)). In the early recession when water levels remain high, groundwater in the upper aquifer (above the depth of 3.5) tends to be more negative indicating event new water influence.

In the later recession, the δD values below 3.5m become a little more negative, whilst the upper aquifer remains the same. As W4 is located at the foot of a steep hillslope, it is likely that lateral flow from the hillslope contributes recharge since the ERT image suggests that this is a high permeability area strongly connected to the hillslope flow path (Fig 3 and Fig 8c(iii)).

(4) **W5**: The water level and isotopic response at W5 contrasts markedly to the other wells, i.e. a transient fluctuation of water level and abrupt change in δD (Fig 8d(i)).

unresponsive groundwater likely reflects that the well is located in an impervious (high resistivity) area (Fig 8d(iii)). After the water level decline, groundwater has a limited decrease in δD (from -58 to -58.4‰) in the early post-event period, and continuous decrease (to -59.6‰) later. The continuous decrease of δD values indicates a longer memory of the much less permeability aquifer to the limited recharge.

Before rainfall, the vertical values of δD are highly varied; water in the upper layer has less negative values of δD, whilst waters at depths of 7.4m and below 8m have more negative values of δD (Fig 8d(ii)). The much older water in the upper layer suggests strong evaporative effect in the non-rainfall period since the top soils and the upper permeability rock layer are thin (Fig 8d(iii)) and thus evaporative effect is strong. Marked changes of the δD values in the lower depths (e.g. below 7.4m) reflects that there are still some permeable fractures, in which new water could be arrival. After the end of the brief water table response, the vertical δD values (13 August in Fig 8d(ii)) in the lower layer (e.g. below 7.4m) are more negative than the upper layer. This would imply that after a short response to rainfall (Fig 8d(i)), event “new” water in the upper layer rapidly mixes with the surrounding “old” water, but “new water” still ingresses into the lower depths. In the later post-event period, the relatively uniform vertical values of δD (14 August in Fig 8d(ii)) show recharged event water mixing in the whole profile.

4 Discussion and conclusions

4.1 Discussion

4.1.1 Conceptualizing hydrological functions in cockpit karst catchment
Variation in hydrological functions (rainfall recharge, horizontal and vertical flow exchange at sites and hillslope-depression connectivity) in the heterogeneous subsurface of our cockpit karst catchment can be summarized as in Fig 9. Fast and frequent rainwater recharge $I_f$ into the epikarst zone on the hillslope (HS) and permeable depression (e.g. W3 and W4) leads to local flow being “active” and young, whilst infrequent rainwater recharge into the less permeable depression (e.g. W1 and W5) leads to local flow being less active or inactive and “old”. For the hillslope-depression-outlet connectivity, the mean isotopic value at outlet ($\delta D$) is between relatively “new” water from fast flows in HS and high permeability depression areas (W3 and W4) ($\delta D_f$) and “old” water from slow flows in low permeability depression areas (W1 and W5) ($\delta D_s$). Thus, outlet water (Q and $\delta D$) can be viewed as a mixture of hillslope ($Q_f$ and $\delta D_f$) and depression flow ($Q_s$ and $\delta D_s$) at various sites.

Fig 8 shows that temporal change in hydrological functions is closely linked to vertical heterogeneity with different composition of the active, less active and inactive areas. The structure of the vertical profile can be generalized as Fig 9 and classified into two types: (a) profiles consisting of two different permeability layers, e.g. a high permeability layer confined by a lower permeability layer at W1 and a high permeability layer with limited thickness perched on a less permeability layer at W5. (b) profiles comprising one layer but their permeability decreasing with depth, e.g. at W3 and W4. Here, the profile flow function can be still illustrated by exchange of fast and slow flow reservoirs (Fig 9) but the relative contribution of fast and slow flow reservoirs is a function of depth $h$, i.e. $Q \delta D(h) = Q_f \delta D_f(h) + Q_s \delta D_s(h)$, in where flows and isotopic values in each reservoir are influenced by rainfall.
recharge $I_f$ and water exchange between fast and slow reservoirs $Q_E = K_E(h)(WT_f - WT_s) (WT_f$

and $WT_s$ represent water table in the fast and slow reservoirs, respectively, and $K_E$ is exchange

coefficient).

