## 1 The value of water isotope data on improving process

## 2 understanding in a glacierized catchment on the Tibetan

## 3 Plateau

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#### 13 Abstract

14 This study integrated a water isotope module into the hydrological model THREW which has been 15 successfully used in high and cold regions. Signatures of oxygen stable isotope (<sup>18</sup>O) of different water 16 inputs and stores were simulated coupling with the simulations of runoff generations. Isotope 17 measurements of precipitation water samples and assumed constant isotope signature of ice meltwater 18 were used to force the isotope module. Isotope signatures of water stores such as snowpack and 19 subsurface water were updated by an assumed completely mixing procedure. Fractionation effects of 20 snowmelt and evapotranspiration were modeled in a Rayleigh fractionation approach. The isotope-aided 21 model was subsequently applied for the quantifications of runoff components and estimations of mean 22 water travel time (MTT) and mean residence time (MRT) in the glacierized watershed of Karuxung River 23 on the Tibetan Plateau. Model parameters were calibrated by three variants with different combinations 24 of streamflow, snow cover area and isotopic composition of stream water. Modeled MTT and MRT was 25 validated by estimate of a tracer-based sine-wave method. Results indicate that: (1) the proposed model 26 performs well on simultaneously reproducing the observations of streamflow, snow cover area, and 27 isotopic composition of stream water, despite that only precipitation water samples were available for 28 tracer input; (2) isotope data facilitate more robust estimations on contributions of runoff components 29 (CRCs) to streamflow in the melting season, as well as on MTT and MRT; (3) involving isotope data for 30 the model calibration obviously reduces uncertainties of the quantification of CRCs and estimations of 31 MTT and MRT, through better constraining the competitions among different runoff processes induced 32 by meltwater and rainfall. Our results inform high value of water isotope data on improving process 33 understanding in a glacierized basin on the Tibetan Plateau.

#### 34 Keywords:

Tracer-aided hydrological model; Contribution of runoff components; Water travel time; Glaciered
 catchment; Tibetan Plateau

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#### 39 1. Introduction

40 Glacierized catchments in mountainous regions are generally headwater catchments, which are of 41 great interest because of its complex runoff generation processes and important role on supplying water 42 sources for downstream regions (Immerzeel et al., 2010). Stable isotopes in water ( $\delta^2$ H and  $\delta^{18}$ O) are 43 powerful tools for investigating the water cycle and hydrological processes (Gat, 1996; Bowen et al., 44 2019). Isotopic composition of water changes with multiple ecological and hydrological processes, and 45 is affected by several environmental factors (Zhao et al., 2012; Wang et al., 2013), thus is frequently used 46 to track the storage and transportation of water. Isotopic compositions generally distinguish among 47 different water bodies and phases (Xi, 2014), thus is widely used to determine the relative dominance of 48 water sources, especially in the glacierized catchments (Kong et al., 2019; He et al., 2020). Water isotope 49 data consequently bears the potential on improving the understanding of hydrological processes in 50 glacierized catchments.

51 The Tibetan Plateau as a high mountainous cryosphere is the source of many major rivers in Asia 52 including Yarlung Tsangpo-Brahmaputra River, Ganges River, Indus River and so on (Report of STEP, 53 2018). Scientific understanding of hydrological processes in this region is critical in predicting the 54 responses of water resources and water hazards to climate changes (Lutz et al., 2014; Immerzeel et al., 55 2010; Miller et al., 2012). River runoff in these basins is prominently fed by multiple water sources 56 including snowmelt, glacier melt and rainfall (Zongxing et al., 2019). Coupling with the strong spatio-57 temporal variabilities of meteorological inputs, the complicated runoff generation processes imply big 58 challenges in understanding the hydrological behaviors in glacierized basins on the Tibetan Plateau.

59 It is, therefore, of critical importance to quantify contributions of runoff components (CRCs) to 60 streamflow in glacierized regions. Estimating CRCs by hydrological models is one of the commonly 61 adopted method (Weiler et al. 2018), which is particularly subject to the following challenges. First, 62 modeled CRCs rely heavily on the model conceptualizations of the mixing and propagations of different 63 water sources in the basin. Model configurations and corresponding parameters representing the storage 64 capacities of soil layers and groundwater aquifers obviously affect the relative proportions of surface and 65 subsurface flow to streamflow (Nepal et al. 2014). CRCs modeled by different hydrological models are 66 thus rarely comparable (Tian et al. 2020). For example, Nepal et al. (2015) and Siderius et al. (2013) 67 compared CRCs estimated by different glacio-hydrological models in glacierized basins in the Himalayan region, and demonstrated considerable variations of the modeled CRCs. They attributed the 68 69 difference to the variations of the model conceptualizations. Second, strong compensatory effects of the 70 simulated runoff induced by precipitation and ice meltwater which were typically not well constrained 71 in the model resulted in large variations of the modeled CRCs. For instance, modeling results from 72 Duethmann et al. (2015) and Finger et al. (2015) indicated that overestimated precipitation-triggered 73 runoff in the model can be easily compensated by an underestimated ice melt runoff and vice versa, 74 especially in high altitude glacierized basins where precipitation input have large uncertainty.

