Title: Effects of passive storage on modelling hydrological function and isotope dynamics in a karst flow system in southwest China Author(s): Guangxuan Li, Xi Chen, Zhicai Zhang, Lichun Wang, and Chris Soulsby MS No.: hess-2021-492 MS type: Research article

Responses to the Reviewers:

Reviewer #2:

dear editor

I thought long and hard before refusing this article, but in the end my arguments are as follows:

the bibliography is not up to date, which is annoying to put the study in an international context.

It remains a very local study and the lack of broadening and conclusive perspectives makes this article inappropriate for your journal.

Finally, the article (some figures illegible, bibliographic references badly cited or missing in the list) suggests that this work was done hurriedly.

Finally, this article can be accepted as a second intention with major revisions depending on your analysis

Reply:

We thank the reviewer for his valuable comments and suggestions.

Although Chenqi catchment is small, the geomorphologic characteristics can represent a broad region of headwater catchments in cockpit karst landscapes in the tropics and sub-tropics areas. The cockpit karst covers an area of about 140,000~160,000 km² in China. Such karst morphology also exists in Southeast Asia, Central America and the Caribbean (Huang et al., 2014).

One of the hydrological characteristics of the cockpit karst landscapes is the hillslope - depression flow connections (H-D). In the karst area, since the flow system can be conceptualized into the fast flow (F) and slow flow (S) reservoirs in each of the hillslope and depression units, the hydrological connections include hillslope fast flow - depression fast/slow flow (HF-DF/DS), and hillslope slow flow- depression fast/slow flow (HS-DF/DS). As hillslope and depression fast flow (HF-DF) primarily moves in the connected conduits of the karst catchment, we neglected the connection of HF-DS in this study. Consequently, we consider three possible connections of hillslope - depression fast flow (HF-DF), and hillslope slow flow (HS)- depression fast/slow flow (HS-DF/DS) with a ratio of r_{hd} of HS contributing to DS (Fig. S1). The optimized r_{hd} is 0.39. It means that about 61% of hillslope slow flow can enter depression fast flow reservoir. The optimal model structure of the passive-active storage connections is the same as the previous result (model f) while the optimized parameter values and hydrological components have some differences (see Table S2~S5 in replies to the

reviewer



Figure S1. Conceptualized structure for the coupled flow-isotope model for hillslope and depression unit connection. The light blue shades indicate active storage, the dark blue shades indicate passive storage.

In reply to the reviewer 1, we have updated the references. Additionally, we summarized the previous studies that account for passive storages in hydrological models using at least one isotopic tracer (Table S1). It shows that number and location of passive storages are dependent on the model structure and the divided geographical units. Generally, the number of passive storages increases with the divided storages and geographical units. Therefore, for the complex karst flow system in the cockpit karst landscapes, the previous model structures with one passive storage (Zhang et al., 2019; Chang et al., 2020) may be insufficient to simulate the function of chemical mixing between active and passive storages. The optimized results from our generalized model structure incorporating all possible passive storages can make up for the deficiency.

Table S1. Summary of the previous studies that account for passive storages in hydrological models using at least one isotopic tracer

		Number				
Scale	Model	of passive	Location of passive storages	Tracer	Function	References
		storages				