Such depth variability of hydrological function can be used to explain the hydrological
response of the observation wells (Fig 9). The profile depth $h$ at W1 and W5 can be divided
into two homogenous layers ($h1$ and $h2$). At W1, we can hypothesize that recharge of the fast
flow reservoir ($I_f$) is negligible due to the upper confining layer. The fast flow $Q_f$ can be
highly connected with the regional flow but has limited exchange with the slow flow reservoir
$Q_s$, i.e. $Q_E$ is small, evidenced by little change of the $\delta$D value in the less permeable layer for
W1 in Fig 8a(i). At W5, groundwater in the profile is dominated by the slow reservoir $Q_s$ in
the lower layer ($h2>>h1$). Before rainfall, when the water level $WT_s$ is low, limited perched
water $Q_f$ in the upper layer ingresses into the slow reservoir ($Q_E$ can be neglected). During
rainfall when the upper water table $WT_f$ rises higher than the low water table $WT_s$, pressure
drives new event water $Q_f$ into the slow reservoir in a short time ($Q_E$ is slight and transient). After rainfall when a short pulse of water table variation ends ($WT_f<=WT_s$), the pressured
new water in the slow reservoir $Q_s$ is slowly released.

The hydrological functions in the profiles W3 and W4 can be still conceptualized into
the fast and slow flow reservoirs in which fast flow contribution ($Q_f$) to the whole profile flow
gradually reduces with depth whilst the slow flow contribution ($Q_s$) increases. As the profile
is highly permeable and unconfined, direct recharge into the fast reservoir ($I_f$) and new event
water quickly mixes with pre-event water in the vertical direction under $WT_f>=WT_s$ during
rainfall (large $Q_E$). In the later recession when water table is low and $WT_f$<=$WT_s$, the older water in the slow flow reservoir is mostly released from the lower layers (Fig.9b), i.e. less negative $\delta D_s(h)$ and larger $Q_s$ in $Q\delta D(h)=Q_f\delta D_f(h)+Q_s\delta D_s(h)$. The unconfined aquifer at W4 is similar to W3 but the aquifer at W4 receives direct recharge ($I_f$) and inputs from hillslope flow ($Q_h$) during events when water levels are high, then quickly infiltrates into the low slow reservoir (large $Q_E$). After rainfall, the upper fast reservoir retains a water level higher than the slow reservoir ($WT_f$>$WT_s$) since the hillslope flow ($Q_h$) continually inputs into the aquifer (evidenced by more negative of the $\delta D$ values in Fig 7c(ii)).

In any specific landscape unit (e.g. depression), outlet flow $Q$ comes from fast and slow flow composition at various sites, i.e. $Q=\sum^n Q_f+\sum^n Q_s$, and $Q\delta D=\sum^n Q_f\delta D_f+\sum^n Q_s\delta D_s$, where $n$ is the total number of the sites. Meanwhile, in each reservoir, the discharge ($\sum^n Q_f$ or $\sum^n Q_s$) and the mass ($\sum^n Q_f\delta D_f$ or $\sum^n Q_s\delta D_s$) are influenced by rainfall recharge $I_f$ and flow exchange $Q_E$. More specifically, considering vertical heterogeneity at the sites, outlet flows $Q$ and mass $Q\delta D$ can be expressed by: $\sum^n \sum^1 Q_f+\sum^n \sum^2 Q_s$ and $\sum^n \sum^1 Q_f\delta D_f+\sum^n \sum^2 Q_s\delta D_s$, respectively, where $n1$ and $n2$ are the respective number of permeable and less permeable layers in a profile.