Tracer data of water stable isotope have been widely used to label runoff components in the popular end-member mixing approach (e. g., Kong et al. 2011; He et al. 2020). Its value for improving modeled CRCs, however, have not been sufficiently investigated. Previous applications of tracer-aided hydrological models which integrated the simulation of water isotopic compositions of different runoff components into the rainfall/melting-runoff processes in snow dominated basins have demonstrated high values of water isotope data on diagnostically improving model structure and recognizing the dominances of runoff processes on streamflow (Capell et al., 2012; Delavau et al. 2017; Son and Sivapalan, 2007; 82 Birkel et al., 2011; Stadnyk and Holmes, 2020). An early test of the isotope-aided hydrological model in 83 a glacierized basin in Tianshan Central Asia of He et al. (2019) indicated that additionally use of isotope 84 data helped to constrain the internal apportionments of runoff components in the model and improved 85 the estimation of CRCs at an event scale. However, exploring the values of water isotope data for 86 hydrological modelling in glacierized basins are still limited to the low availability of water tracer data 87 from field water sampling due to the harsh environment, especially for glacierized basins on the Tibetan 88 Plateau. As far as we know, glacio-hydrological model coupled with the simulations of isotope signatures 89 have not been developed and tested in the Tibetan Plateau yet.

90 Quantifying the time from entrance of water to its exits is fundamental to understandings of flow 91 pathways and the storage and mixing processes (McGurie and McDonnell, 2006). Characterizing water 92 travel time distribution (TTD) and mean travel time (MTT) in addition to the traditional focus on 93 streamflow response allows us to be closer to getting the right answers for the right reasons (Hrachowitz 94 et al. 2013). Despite that TTD and MTT serve good tools to diagnose unsuitable model structures and 95 parameterizations (McMillan et al. 2012), it has been rarely quantified in glacierized basins. Plenty of 96 convenient tools have been developed based on lumped parameter models, but their practical applications 97 in glacierized basins are restricted by the time invariant assumption and the weakness on considering the 98 strong spatio-temporal variability of runoff processes (van Huijgevoort et al. 2016) as well as the seasonal 99 water inputs from snowmelt and glacier melt. Fully physically-based water particle tracking approaches 100 coupling with hydrological processes whereas are only limited to small basins due to the heavy 101 computation cost (Remondi et al. 2018). Conceptual models that used additional tracer storage 102 compartments along with the flow and transport processes have provided crucial information on the 103 dynamics of flow pathways and storages, but rely heavily on the prior definitions of function shape (e.g., 104 travel time distribution (TTD) in van der Velde et al. 2015; StorAge Selection function (SAS) in Benettin 105 and Bertuzzo 2018; age-ranked storage-discharge relation in Harman 2019). In contrast, tracer-aided 106 hydrological models that integrated the storage and transportation of conservative water tracers into the 107 runoff generation processes have been demonstrated as successful on estimating TTD and water ages as 108 well as their time variances with in snowmelt influenced basins (e.g., Soulsby et al. 2015; Ala-Aho et 109 al.2017). However, such hydrological models have not been applied in glacierized basins for estimations 110 of TTD and MTT yet.

111 For process understanding in glacierized basins, glacio-hydrological models that additionally 112 represented the snow processes and glacier evolution have been widely used (e.g. Immerzee et al. 2013; 113 Lutz et al. 2014 and 2016; Luo et al. 2018). The more complex integration of water sources from different 114 flow pathways and units whereas resulted in expanded parameter space of these hydrological models 115 which introduced large uncertainty in the model calibration (Finger et al. 2015). Equifinality is serious 116 in these regions when calibrating hydrological model by streamflow solely, indicating that different 117 parameters and runoff component proportions could perform similarly in discharge simulation (Beven 118 and Freer, 2001; Chen et al., 2017), despite of the general good performance for streamflow simulation. 119 Therefore, multiple datasets including glacier observation and remotely sensed snow products have been 120 frequently used in addition to streamflow measurements in vast glacio-hydrological simulations (e.g., 121 Parajka and Blöschl, 2008; Konz and Seibert, 2010; Schaefli and Huss 2011; Duethmann et al. 2014; 122 Finger et al., 2015; He et al. 2018). However, both discharge and snow/glacier measurements provide 123 insufficient constraints on distributions of flow pathways and the parameterizations of subsurface water 124 storages (He et al. 2019). Although application in a glacierized basin in Central Asia of He et al. (2019) 125 indicated high utility of isotope data on constraining the complex interactions of multiple runoff 126 processes for the quantifications of CRCs, the values of water tracer such as stable isotope on reducing 127 uncertainties on the estimations of TTD and MTT in glacierized basins on Tibetan Plateau have not been 128 investigated.

129 In light of those backgrounds, this study integrated the simulation of oxygen isotope signatures into 130 a hydrological model that has been proved effective to simulate the runoff processes on the Tibetan 131 Plateau. The developed tracer-aided hydrological model was applied to the Karuxung River catchment 132 (286 km<sup>2</sup>, 4550 to 7206 m a.s.l.) on Tibetan Plateau. The objectives of this study are: (1) to test the 133 capability of the proposed tracer-aided model on simultaneously reproducing streamflow and isotope 134 signatures of stream water in the study basin where only precipitation water samples are available for 135 isotope input, (2) to evaluate the values of tracer-aided method on improving the estimation of CRCs and 136 TTD/MTT in the study basin, and (3) to assess and interpret the differences between modeled TTD/MTT 137 and estimates by a lumped parameter method.