	Models with					Domes and Donall	
25 ha	fast and slow	1	One storage	$^{2}\mathrm{H}$	А		
	flow reservoirs					1990	
	Chemical-						
2.5 km^2	mixing	r	Shallow and deep	Chlorida	A and P	Page at $a1 - 2007$	
5.5 KIII	dynamic	2	storages	Chioride	A allu D	Page et al., 2007	
	TOPMODEL						
23.6	The multiple	2	C = 11 = t = m = = =	211	٨	Sam an al 2007	
km ²	bucket model	3	Soll storage	-H	А	Son er al., 2007	
2.0.1.	The	2	Upper and lower	211	٨	E-mining at al. 2009	
5.8 na	SoftModel _i	Z	hillslope storages	п	А	Femicia et al., 2008	
	Complete						
2.0.1.	mixing	1	0	211	D	E-mining et al. 2010	
3.8 ha	and partial-	1	One storage	-Ή	В	Fenicia et al., 2010	
	mixing model						
			2 for upper and low				
0.2			storages in upper				
2.5 and	Lunan-CIM	25	catchment, and 3	211	A	Birkel et al., 2011a	
122	(L-CIM)	2~5	for upper, low and	-H			
Km-			deep storages in				
			lower catchment				
3.6 and			The total of				
30.4	SAM ^{dyn} model	1	The total of	18 O	С	Birkel et al., 2011b	
km ²			catchment storages				
740	The tracer		Shallow and deep	² LI			
149	aided model	4	storages for uplands	П, allvalinity	А	Capell et al., 2012	
KIII	alded model		and lowlands	arkannity			
118	The		Unsaturated zona				
1.4, 0	DYNAMIT	2	and slow flow	Chlorida	A and D	Hrachowitz et al.,	
1 m ²	(DYNAmic	2		Chionde	A allu D	2013	
KIII	MIxing Tank)		reservoir				
	Tracer-aided						
	hydrological		Three store res	180	B and C		
201^{2}	model for a	3	(upper lower and			Dirizol et al. 2015	
JU KIII	wet Scottish	5	(upper, lower and	0		Dirker et al., 2015	
	upland		saturation areas)				
	catchment						
	Hydrochemical		Shallow and				
3.7 km ²	model of	2	groundwater	Chloride	A and B	Benettin et al., 2015	
	Upper Hafren		storage				
	The landscape-		Three storages				
$3.2 \mathrm{km}^2$	hased dynamic	3	(hillslope,	² н	R	Soulsby et al 2015	
3.2 XIII	model	с <u>э</u>	groundwater, and	11	Б	500150y et al., 2015	
	model		saturation area)				

3.2 km ²	STARR (Spatially Distributed Tracer-Aided Rainfall-	2	Soil and groundwater storage	² H	A, B and C	van et al., 2016
3.2, 0.6 and 0.5 km ²	Runoff model) STARR (Spatially Distributed Tracer-Aided Rainfall- Runoff model)	1	Soil storage	¹⁸ O	A and B	Ala-Aho et al., 2017
3.2 km ²	STARR model for the humid	2	Soil and groundwater	² H	A and C	Dehaspe et al., 2018
10.2 ha	A conceptual catchment model	2	Shallow and groundwater storage	¹⁸ O	A, B and C	Rodriguez., 2018
1.25 km ^{2*}	Tracer-aided hydrological model for karst	1	Hillslope storage	² H	A, B and C	Zhang et al., 2019
7.8 km ²	STARR (Spatially Distributed Tracer-Aided Rainfall- Punoff model)	2	Soil and groundwater storage	² H	A, B and C	Piovano et al., 2019
Spring*	Lumped Model for karst	1	Fast flow reservoir	EC	A	Chang et al., 2020
0.23, 0.5, 0.6, 3.2 and 7.8 km ²	A spatially distributed tracer-aided hydrological model (STARR)	1	Soil storage	² H and ¹⁸ O	A, B and C	Piovano et al., 2020
1.44 km ²	The EcH ₂ O-iso Model	1	The extra groundwater storage	² H and ¹⁸ O	A, B and C	Yang et al., 2021

Note: A represents that passive storage can help reproduce the main isotope dynamics and improve simulation accuracy; B represents that passive storage can help track flux, resident or transit time; C represents that passive storage can help estimate catchment storage. *refers to karst catchment.

We will redraw the figures in high quality and add the missing references.

References in Table S1:

- Ala-Aho, P., Tetzlaff, D., Mcnamara, J.P., Laudon, H., Soulsby, C.: Using isotopes to constrain water flux and age estimates in snow-influenced catchments using the STARR (Spatially distributed Tracer-Aided Rainfall-Runoff) model, Hydrol. Earth Syst. Sci., 21, 5089-5110, https://doi.org/10.5194/hess-21-5089-2017, 2017.
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- Benettin, P., J. W. Kirchner, A. Rinaldo, G. Botter.: Modeling chloride transport using travel time distributions at Plynlimon, Wales, Water Resour. Res., 51, 3259-3276, https://doi.org/10.1002/2014WR016600, 2015.
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- Birkel, C., Soulsby, C., Tetzlaff, D.: Conceptual modelling to assess how the interplay of hydrological connectivity, catchment storage and tracer dynamics controls nonstationary water age estimates, Hydrol. Process., 29, 2956-2969, https://doi.org/10.1002/hyp.10414, 2015.
- Capell, R., Tetzlaff, D., Soulsby, C.: Can time domain and source area tracers reduce uncertainty in rainfall-runoff models in larger heterogeneous catchments? Water Resour. Res., 48, W09544, https://doi.org/10.1029/2011wr011543, 2012.
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- Dehaspe, J., Birkel, C., Tetzlaff, D., Sánchez-Murillo, R., Dur á Quesada, A.M., Soulsby, C.: Spatially-distributed tracer-aided modelling to explore water and isotope transport, storage and mixing in a pristine, humid tropical catchment, Hydrol. Process., 32, 3206-3224, https://doi.org/10.1002/hyp.13258, 2018.
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- Hrachowitz, M.; Savenije, H.; Bogaard, T. A.; Tetzlaff, D.; Soulsby, C.: What can flux tracking teach us about water age distribution patterns and their temporal dynamics?, Hydrol. Earth Syst. Sci., 17, 533-564, https://doi.org/10.5194/hess-17-533-2013, 2013.