4.1.2 Benefits of an integrated approach

Karst aquifers have distinct hydraulic structures and behaviors and therefore require specific investigation methods (Goldscheider and Drew 2007). Capturing the hydrological functions requires hydrometric observations and tracer sampling at sub-hourly intervals as shown in our study. Current techniques that deploy loggers with the capacity to monitor in
real-time and the capability to transfer data remotely are particularly useful (Luhmann et al., 2015). In general, monitoring water level data is easy and cheap and it reflects the relatively strong control of rainfall frequency on hydrograph shape. Nevertheless, it has limited value in interpreting karst aquifer structures (Jeannin and Sauter, 1998) and identifying the contributions of event and pre-event water. The non-conservative nature of water temperature facilitates insights into conduit size, and the damping and retardation in porous media via an analysis of input and output thermographs (Covington et al., 2011, 2012; Luhmann et al., 2012; Birk et al., 2014; Luhmann et al., 2015). However, reliable identification of the hydrological functions from input and output thermographs need integrate information on heat exchange within karst conduits that may introduce a retardation in the residence times (Luhmann et al., 2015). Comparison of the temporary variation of isotopic values in rainfall with the observed variability in karst spring waters allows not only quantification of mixing processes in discharge as shown in our study, but also to quantify transit time distributions (Hu et al., 2015) and determine groundwater ages.

Selecting representative sites is challenging and important in capturing the large-scale hydrological functions in the karst catchment due to strong heterogeneity, hydraulic discontinuity and anisotropy. Traditionally, tracing and hydrometric observations have been mostly undertaken in the conduit network to study the rapid flow (Goldscheider et al. 2008) and at outlet springs for the overall characterization of karst systems (Kovacs and Perrochet, 2008). However, fractured rocks in the karst critical zone have the permeability ranging over several orders of magnitude. Even at small spatial scales (e.g. the four wells in our study
catchment), there is relatively less active subsurface flow in the high permeability zones, and relatively active subsurface flow in the less permeable zones. Additionally, our analyzed results of new event water recharge and the degree of mixing with pre-event old water indicates that water samples at representative depths to characterize subsurface inflows to wells are necessary for assessing different flow paths in the karst and temporal changes in their hydrological dynamics.

A key contribution of this study was to show how geophysical techniques like ERT surveys can help identifying structural differences which can be incorporated in designing targeted monitoring networks. ERT images combined with tracer characteristics of subsurface water and the associated mixing processes identify which depths/sites in the catchment are representative for monitoring in order to reliably quantify fast and slow flows. As shown in Fig 8, water sampling at permeable layers/sites during rainfall events are required for capturing variability of the hydrological functions in the karst catchment. If the permeability varies with depth, water sampling at various depths are particularly important for quantifying the depth-dependent variability of the hydrological function.

Moreover, ERT images combined with details observations provide insights into how to conceptualize complex karst systems for lumped and distributed modeling. ERT images provide high-resolution visualization of the subsurface, and the relationship between these images and parameters affecting flow and transport (Hubbard et al., 1999). The spatially distributed information from geophysics and isotopic characterisation of subsurface water and the associated mixing processes facilitate tracing water flow sources (e.g. rainfall recharge.
and exchange between high and low permeable sites/layers). This understanding can inform hydrological model structures at different scales (e.g. as conceptualized fast and slow flow reservoirs) and their hydrological connectivity (e.g. hillslope-depression-outlet). The inferred hydrologic characteristics can be then used either independently or combined with direct hydrologic observations to constrain hydrologic properties and reduce uncertainty in hydrological models (Hinnell et al., 2010).

4.2 Conclusions

Understanding the function of the water cycle is a key issue for critical zone science since water is a unifying theme for understanding complex environmental systems (Lin, 2010). Nevertheless, it is a significant challenge to identify active subsurface processes that determine water flows and travel times. In this study, geophysical, hydrometric and tracer tools are used to characterize the hydrological function of the cockpit karst critical zone in the small catchment of Chenqi, Guizhou province, China. The ERT surveys (Fig 2) identified structural features that likely control aquifer permeability and the heterogeneity in observed hydrodynamic response. Hydrometric observations and using water temperatures (Figs 2 and 3) as a tracer clearly identified rainfall recharge and flow recession induced by new rainwater recharge and the constraints of aquifer permeability. Stable water isotopes (δD and δ¹⁸O) (Fig 5) largely corroborated geophysical, hydrometric and thermal data and provided detail qualitative insight into event water recharge into, and pre-event storage release from, the heterogeneous aquifer. In particular, this information can help identify flow paths of new
event water recharging through high permeability zones in both horizontal and vertical
directions in the catchment.