#### 138 2. Materials and methodology

#### 139 2.1 Study area and data

140 This study focuses on the Karuxung catchment, which is located in the upper region of the Yarlung 141 Tsangpo River basin, on the northern slope of the Himalayan Mountains (Figure 1). Digital elevation 142 model (DEM) data in the study catchment with a spatial resolution of 30-m was downloaded from the 143 Geospatial Data Cloud (www.gscloud.cn). The Karuxung river originates from the Lejin Jangsan Peak 144 of the Karola Mountain at 7206 m above sea level (a.s.l.), and flows into the Yamdrok Lake at 4550m 145 a.s.l. (Zhang et al., 2006). The catchment covers an area of 286 km<sup>2</sup>. The river discharge is significantly 146 influenced by the headwater glaciers which cover an area of around 58 km<sup>2</sup> (Mi et al., 2001). This 147 catchment is dominated by a semi-arid climate. The mean annual temperature and precipitation at 148 Langkazi Weather Station were 3.4°C and 379 mm, respectively. Due to the effect of the South Asian 149 Monsoon, more than 90% of the annual precipitation falls between June and September. Precipitation 150 occurs mostly in form of snow from October to the following March at high elevations (Zhang et al., 151 2015).

152

#### [Figure 1]

153 Daily temperature and precipitation data from 1<sup>st</sup> January 2006 to 30<sup>th</sup> September 2012 were 154 collected at the Langkazi Weather Station (4432 m a.s.l.). Altitudinal distributions of temperature and 155 precipitation across the catchment were estimated by the lapse rates reported in Zhang et al. (2015). 156 Runoff were measured daily from 1st April 2006 to 31st December 2012 at the Wengguo Hydrological 157 Station at the catchment outlet. The coverages of glaciers were extracted from the Second Glacier 158 Inventory Dataset of China (Liu, 2012). The 8-day snow cover extent data from MODIS product of 159 MOD10A2 (500m×500m, Hall and Riggs, 2016) were used to denote the fluctuations of the snow cover 160 area (SCA). The 8-day Leaf Area Index (LAI) and the monthly normalized difference vegetation index 161 (NDVI) data were downloaded from MODIS product of MOD15A2H (500m×500m, Myneni et al., 2015) 162 and MOD13A3 (1km×1km, Didan, 2015). Soil hydraulic parameters were estimated based on the soil 163 properties extracted from the 1km × 1km Harmonized World Soil Database (HWSD, 164 http://www.fao.org/geonetwork/).

165 Grab samples of precipitation and stream water were collected at the Wengguo Station in 2006-166 2007 and 2010-2012, for analysis of  $\delta^{18}$ O and  $\delta^{2}$ H, and the characteristics of samples are summarized in 167Table1. In the dry seasons when precipitation water was not sampled due to small event amounts,168precipitation isotope data from monthly Regionalized Cluster-based Water Isotope Prediction (RCWIP169with a pixel size of  $10' \times 10'$ , Terzer et al., 2013) were used as proxy for model input. The effect of170elevation on the isotopic composition of precipitation was estimated using a lapse rate of -0.34%/100m171based on Liu et al. (2007). The stream water samples were collected weekly every Monday from the river

172 channel near the Wengguo Station. Isotopic composition of glacier meltwater was assumed to be constant

- 173 during the entire study period and the value reported in Gao et al. (2009) was adopted.
- 174

#### [Table 1]

#### 175 2.2 Tracer-aided hydrological model

176 The THREW (Tsinghua Representative Elementary Watershed) model was originally developed by 177 Tian et al. (2006), and has been successfully applied to a wide range of catchments (e.g., Tian et al., 2012; 178 Yang et al., 2014), including glacierized basins in the Alps, Tianshan, and the Tibet Plateau (He et al., 179 2014; 2015; Xu et al., 2019). The THREW model uses the Representative Elementary Watershed (REW) 180 method for the spatial discretization of catchment, in which the study catchment is divided into REWs 181 based on the catchment DEM, and then each of the REWs is divided into sub-zones as the basic units for 182 hydrological simulation. More details of the model set up are given in Tian et al. (2006). In this study, 183 the Karuxung catchment was divided into 41 REWs.

The snowmelt and glacier melt are differentiated according to the glacier coverage data. The meltwater in non-glacier area is defined as snowmelt, and the meltwater in glacier covered area is defined as glacier melt, which includes the meltwater of both ice and snow. The two water sources are assumed to be melt in different rates, as represented by different degree-day factors. Meltwater from snow and glacier are simulated using a temperature-index method as given in Eqs. (1) and (2):

189 
$$M_{S} = \begin{cases} DDF_{S} * (T - T_{S0}) & for T > T_{S0} \\ 0 & for T \le T_{S0} \end{cases}$$
(1)

190 
$$M_{G} = \begin{cases} DDF_{G} * (T - T_{G0}) & for \ T > T_{G0} \\ 0 & for \ T \le T_{G0} \end{cases}$$
(2)

191 where, the subscripts S and G represent snow and glacier, respectively. M is the melt amount, T is 192 temperature and  $T_0$  refers to temperature threshold above which snow/ice starts to melt. DDF is the 193 degree-day factor, representing the melt rate. Glacier meltwater ( $M_G$ ) in this study includes both ice melt 194 and snowmelt on the glacierized area.

195 The fraction of snowfall  $(P_s)$  of the total precipitation P is determined by a temperature threshold  $T_s$ 196 in Eq. 3. Snow water equivalent (SWE) of each REW is thus updated by Eq. 4. The snow cover area 197 (SCA) of the corresponding REW is determined by a SWE threshold value (SWE<sub>0</sub>): when the calculated 198 SWE is higher than SWE<sub>0</sub>, the SCA of this REW is recorded as 1, otherwise the SCA is assumed to be 0 199 (similarly to Parajka and Blöschl, 2008; Zhang et al., 2015; He et al., 2014). The SCA of the whole study 200 catchment is calculated as the ratio of the sum of the areas of snow covered REWs to the total catchment 201 area. Values of  $T_N$  and  $SWE_{\theta}$  are set based on prior knowledge from Dou et al. (2011), Marques et al. 202 (2011) and He et al. (2014):  $T_s = 2^{\circ}C$ ,  $SWE_0 = 20mm$ .