- Page, T., Beven, K. J., Freer, J., Neal, C.: Modelling the chloride signal at Plynlimon, Wales, using a modified dynamic TOPMODEL incorporating conservative chemical mixing (with uncertainty), Hydrol. Process., 21, 292-307, https://doi.org/10.1002/hyp.6186, 2007.
- Piovano, T. I., Tetzlaff, D., Carey, S. K., Shatilla, N. J., Smith, A., Soulsby, C.: Spatially distributed tracer-aided runoff modelling and dynamics of storage and water ages in a permafrost-influenced catchment, Hydrol. Earth Syst. Sci., 23, 2507-2523, https://doi.org/10.5194/hess-23-2507-2019, 2019.
- Piovano, T. I., Tetzlaff, D., Maneta, M., Buttle, J. M., Carey, S. K., Laudon, H., McNamarah, J., Soulsby, C.: Contrasting storage-flux-age interactions revealed by catchment inter-comparison using a tracer-aided runoff model, J. Hydrol., 590, https://doi.org/10.1016/j.jhydrol.2020.125226, 2020.
- Rodriguez, N. B., McGuire, K. J., Klaus, J.: Time-varying storage-Water age relationships in a catchment with a Mediterranean climate, Water Resour. Res., 54, https://doi.org/10.1029/2017WR021964, 2018.
- Son, K., Sivapalan, M.: Improving model structure and reducing parameter uncertainty in conceptual water balance models through the use of auxiliary data, Water Resour. Res., 43, W01415, https://doi.org/10.1029/2006wr005032, 2007.
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- Zhang, Z., Chen, X., Cheng, Q., Soulsby, C.: Storage dynamics, hydrological connectivity and flux ages in a karst catchment: conceptual modelling using stable isotopes, Hydrol. Earth Syst. Sci., 23, 51-71, https://doi.org/10.5194/hess-23-51-2019, 2019.

(1) This article raises the problem of how to improve the knowledge of the functioning of karst aquifers by combining field data and a numerical model that wants to consider all flows reflecting different modes of transfer.

This study relies on numerous oxygen-18 isotopic data to better constrain the different volumes of water present in karst systems.

This study thus proposes an interesting approach but remains very local and does not propose interesting perspectives to other contexts. Reply: Please see the above explanations.

(2) The figures are not of good quality and are often too small for the information to be used quickly.

Reply:

We will deliver the improved figures with high quality and clear information in the revised manuscript.

(3) The bibliography lacks recent references and sometimes is not appropriate to support an argument. The introduction really needs to be improved by referring to more recent and relevant work.

Reply:

We will revise the introduction to focus on hydrological connections of hillslope depression fast/slow flow in cockpit karst landscapes, and functions of passive storages incorporated into the total storage, particularly in karst flow systems, as summarized in Table S1. We have added associated publications in the most recent 10 years as follows:

The residence time:

- Brki, Z., Kuhta, M., Hunjak T.: Groundwater flow mechanism in the well-developed karst aquifer system in the western Croatia: Insights from spring discharge and water isotopes, CATENA., 161,14-26, https://doi.org/10.1016/j.catena.2017.10.011, 2018.
- Zhang, Z., Chen, X., Cheng, Q., Soulsby, C.: Characterizing the variability of transit time distributions and young water fractions in karst catchments using flux tracking, Hydrol. Process., 34, 15, https://doi.org/10.1002/hyp.13829, 2020b.