The study illustrated that even in a highly heterogeneous catchment, hydrological functions
can be conceptualized simply into fast and slow flow reservoirs. Nevertheless, using such a
dual flow system in simulation of hydrological processes should be based on detail
observations at representative sites within a catchment. Particularly, since hydrological
functions can vary with depth, observations and water sampling from various permeability
layers are extremely important.

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multiple scales, Water Resources Research, 51(6), 3837-3866, DOI:10.1002/2015WR017016


Table 1 Statistical summary of water level and flow discharge

<table>
<thead>
<tr>
<th></th>
<th>W1</th>
<th>W3</th>
<th>W4</th>
<th>W5</th>
<th>Hillslope</th>
<th>Outlet</th>
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<tbody>
<tr>
<td><strong>Min</strong></td>
<td>1267.4</td>
<td>1272.6</td>
<td>1280.0</td>
<td>1276.4</td>
<td>0</td>
<td>0</td>
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<td><strong>Max</strong></td>
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<td>1279.9</td>
<td>1285.2</td>
<td>1284.0</td>
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<td><strong>Range</strong></td>
<td>8.5</td>
<td>7.3</td>
<td>5.2</td>
<td>7.6</td>
<td>1.4×10^{-3}</td>
<td>0.15</td>
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<tr>
<td><strong>Mean</strong></td>
<td>1273.6</td>
<td>1275.9</td>
<td>1281.8</td>
<td>1278.9</td>
<td>8.5×10^{-5}</td>
<td>4.7×10^{-3}</td>
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<tr>
<td><strong>Cv</strong></td>
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<td>0.21</td>
<td>0.61</td>
<td>0.07</td>
<td>1.73</td>
<td>2.83</td>
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Table 2 Statistical summary of air and water temperature

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Table 3 Statistical summary of isotope data for rainfall, hillslope spring (HS), catchment outlet and depression wells

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<th>Obs</th>
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<th>δ18O (‰)</th>
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<td></td>
<td>Max</td>
<td>Min</td>
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<tr>
<td>outlet</td>
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<tr>
<td>HS</td>
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</tr>
<tr>
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<tr>
<td>W4</td>
<td>6/7~20/8</td>
<td>-55</td>
<td>-70.2</td>
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<tr>
<td>W5</td>
<td>6/7~20/8</td>
<td>-55.7</td>
<td>-67.5</td>
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Figure 1 Sketch map of karst hydrological processes.
Figure 2 Map of geomorphology and hydrological monitoring locations in the Chenqi catchment.

59x53mm (600 x 600 DPI)
Figure 3 ERT image in the study depression corresponding to the survey profiles in Fig 1.

37x21mm (600 x 600 DPI)
Figure 4 Variations of flow discharge for hillslope spring (HS) and catchment outlet and water levels at depression wells (W1, W3, W4 and W5).

52x41mm (600 x 600 DPI)
Figure 5 Variations of air temperature and water temperatures for hillslope spring (HS), outlet discharge and depression wells (W1, W3, W4 and W5).
Figure 6 Variation of the δD values for rainfall, hillslope spring (HS), catchment outlet and depression wells.
Figure 7 Variability of the δD values for depression wells (the bar represents range of δD values along various depths to water surface; black block represents its mean value; solid line represents depth to water table).

85x80mm (600 x 600 DPI)
54x35mm (600 x 600 DPI)
(b)

52x36mm (600 x 600 DPI)
(c)

51x39mm (600 x 600 DPI)
Figure 8 Variability of rainfall, water level and δD values (i), vertical distribution of groundwater δD values (ii) and ERT image (iii) of W1 (a), W3 (b), W4(c) and W5(d). The colour dots in the (i) plots are the mean δD value of different depths.

58x40mm (600 x 600 DPI)
Figure 9 Conceptual model of groundwater aquifer at depression wells of W1, W3, W4 and W5. The high permeability layers in the grey color areas and low permeability layers in the deep grey color areas. WTf on solid lines is water table of fast flow reservoir, WTs on dotted lines is water table of slow flow reservoir, QE is exchange flow between fast (Qf) and slow flow (Qs) reservoirs and If is rainfall recharge.

36x43mm (600 x 600 DPI)