203 
$$P_s = \begin{cases} 0 & T \le T_s \\ P & T > T_s \end{cases}$$
(3)

$$\frac{dSWE}{dt} = P_s - M_s \tag{4}$$

Meltwater of ice and snow, and rainfall over the glacier area are assumed to flow directly into the channel near the glacier tongue in form of surface runoff, based on the low permeability of the glacier surface. Snowmelt in the non-glacier area is assumed to generate runoff in a similar way to rainfall (Schaefli et al., 2005). For model simplicity, the evolution of the glacier area is not simulated in the model for the short simulation period of seven years.

210 Simulation of  $\delta^{18}$ O of multiple water sources was integrated into the runoff generation processes 211 in the THREW model (hereafter abbreviated as a THREW-t model). The  $\delta^{18}$ O of water sources in each 212 of the sub-zones was assumed to be conservative, meaning that no chemical reactions occurred during 213 the mixing of water sources. We assumed that the isotopic compositions of precipitation and glacier 214 meltwater are linearly dependent on elevation, and used linear gradients reported in Liu et al. (2007) 215 to estimate the initial isotopic compositions of precipitation and glacier meltwater in individual REWs 216 (similarly to He et al. 2019). The isotopic compositions of the snowpack and subsurface water storages 217 were initialized by a "spin-up" running for three hydrological years, assuming the isotopic 218 compositions of water storages would reach steady levels after three years' running. Isotope 219 composition of event snowfall on the snowpack was assumed to be the same as that of precipitation 220 occurring in the corresponding REW.

The fractionation effects of evaporation on isotope composition of water were estimated by a
Rayleigh fractionation method in Eqs. (5) to (7) (Hindshaw et al., 2011; Wolfe et al., 2007; He et al.,
2019):

224 
$$\delta^{18}O_{x}{}' = \delta^{18}O_{x} * \frac{1 - f^{CF(\frac{1}{\alpha} - 1) + 1}}{1 - f}$$
(5)

225 
$$ln\alpha = -0.00207 + \frac{-0.4156}{T} + \frac{1137}{T^2}$$
(6)

$$f = 1 - \frac{w'_x}{w_x} \tag{7}$$

where,  $\delta^{18}O_x'$  is the isotope composition of the evaporated water,  $\delta^{18}O_x$  is the isotope composition of water before evaporation,  $\alpha$  is the Rayleigh fractionation factor, T(K) is air temperature in the corresponding catchment unit, *CF* is a correction factor, and *f* is the ratio of remaining water volume to the original water volume before evaporation.

231 A complete mixing assumption was used for the tracer signatures in each water storage. 232 Consequently,  $\delta^{18}$ O of soil water and groundwater were updated according to the following equation:

233 
$$\delta^{18}O_t = \frac{w_o \delta^{18}O_o + \sum w^i \delta^{18}O^i}{w_o + \sum w^i}$$
(8)

where,  $w_o$  and  $\delta^{18}O_o$  are the water quantity and isotopic composition of the subsurface storages at the prior step, respectively.  $w^i$  refers to the infiltration into the soil storage from water source *i*. For groundwater storage,  $w^i$  refers to the seepage from upper soil water.  $\delta^{18}O^i$  stands for the isotopic composition of input water source *i*. The isotope signature of snowpack was simulated similarly as subsurface water storages according to Eq. 8. Stream water in each of the REWs was considered as a mixture of three components including inflow from the upstream REWs, runoff generated in the current REW, and the water storage in the river channel. Consequently, the isotopic composition of stream water in each REW ( $\delta^{18}O_r$ ) was estimated based on the following conservative mixing equation:

243 
$$\delta^{18}O_r = \frac{\delta^{18}O_{r,0} * w_r + \sum \delta^{18}O_{r,up} * {}_{*I^k} + \delta^{18}O_{sur}R_{sur} + \delta^{18}O_{gw}R_{gw}}{w_r + \sum I^k + R_{sur} + R_{gw}}$$
(9)

where,  $\delta^{18}O_{r0}$  is the isotopic composition of stream water and  $w_r$  is the water storage in the river channel at the time step before the mixing of runoff components.  $\delta^{18}O_{r,up}^{\ \ k}$  is the isotopic composition of stream water coming from the upstream REW k, and  $l^k$  is the inflow from the corresponding upstream REW. Subscripts of *sur* and *gw* refer to the surface runoff and subsurface flow from groundwater outflow generated in the current REW.

#### 249 2.3 Model calibration

250 The physical meaning and value ranges of the calibrated parameters in the THREW-t model are 251 described in Table 2. Parameter values were optimized using three calibration variants: (1) single-252 objective calibration using only the observed discharge at the catchment outlet, (2) dual-objective 253 calibration using both observed discharge and MODIS SCA estimates, and (3) triple-objective calibration 254 using observed discharge, MODIS SCA estimates and  $\delta^{18}$ O measurements of stream water. Considering 255 the data availability, we chose April 1st 2006 to December 31st 2010 as the calibration period, and January 256 1<sup>st</sup> 2011 to September 30<sup>th</sup> 2012 as the validation period. For SCA, we used only the MODIS SCA 257 estimates during the ablation period (1st May to 30th July) of each year for the model calibration, because 258 simulations of runoff processes are mostly sensitive to the dynamics of snow cover extent in the melting 259 period (Duethmann et al., 2014). Only the  $\delta^{18}$ O measurements of stream water in the rainy season (from 260 the first rainfall event to the last rainfall event of each year, as shown in Table 1) were used to optimize 261 the model parameters, because the measured isotope data for precipitation were only available in this 262 season. We chose the objective functions of Nash Sutcliffe efficiency coefficient (NSE) (Nash and 263 Sutcliffe, 1970) and mean absolute error (MAE) to optimize the simulations of discharge, SCA and 264 isotope respectively (Eqs. 12-14).