Modeling in karst:

- Dubois, E., Doummar, J., Pistre, S., Larocque, M.: Calibration of a lumped karst system model and application to the Qachqouch karst spring (Lebanon) under climate change conditions, Hydrol. Earth Syst. Sci., 24, 4275-4290, https://doi.org/10.5194/hess-24-4275-2020, 2020.
- Husic, A., Fox, J., Adams, E., Ford, W., Agouridis, C., Currens, J., Backus, J.: Nitrate Pathways, processes, and timing in an agricultural karst system: Development and application of a numerical model, Water Resour. Res., 55, 2079-2103, https://doi.org/10.1029/2018wr02370, 2019.
- Xu, C., Xu, X., Liu, M., Li, Z., Zhang, Y., Zhu, J., Wang, K., Chen, X., Zhang, Z., Peng, T.: An improved optimization scheme for representing hillslopes and depressions in karst hydrology, Water Resour. Res., 56, e2019WR026038, https://doi.org/10.1029/2019WR026038, 2020.
- Ollivier, C., Mazzilli, N., Olioso, A., Chalikakis, K., Carrière, S.D., Danquigny, C., Emblanch, C.: Karst recharge-discharge semi distributed model to assess spatial variability of flows, Sci. Total Environ., 703, 134368, https://doi.org/10.1016/j.scitotenv.2019.134368, 2020.
- Wunsch, A., Liesch, T., Cinkus, G., Ravbar, N., Chen, Z., Mazzilli, N., Jourde, H., and Goldscheider, N.: Karst spring discharge modeling based on deep learning using

spatially distributed input data, Hydrol. Earth Syst. Sci., 26, 2405-2430, https://doi.org/10.5194/hess-26-2405-2022, 2022.

Jeannin, P.Y., Artigue, G., Butscher, C., Chang, Y., Charlier, J.B., Duran, L., Gill, L., Hartmann, A., Johannet, A., Jourde, H., Kavousi, A., Liesch, T., Liu, Y., Lüthi, M., Malard, A., Mazzilli, N., Pardo-Igúzquiza, E., Thi éry, D., Reimann, T., Schuler, P., W öhling, T., Wunsch, A.: Karst modelling challenge 1: Results of hydrological modelling, J. Hydrol. 600, 126508, https://doi.org/10.1016/j.jhydrol.2021.126508, 2021.

Hydraulics in karst:

- Ding, H., Zhang, X., Chu, X., Wu, Q.: Simulation of groundwater dynamic response to hydrological factors in karst aquifer system, J. Hydrol., 587, 124995, https://doi.org/10.1016/j.jhydrol.2020.124995, 2020.
- Huang, W., Deng, C.B., Day, M.J.: Differentiating tower karst (fenglin) and cockpit karst (fengcong) using DEM contour, slope, and centroid, Environ. Earth Sci., 72, 407-416, https://doi.org/10.1007/s12665-013-2961-3, 2014.
- Jourde, H., Massei, N., Mazzilli, N., Binet, S., Batiot-Guilhe, C., Labat, D., Steinmann, M., Bailly-Comte, V., Seidel, J. L., Arfib, B., Charlier, J. B., Guinot, V., Jardani, A., Fournier, M., Aliouache, M., Babic, M., Bertrand, C., Brunet, P., Boyer, J. F., Bricquet, J. P., Camboulive, T., Carrière, S. D., Celle- Jeanton, H., Chalikakis, K., Chen, N., Cholet, C., Clauzon, V., Soglio, L. D., Danquigny, C., Défargue, C., Denimal, S., Emblanch, C., Hernandez, F., Gillon, M., Gutierrez, A., Sanchez, L. H., Hery, M., Houillon, N., Johannet, A., Jouves, J., Jozja, N., Ladouche, B., Leonardi, V., Lorette, G., Loup, C., Marchand, P., de Montety, V., Muller, R., Ollivier, C., Sivelle, V., Lastennet, R., Lecoq, N., Mar échal, J. C., Perotin, L., Perrin, J., Petre, M. A., Peyraube, N., Pistre, S., Plagnes, V., Probst, A., Probst, J. L., Simler, R., Stefani, V., Valdes-Lao, D., Viseur, S., Wang, X.: SNO KARST: A French Network of Observatories for the Multidisciplinary Study of Critical Zone Processes in Karst Watersheds and Aquifers, Vadose Zone J., 17, 180094, https://doi.org/10.2136/vzj2018.04.0094, 2018.
- Zhang, R., Chen, X., Zhang, Z., Soulsby, C.: Using hysteretic behavior and hydrograph classification to identify hydrological function across the "hillslope-depressionstream" continuum in a karst catchment, Hydrol. Process., 34, 3464-3480, https://doi.org/10.1002/hyp.13793, 2020a.