265 
$$NSE_{dis} = 1 - \frac{\sum_{i=1}^{n} (Q_{o,i} - Q_{s,i})^2}{\sum_{i=1}^{n} (Q_{o,i} - \overline{Q_o})^2}$$
(10)

$$MAE_{SCA} = \frac{\sum_{i=1}^{n} |SCA_{o,i} - SCA_{s,i}|}{n}$$
(11)

267 
$$MAE_{iso} = \frac{\sum_{i=1}^{n} |\delta^{18} O_{o,i} - \delta^{18} O_{s,i}|}{n}$$
(12)

where, *n* is the total number of observations. Subscripts of *o* and *s* refer to observed and simulated variables, respectively.  $\overline{Q_o}$  is the average value of observed streamflow during the assessing period.

An automatic procedure based on the pySOT optimization algorithm developed by Eriksson et al. (2015) was implemented for all the three calibration variants to identify the behavioral parameters. pySOT used surrogate model to guide the search for improved solutions, with the advantage of requiring few function evaluations to find a good solution. An event-driven framework POAP were used for building and combining asynchronous optimization strategies. The optimization was stopped if a maximum number of allowed function evaluations was reached, which was set as 3000 in this study. For 276 the single-, dual- and triple-objective calibration variants, NSEdis, NSEdis - MAESCA, NSEdis - MAESCA -277 MAE<sub>iso</sub> were chosen as combined optimization objectives, respectively. The pySOT algorithm was 278 repeated 150 times for each calibration variant. The 150 final results were further filtered according to 279 the metric of  $NSE_{dis}$ , i.e., only the parameters producing  $NSE_{dis}$  higher than a threshold were regarded as 280 behavioral parameter sets. For single- and dual-objective calibration, the threshold was selected as 0.75. 281 Considering the trade-off between discharge and isotope simulation, the threshold was chosen as 0.70 282 for triple-objective calibration. For each calibration variant, the parameter producing highest combined 283 optimization objective was regarded as the best parameter set.

284

#### [Table 2]

#### 285 2.4 Quantifications of the contributions of runoff components to streamflow

286 The contributions of individual runoff components to streamflow were quantified based on two 287 definitions of the runoff components. In the first definition, we quantified the contributions of individual 288 water sources including rainfall, snow meltwater and glacier meltwater to the total water input, which 289 were commonly reported in previous quantifications of runoff components on the Tibetan Plateau (Chen 290 et al., 2017; Zhang et al., 2013). To be noted, the sum of the three water sources should be larger than the 291 simulated volume of runoff because of the evaporation loss. Thus, contributions quantified in this 292 definition only refer to the fractions of the water sources in the total water input forcing runoff processes, 293 rather than the actual contributions of water sources to streamflow at the basin outlet. In the second 294 definition, runoff components were quantified based on the runoff generation processes including surface 295 runoff and subsurface flow. Surface runoff consists of runoff triggered by rainfall and meltwater that feed 296 streamflow through surface paths, and the precipitation occurring in river channel and contributes to 297 runoff directly. Subsurface flow is the interflow from groundwater outflow.

#### 298 2.5 Estimation of the water travel time and residence time

299 In this study, the water travel time is estimated by three methods, a lumped analytical method and 300 two distributed model-based methods. A simplified lumped method, sine-wave method (SW) was used 301 to provide a reference value of mean travel time (MTT) and mean residence time (MRT) in the catchment. 302 The adopted model-based methods were developed by van Huijgevoort et al. (2016) and Remondi et al. 303 (2018), which were referred to as mass-mixing method (MM) and flux-tracking method (FT), 304 respectively. SW method is based on the isotope data of precipitation and stream water.MM and FT 305 methods were conducted by the tracer-aided hydrological model using behavior parameter values 306 identified by the calibration scenarios.

307 SW method has a stationarity assumption that a constant flow field gives constant travel time 308 distribution (TTDs) (van der Velde et al., 2015). It assumes the form of TTD, and derives the MTT 309 directly from the series isotopic data (McGuire and McDonnell, 2006). Although the assumption is rather 310 stringent, SW is widely used in the studies when an approximate estimation of MTT is required (e.g., 311 Kirchner, 2016; Garvelmann et al., 2017). Here we assumed the form of TTD as the exponential function, 312 and the MTT can be estimated according to Eqs. 13-14 (McGuire and McDonnell, 2006; Garvelmann et 313 al., 2017):

314 
$$\delta_t = \bar{\delta} + A * \sin\left(\frac{2\pi}{365} * t + \varphi\right) \tag{13}$$

315 
$$MTT = \frac{\sqrt{\left(\frac{1}{A_r/A_p}\right)^2 - 1}}{2\pi}$$
(14)

316 where,  $\delta_t$  is the calculated  $\delta^{18}$ O of stream water or precipitation on day *t* of the year.  $\bar{\delta}$  is the mean  $\delta^{18}$ O 317 of stream water or precipitation measured in different seasons. *A* and  $\varphi$  are parameters controlling the 318 amplitude and phase lag, and are estimated based on the fitness between the sine-wave curve and the 319  $\delta^{18}$ O measurements. Subscripts of *r* and *p* in Eq. 14 represent river and precipitation, respectively.

320 MM method was used to estimate the water age of outflow and water storage in the catchment. For 321 the outflow (e.g., stream water, evaporation), the concept of water age is consistent with the concept of 322 "travel time conditional on exit time" by Botter et al. (2011), "flux age" by Hrachowitz et al. (2013), and 323 "backward travel time" by Harman and Kim (2014). For the water storage (e.g., soil water, groundwater, 324 snowpack), the concept of water age is consistent with the concept of "residence time" by Botter at al. 325 (2011) and "residence age" by Hrochowitz et al. (2013). MM method regarded the water age as a kind 326 of tracer, and simulated the "concentration" of this tracer of the water bodies including snowpack, soil 327 water and stream water (van Huijgevoort et al., 2016; Ala-aho et al., 2017). The "mass" and 328 "concentration" of the water age were simulated similarly in Eqs. 8-9, by replacing  $\delta^{18}$ O with water age 329 of the multiple terms. Event precipitation entering the catchment was treated as new water with a 330 youngest age equaling to the simulation step of model. The glacier meltwater was regarded as very old 331 water, and a constant age of 1000 days was adapted in this study. Meanwhile, the age of water stored in 332 snowpack, soil and river channel were assumed to increase with the ongoing simulation time: water age 333 increased by one day after each model running at a daily step.

334 FT method ran the model multiple times in parallel to track the fate of each precipitation event 335 separately (Remondi et al., 2018). All days with precipitation were individually labeled and tracked over 336 the simulation period by adding an artificial tracer to the water amounts which was assumed to not 337 otherwise exist anywhere. The snow meltwater was tracked from the time when the snow entered the 338 catchment as solid precipitation (i.e., snowfall), rather than the time when the snowpack melted. Glacier 339 meltwater was not tracked, because the evolution of glacier was not simulated in the model, and the travel 340 time of glacier melt as surface runoff was negligible. Similar as MM method, the MTT of glacier melt 341 runoff was assumed as a constant value as 1000 days. The mixing and transport processes of the tracer 342 were also simulated similarly in Eqs 8-9 by replacing  $\delta^{18}$ O with the concentration of the artificial tracer. 343 By summarizing the mass of labeled precipitation in the water storage and stream water, the TTD 344 conditional on exit time (backward TTD), TTD conditional on injection time (forward TTD) and 345 residence time distribution (RTD) can be derived.

346 In summary, this study estimated the water travel time and residence time using a lumped method 347 (SW), and two model-based methods (MM and FT), and the results of three methods were compared to 348 test the robustness of travel time estimation in this glacierized basin. Specifically, SW method estimated 349 the MTT of total discharge and the MRT of water storage directly based on the isotopic data in stream 350 water and precipitation. MM method estimated the water age of stream water and groundwater storage, 351 representing the daily backward MTT and MRT respectively, and all the 19 behavioral parameter sets of 352 triple-objective calibration were used to illustrate the uncertainty of MTT. FT method estimated the time-353 varying precipitation-triggered TTD and RTD, only using the parameter set producing best metric. To 354 make the result of FT method comparable to MM method, the glacier melt runoff was also assumed to

have MTT (water age) of 1000 days to calculate the MTT of the total runoff generation as the weighted average value of the MTT of precipitation runoff (including rainfall and snowmelt) and glacier melt runoff according to the contribution of water sources. The glacier melt was assumed to only contribute

358 to surface runoff directly and exit the catchment rapidly, thus had no influence on the MRT estimation.

#### 359 **3. Results**

#### 360 **3.1 Model performance on the simulations of discharge and isotopic composition**

361 For the calibration period, the single-objective calibration produced good performance for the 362 simulation of discharge, but had an extremely poor performance for the simulations of SCA and  $\delta^{18}$ O 363 (Table 3). Involving SCA in the calibration objective, the dual-objective calibration significantly 364 improved the simulation of SCA, and kept a good behavior on discharge simulation, but brought no 365 benefit to the isotope simulation. The triple-objective variant led to a good performance for all the three 366 metrics. The NSE<sub>dis</sub> produced by triple-objective calibration was slightly lower than that of another two 367 variants because of the lower threshold for behavior parameter sets. The simulation of isotopic 368 composition of stream water was significantly improved by triple-objective calibration compared to the 369 other two variants. For the validation period, the NSE<sub>dis</sub> of triple-objective calibration was significantly 370 improved, even better than the single-objective, indicating the improved process representation of the 371 behavior parameters by the triple-objective calibration. Through 150 runs of calibration program, triple-372 objective calibration got the smallest behavior parameter sets, indicating that involving additional 373 calibration objectives increase the identifiability of model parameters and reduce the equifinality.

#### 374

#### [Table 3]

375 Fig. 2 shows the uncertainty ranges of the simulations for the behavioral parameters obtained by 376 the three calibration variants. The three variants generally produced similar hydrographs in terms of the 377 magnitudes and timing of peak flows with averaged behavioral parameter sets, but the triple-objective 378 had a narrower uncertainty range, especially for the baseflow dominated periods (Figs. 2a-c). The single-379 objective variant resulted in rather large uncertainty ranges for the simulations of SCA and isotopic 380 composition (Figs. 2d and g). The good fitness between the simulated and observed streamflow in 381 summer is likely due to the largely overestimated rainfall-triggered surface runoff, because of the 382 underestimated reduction of SCA in spring. The dual-objective calibration significantly reduced the 383 uncertainty range of the SCA simulation, and captured the declining SCA in summer very well (Fig. 2e). 384 Including SCA in the model calibration, however, only provided small benefits for the simulation of  $\delta^{18}$ O 385 in stream water (Fig. 2h). Simulations of the triple-objective variant properly reproduced the temporal 386 variation in SCA in the melt season, despite the slightly reduced performance compared to that of the 387 dual-objective variant (behaving as higher MAE<sub>SCA</sub> of triple-objective calibration in Table 3). Meanwhile, 388 the seasonal variations of  $\delta^{18}$ O of stream water were reproduced well by the triple-objective calibration 389 (Fig. 2i).