Mixing processes in karst:

- Dar, F., Jeelani, G., Perrin, J, Ahmed, S.: Groundwater recharge in semi-arid karst context using chloride and stable water Isotopes, Groundwater Sustain. Dev., 14, 100634, https://doi.org/10.1016/j.gsd.2021.100634, 2021.
- Lorette, G., Viennet, D., Labat, D., Massei, N., Fournier, M., Sebilo, M., Grancon, P.: Mixing processes of autogenic and allogenic waters in a large karst aquifer on the edge of a sedimentary basin (Causses du Quercy, France), J. Hydrol., 593, 125859, https://doi.org/10.1016/j.jhydrol.2020.125859, 2021.
- Mayer-Anhalt, L., Birkel, C., Sánchez-Murillo, R., Schulz, S.: Tracer-aided modelling reveals quick runoff generation and young streamflow ages in a tropical rainforest catchment, Hydrol. Process., 36, e14508, https://doi.org/10.1002/hyp.14508, 2022.

(4) For example, citing the 2003 paper by Batiot et al. to refer to the fact that oxygen isotopes can provide information on water residence times is a misuse of this work since in this paper Batiot et al. use TOC and Mg as a tracer of fast transit times versus long residence times. There are no references to isotopes in this paper. Again, the citations should be reviewed as there is recent work on the use of isotopes to improve knowledge of karst systems.

Reply:

We will delete this reference (Batiot et al., 2003) and cite the latest references about the use of isotopes to improve knowledge of karst systems as listed above (e.g., Brki et al., 2018; Zhang et al., 2020b).

(5) In line 62, the authors refer to work from 2010 and 2013 as the state of the art of models at different scales of study that have been developed to describe flows in karst. There is recent work on tracing-modelling coupling by the Montpellier team that could have been used to support the authors' argument. Reply:

We have updated the references and added more recent works on hydrological modelling such as Jourde et al.(2018), Dubois et al.(2020), Jeannin et al.(2021), and Wunsch et al.(2022) from recent works by the Montpellier team.

(6) Finally, to end these comments on bibliographic references, the work of Rodriguez et al. (2017) is cited on line 127 but the reference does not appear in the bibliographic list.

Reply:

We will add this reference as shown below:

Rodriguez, N. B., McGuire, K. J., Klaus, J.: Time-varying storage-Water age relationships in a catchment with a Mediterranean climate, Water Resour. Res., 54, https://doi.org/10.1029/2017WR021964, 2018.

(7)

On the background of the article Introduction

In my opinion, the introduction is a bit confusing and would benefit from being reworked and clarified especially in the justification section of the study. The authors go directly from the general idea to the application on their site without explaining why their site will allow them to answer their problem if only because there are isotopic and hydrological data (which ones).

<u>Reply:</u>

We will revise the introduction. Our selected catchment of Chenqi is a karst experimental catchment focused on investigations of hydrological, ecological and geological (carbonate dissolution) changes under climate change and human activities. So there are detailed observational data and field investigations in this catchment. The flow discharge was observed at intervals of 15 min, and water was sampled for isotope analysis at intervals of daily (dry season) and hourly (wet season). As we know, there are seldom detailed observations of isotope signatures. The previous coupled models of hydrological and isotopic processes (listed in Table S1) are mostly calibrated and validated against daily and weekly isotope signatures. In karst catchment, as flow discharge and isotope concentration vary dramatically fast, the coarse resolution data used in this study offers an opportunity to optimize our new model structure, such as hydrological connections of hillslope-depression fast/slow flow, and the functioning of passive storages in the karst flow system.

(8) Page 88; can the authors clarify this concept "Hence, the storage...." How do they account for the seasonality of water isotopic levels and their notion of storage? <u>Reply:</u>

Here the storage volume refers to the total storage (active storage and passive storage) for the isotope mixing (see Fig. S1). Passive storage does not directly contribute to streamflow, but it participates in stable isotope simulation (Hrachowitz et al., 2013). As shown in Eqs. $(7)\sim(9)$ in the original manuscript, the passive storage added in the total storage takes a function of the isotope mixing and transport between active storage and passive storage and thereby can reduce the seasonality of isotopic composition in stream water.