#### 390

#### [Figure 2]

Fig. 3a shows median value of the simulated daily inputs of water sources (rainfall, snowmelt, and glacier melt) for the calibration period obtained by the behavioral parameter sets of the triple-objective variant. All the three water sources started to contribute to stream water in around April. The volume of snowmelt peaked around June, and then decreased rapidly in July as the catchment SCA decreased significantly. The volumes of rainfall and glacier melt peaked in mid-summer which was the wettest and warmest period in the year. The fluctuations of the simulated  $\delta^{18}O$  of stream water in Fig. 3b are generally consistent with the varying contributions of these water sources to runoff. At the beginning of the wet season,  $\delta^{18}O$  of stream water increased rapidly in response to the dominance of the isotopic enriched precipitation. The  $\delta^{18}O$  of stream water began to decease in the late wet season, likely because of the reduced  $\delta^{18}O$  of precipitation reported as "temperature effect" (Dansgaard, 1964) which is mainly due to the effect of southwest monsoon (Yin et al., 2006), as well as the increased contributions of isotopic depleted glacier melt.

403

#### [Figure 3]

#### 404 **3.2** Contributions of runoff components

405 The results of runoff component quantification reported in this section were based on the behavioral 406 parameter sets of the three calibration variants. Table 4 and Figure 4 shows the proportions of water 407 sources in the mean annual water input during 1<sup>st</sup> January 2007 to 31<sup>st</sup> December 2011. In all the three 408 calibration variants rainfall provided most of the water quantity for runoff generation (44.2% to 48.0%), 409 because of the high partition of rainfall (around 347mm) in the annual precipitation (around 587mm). 410 The single-objective variant estimated the lowest proportion of snowmelt (19.7%), because the 411 simulation of SCA was not constrained in the calibration, leading to largely overestimated SCA in 412 comparison to the MODIS SCA estimates due to less melting (Fig. 2d). The dual-objective variant 413 estimated the highest proportion of glacier melt (33.8%), resulting in a lower proportion of rainfall 414 (44.2%). Involving the calibration objective of isotope, the triple-objective variant estimated the lowest 415 proportion of glacier melt (29.2%) by rejecting the parameter sets that produced high contribution of 416 glacier melt (as shown in Fig. 4), which will be discussed more in detailed in the discussion section. To 417 be noted, despite above differences, the results of three calibration variant were quite similar, with the 418 maximum difference lower than 5%. However, the uncertainties of the simulated water proportions 419 decreased substantially with the increase of data that was involved in the calibration, showing as a 420 decreasing uncertainty (12.4% to 6.2%, Table 4) and fewer outliers (Fig. 4), demonstrating considerable 421 values of additional datasets for constraining the simulations of corresponding runoff generation 422 processes.

#### 423

#### [Table 4]

424

## [Figure 4]

425 Fig. 5 and Table 4 compare the seasonal proportions of water sources in the total water input of the 426 three calibration variants. The seasonal dominance of the water sources on runoff estimated by the three 427 calibration variants are similar. In particular, the proportion of rainfall was large (around 55%) in summer 428 but small in winter when rainfall rarely occurred. Snowmelt and glacier melt dominated the total water 429 input in winter with proportions of around 60% and 40%, respectively. The proportion of meltwater in 430 summer was relatively low because of the dominance of rainfall during the summer monsoon. Snowmelt 431 could only account for around 15% in the total water input in summer because of the significantly reduced 432 snowpack. The proportion of glacier melt was higher than that of snowmelt in summer because of the 433 decreasing snow cover area. In the spring months, snowmelt and glacier melt contributed around 55%-434 60% and 35%-30% to the total water input, respectively, and rainfall provided the remaining 5%. The 435 glacier melt provided a steady contribution of around 30%-40% throughout the entire hydrological year. 436 The seasonal proportions of water sources show slightly different among the calibration variants. 437 Specifically, the triple-objective calibration estimated not only the highest snowmelt and lowest glacier

438 melt in the three seasons except winter, but also the highest contribution of rainfall in summer (Fig. 5c), 439 which was consistent with the lowest contribution of glacier melt in total water input estimated by triple-440 objective calibration. Single-objective calibration produced the highest contribution of rainfall in autumn, 441 and the highest contribution of snowmelt in winter (Fig. 5a). Although the contribution of water sources 442 exhibited a large uncertainty in winter, and significant difference existed among the calibration variants, 443 it had negligible effect on the annual result, because of the extremely low contribution of water input 444 during winter (<1%). The uncertainty ranges of the seasonal proportion during summer and autumn were obviously reduced by the triple-objective calibration (Fig. 5c).

- 445
- 446

#### [Figure 5]

447 Table 5 shows the contributions of runoff components to annual and seasonal runoff. Three 448 calibration variants resulted in rather similar contributions of surface runoff and subsurface runoff 449 (around 65% and 35% respectively). Surface runoff was the dominant component in this catchment, 450 because of the large glacier covered area (around 20%) and the large saturation area (around 20%). The 451 triple-objective calibration estimated relatively lowest surface runoff (64.9%) and highest subsurface 452 runoff (35.1%). Surface runoff dominated streamflow during spring and summer (with proportions of 453 around 65% and 80%, respectively), when large rainfall and snowmelt events occurred frequently and 454 the catchment was rather wet. Subsurface runoff played a more important role during autumn, accounting 455 for about 60% of the runoff. The runoff in winter was dominated by baseflow, because of the very rare 456 water input events. Again, the triple-objective calibration resulted in lowest uncertainty ranges for the 457 contributions of the runoff components compared to the other two calibration variants (4.1% compared 458 to 12.1%). Isotope data used in the triple-objective variant provided additional constraints on the 459 estimation of parameters controlling the generation of subsurface flow (such as KKA and KKD in Table 460 2) and the saturation area where surface runoff occurred (such as WM and B), thus constraining the 461 partitioning between surface runoff and subsurface runoff.