(9) On the study site part

This paragraph should also be reworked, especially figure 1 which is unclear. It is difficult to distinguish the sources on the figure.

I would have liked to have a more complete description of their karstic system. <u>Reply:</u>

We have redrawn Fig. 1 as shown below (Fig. S2). There is a main underground channel in the depression with an ascending spring at the catchment outlet, and high flows can spill over the bottom of the depression ditches (referring to the surface stream in Fig. S2). So, in Fig. S2, the two points at the outlet refer to the observation sites of underground channel and surface stream at the catchment outlet. The discharge used for simulations is the total of underground channel and surface stream discharge.

Two hillslope springs can be observed in the study catchment (see Fig. 1 in Zhang et al. (2013)). We selected a perennial spring at the hillslope foot in this study. The location has been added in the figure (see Fig. S2). Water samples at two depression wells (W1 and W4 in Fig. S2) are analyzed, and the isotope compositions of W1 and W4 in comparison to those at the hillslope spring and the outlet discharge are used to indicate flow connections between hillslope and depression units.



Figure S2. The location of Chenqi catchment (a), stratigraphic profile (b), topography (c), photo (d), and observations at surface stream outlet (e), underground channel outlet (f) and hillslope spring(g).

Reference

Zhang, Z., Chen, X., Chen, X., Shi, P.: Quantifying time lag of epikarst-spring hydrograph response to rainfall using correlation and spectral analyses, Hydrogeol. J., 21, 1619-1631, https://doi.org/10.1007/s10040-013-1041-9, 2013.

(10) On the study site part

Where are located the two epikarst springs mentioned in line 168? Are they the two pink triangles?

Reply:

See the above reply and Fig. S2. Since the springs are formed by the shallow permeability zones (fractures and conduits) overlying the impervious bedrock (marlite), the hillslope springs are also called epikarst springs in the previous study (Zhang et al. (2013)).

(11) Where is the main outlet of this system located, are there any isotopic and hydrological data? I asked myself this question while reading the description of the hydrological response of epikarst springs to precipitation. It is difficult to say that the behaviour of epikarst springs reflects the behaviour of the karst system itself.

Reply:

Please see the explanations above. Discharge at the catchment outlet and hillslope springs was measured by v-notch weirs with a time interval of 15 min. Hillslope springs, catchment outlet flows, and rainfall were regularly sampled at daily intervals. They were intensively sampled during the wet season (May-August) using an autosampler set at an hourly interval (see lines 197-184 in the original manuscript).

Here, epikarst springs refer to hillslope springs, and the discharge and isotope dynamics are used to indicate shallow (fast) flow behavior in the hillslope unit. For the whole catchment, the discharge and isotope dynamics at the catchment outlet can reflect the behavior of the karst system.

We will revise this portion and describe the associated contents more clearly.

(12) This raises the question of what the authors want to identify in their article, is it to work on flows in the epikarst or in the karst? In which case the problematic of the introduction must be reoriented and the bibliography better targeted. <u>Reply:</u>

As shown in Fig. 4 in the original manuscript, the profile in each unit was vertically separated into an unsaturated zone in the upper soil and epikarst layers and a saturated zone representing the deep aquifer. The saturated flow can be produced in the epikarst and deep saturated zone. In our model, we merged flows in the two layers and conceptualized them into fast flow and slow flow reservoirs. A large portion of the shallow flow (or epikarst flow) together with deep conduit flow in hillslope unit is categorized into fast flow reservoir (see over 70% of the fast flow in the hillslope unit shown in Table 6 in the original manuscript).

We will revise the introduction to clarify hydrological behaviors and connections in the cockpit karst landscapes.

(13) In the "Obervationnal dataset" section, it would have been nice to structure this paragraph better between data collection and isotopes analysis

The first part of this paragraph concerns data acquisition

Were the samples collected in the automatic samplers analysed quickly to avoid evaporation problems?

Can you provide details on how the groundwater was collected?

Is it possible to have a little more detail on the dates of sampling? Which samples were taken at the same time, what is the time lag between rainwater and groundwater?

Reply:

We will revise this portion description according to your suggestions.

The hillslope springs, the catchment outlet flows, and rainfall were sampled using an autosampler set. The sampled water was sealed by using plastic bags to avoid evaporation (see Fig. S3). Water samples were taken to our laboratory every day and stored at about 4 ∞ .

The depression groundwater at two wells was manually sampled. The sampling was taken two times before and after the four rainfall events from 6 July 2017 to 20 August 2017.

We have listed the sampling time in the study period in Table S2.

Fable S2. Statistical characteristics of isotope data for rainfall, hillslope spring,
catchment outlet discharge and depression groundwater in the study period (note: water
sampled at an hourly intervals from 12 June 2017 to 20 August 2017, and at daily
nterval in other times)

Obs	Sampling	Numborg	δD (‰)			$\delta^{18} { m O}$ (‰)		
Obs	time	Numbers	Range	Mean	CV	Range	Mean	CV
	Oct. 8	253	-120.2~29	-64.9	0.49		-9.1	0.42
Deinfall	2016 to					166.10		
Kainiali	June 12					-10.0~1.0		
	2018							
	Oct. 8							
Catchment	2016 to	1096	-76.8~-39.3	-60.6	0.07	-11~-4.1	-8.6	0.09
outlet discharge	June 12							
	2018							
	Oct. 8	1095	-77~-37.8	-63.7	0.05	-10.8~-5.9	-9.2	0.06
Uillalana anring	2016 to							
Hinsiope spring	June 12							
	2018							
Crowndwyston	July 6~							
W1	Aug. 20,	175	-65.7~-50.7	-60.8	0.03	-9.6~-6.3	-8.7	0.05
W I	2017							
Crowndwyston	July		-70.2~-55	-62.5	0.07	-10.1~-7.9	-8.9	0.07
W/4	6~Aug.	47						
W4	20, 2017							



Figure S3. The automatic sampling instruments we designed.

(14) The second part of this paragraph concerns the analysis of isotopic data. Figure 2 really needs to be taken back because it is unreadable. I can't follow their reasoning based on this figure.

What is the significance of some correlations that have coefficients at 0.21? <u>Reply:</u>

We have redrawn Fig. 2 (see Fig. S4).

The figure shows that (1) all the hillslope and depression flows undergo evaporative effect as their isotopes are more enrichment than those of precipitation; (2) the catchment outlet flow is primarily contributed to the hillslope flow as the fitted isotope lines of the two flows are close; (3) fast and slow flows at the depression unit are strong variable in space. The depression groundwater at W1 and the catchment outlet flow is more enriched compared to that at W4. As W4 is located at the hillslope foot, and groundwater there receives more new water (fast flow) from the hillslope spring and rainfall. W1 is located in a locally confined aquifer surrounded by rocks with poor permeability, and the flow seldom mixes with new water (rainfall) (Chen et al., 2018).

The correlation between δ^{18} O and δ D at W1 is 0.21, and tested to be significant at the significance level of *p*<0.001.



Figure S4. Plot of δ^{18} O- δ D for rainwater, catchment outlet discharge, hillslope spring and depression groundwater at wells W1 and W4.

Chen, X., Zhang, Z. C., Soulsby, C., Cheng, Q. B., Binley, A., Jiang, R., Tao, M.: Characterizing the heterogeneity of karst critical zone and its hydrological function: an integrated approach, Hydrol. Process., 32, 1-15, https://doi.org/10.1002/hyp.13232, 2018.

(15) Where are the sources of the hillslope?

Line 216 "this phenomenon....recharge" is this really surprising? do we need so much isotopic analysis to reach this conclusion? What do the authors want to demonstrate? Or rather, what do they bring that is new? Reply:

The hillslope discharge and δD values come from the observations and water sampling at the hillslope spring (see Figs. S2 and S3). We used the daily δD and lc-excess values to draw the box plot of their monthly variations.

As shown in Fig. S5, the similar seasonality pattern between hillslope spring and catchment outlet discharge proves that hydrological variability (e.g., evaporation and fresh water recharge) at the catchment outlet is primarily controlled by the hillslope hydrological processes. The difference of the monthly mean δD and lc-excess values between hillslope spring and catchment outlet discharge demonstrates that flow composition (i.e., fast and slow flows) has been regulated by depression unit when

hillslope flow mixes with depression flow. The more enriched δD and less lc-excess at the catchment outlet indicate a stronger mixture of hillslope fresh flow with depression old flow (indicating the HF-DS connection in Fig. S1), while the evaporation effect on depression groundwater flow is relatively weaker due to thick soils in the depression unit.



Figure S5. Monthly observed δD and lc-excess of outlet discharge and hillslope spring during the study period.

(16) I think that this paragraph really needs to be reworked by providing information on the geometry of their system, to make figure 2 readable, and to explain the variability of the results of each analysis point. This figure brings more confusion than help in the argumentation.

Reply:

This figure has been redrawn as shown in Fig. S4. We will revise the descriptions as shown in our reply to question (14).

(17) It would also be necessary to specify the precautions of the mode of sampling especially for the analysis of isotopes. Finally, it would be necessary to have a

temporal idea of the samples at each sampling site. This could help in the analysis of the results.

Reply:

Please see our reply to question (11) and Table S2.

(18) Finally, how can we consider a flow model, a tracer that is not conservative? Doesn't this call into question their initial hypothesis concerning the fact of using a tracer to identify stored water volumes

Reply:

The stable isotopes (δ^{18} O and δ^{2} H) belong to the conservative tracer when their isotopic fractionations are taken into account in our developed model. So, the spatial and temporal data offer ideal information to trace flow dynamics (e.g., the residence time, storage, flux, and age).

(19) Model development part

I am well aware that one has to start from hypothesis to build a conceptual model that helps to lay the foundations of a numerical model, but I am not sure that considering the epikarst as an analog of a karst system is really relevant. A better justification than the one given is really needed. The calibration of the model with a tracer which is supposed to be conservative, and which is not, given the evaporation curves. Even if the results between calibration and validation are satisfactory, it is the very design of the model that is problematic.

Reply:

We agree that the saturated flows can be further divided into the shallow aquifer (epikarst) and deep aquifer (conduit). Since the epikarst flow in response to rainfall is generated locally and intermittently, most of the epikarst flow recharges into deep aquifer. In this study, the flows in these two aquifers are merged into a flow system consisting of fast and slow flows (the dual flow system).

The stable isotope is a conservative tracer, but its concentration is affected by evaporation fractionation. The evaporation fractionation (see parameters of ls_h and ls_d) has been added to the model calibration and validation (please see our reply to question (9) for the reviewer 1).

We will revise the relevant descriptions to be more clear.

(20) Where do the hydrographs in figure 5 come from? This was not mentioned in the data section. Or how was it measured?

Reply:

Does this question refer to Fig. 6? If yes, the hydrographs in Fig. 6 show a comparison between the daily simulated and observed discharge in calibration and validation periods. The simulated discharge comes from the 30 sets of optimal solutions by model *f*. We will correct the figure explanations.

(21) Is taking into account a certain number of passive storages until arriving at a satisfactory modelling result representative of reality? Reply:

Yes, we set fourteen schemes (scenarios) that incorporate 0~4 passive storages into different positions within the karst flow system, i.e., fast and/or slow flow reservoirs in combination with the hillslope and/or depression units (see lines 343~347 and Table 3 in the original manuscript). The optimized model (model *f*) captures the sharp rise and decline of high flow and isotopic variations. As listed in Table S4 (see reply to the reviewer 1), the simulation results of model *f* suggest that the proportion of the total subsurface flow (slow flow and fast flow at underground channel) is 58%, and surface flow from the surface channel is 42% of the total catchment flow. These proportions are consistent with 55% and 45% from the observations at the underground conduit outlet and surface channel outlet, respectively.

(22) The conclusion also needs to be reviewed and above all, what prospects are there for extending this study to other cases? It would have been nice to analyze the relevance of the conceptual model (epikarst as an analog of a karst) to give some weight to their study and try to bring some opening elements. Reply:

We will revise the discussion and conclusion according to your comments and suggestions. In particular, we will discuss the connections between hillslope flow and depression fast/slow flow (indicated by a weight r_{hd} , see replies to the reviewer 1) and the functioning of epikart flow to the karst flow (indicated by a weight *ks*, see lines 278~283 in the original manuscript). We will compare our modeling results with other associated results in karst areas.

(23) It remains a very local study, with results that seem coherent, but on what assumptions?

Reply:

Please refer to our reply at the beginning. We will thoroughly revise the manuscript according to your comments and suggestions. An assumption in this study is that more passive storages are needed to improve flow and isotope simulations since there are different flow components and connections in two geographical units of the cockpit karst landscapes.