462

#### [Table 5]

#### 463 3.3 Estimations of water travel time and residence time

464 The travel time and residence time were estimated for the five water years (2007/1/1 - 2011/12/31)465 during the simulation period. The result produced by the best parameter set ((NSE<sub>dis</sub> = 0.72, MAE<sub>SCA</sub> = 466 0.079, MAE<sub>iso</sub> = 0.484) was used to test the consistency between the two model-based methods. Based 467 on the assumption that glacier melt water had an age of 1000 days, the backward MTT and MRT 468 estimated by MM (FT) method were 1.70 (1.72) and 1.22 (1.17) years, respectively. Fig. 6 shows the 469 comparison between the results of MM and FT methods. As shown in Fig. 6a and 6d, there were strong 470 correlation between the daily MTTs (MRTs) estimated by the two methods with a high correlation 471 coefficient of 0.96 (0.98). The daily MTT and MRT series also showed similar temporal variability 472 between the two methods as shown in Fig. 6b and 6e. The MRT increased steadily during dry season, 473 and decreased rapidly during wet season due to the recharge of young precipitation. The daily MTT also 474 showed steady increasing trend during dry season, but showed significant fluctuation during wet season 475 because of the combined effect of young precipitation and old glacier meltwater. Fig. 6c and 6e shows 476 the probability density function of the daily backward MTT and MRT produced by the two methods. The 477 daily MTT had a large range from 0.42 to 2.75 years, with several peak density values at around 1, 1.5 478 and 2.67 years, including the influence of the multiple water sources with different ages. On the contrary, 479 the daily MRT only had a narrow range from 0.75 to 1.75 years, with a significant peak value at around 480 1.25 years, similar with the MRT. Excluding the effect of glacier meltwater, FT method estimated the
481 precipitation-triggered backward MTT of runoff as 263 days, significantly smaller than the MRT,
482 indicating the incomplete mixing in the catchment scale caused by the distributed modelling framework.

483

#### [Figure 6]

484 The lumped SW method estimated the MTT and MRT as 1.68 years ( $A_r$  and  $A_p$  were estimated as 485 0.58 and 6.19, respectively). Based on the average result of 19 behavioral parameter sets, the model-486 based methods estimated the MTT and MRT as 1.61 and 1.28 respectively. The two kinds of methods 487 produced similar MTT, indicating the robustness of travel time estimation in this catchment. The 488 precipitation-triggered MTT (shorter than 1 year) was significantly smaller than the MTT of total runoff 489 estimated by the lumped method, indicating the effect of old glacier melt water. The glacier melt 490 contributed to stream water through surface runoff directly, and had no contribution to the water storage, 491 leading to a smaller model-based MRT compared to MTT. The uncertainty of MTT and MRT estimation 492 could be reflected by the range produced by MM method by the 19 behavioral parameter sets as shown 493 in Fig. 7. The standard deviation of the estimated MTT and MRT were 74 and 79 days, respectively. The 494 uncertainty range during June to August was relatively small (Fig. 7a), indicating that different behavioral 495 parameters produced similar precipitation-triggered processes during the wet season, and the uncertainty 496 mainly came from the large range of MRT, i.e., the age of water storage including soil water and 497 groundwater.

#### [Figure 7]

499 Based on the best parameter set, FT method tracked the transportation of precipitation and produced 500 time-varying forward TTD, backward TTD and RTD. For simplicity, Fig. 8 shows the average 501 distributions weighted by the precipitation amount (for forward TTD), runoff generation (for backward 502 TTD) and water storage (for RTD). As shown in Fig. 8a and 8b, the forward and backward TTD were 503 similar, behaving as a high proportion ( $\sim 0.3$ ) of the youngest water, which was consistent with the high 504 proportion of rapid surface runoff. The high proportion of young water led to a similar TTD form as 505 exponential model. The relative peaks of TTD were mainly around the travel time of integral years, 506 indicating the influence of baseflow from groundwater, which were recharged by precipitation in the wet 507 seasons of previous years. The simulated RTD was significantly different from the TTD, behaving as the 508 low probability density of young water. The young water was mainly recharged by the infiltrating rainfall 509 and snowmelt, which was negligible compared to the total water storage. Again, the difference between 510 TTD and RTD indicated the incomplete mixing processes, behaving as affinity for young water due to 511 the rapid flow pathways such as surface runoff.

512

498

#### [Figure 8]

#### 513 **4. Discussions**

# 4.1 The values of tracer on constraining flow pathways and water storages in the hydrologicalmodel

This study developed a tracer-aided hydrological model and tested its behavior in a glacierized catchment. Because of the sampling difficulties on the Tibetan Plateau, the tracer data of the water sources (e.g., snow, glacier, groundwater) was rather limited compared to other tracer-aided modelling works (e.g., Ala-aho et al., 2017; He et al., 2019). Nonetheless, the model developed in this study performed well on producing the tracer signature of stream water, producing a tool for applying the tracer-aided method to the areas with limited tracer data. Although it was widely accepted that simple input-output tracer measurements provided limited insight into catchment function, and sampling source water components would be helpful (Birkel et al., 2014; Tetzlaff et al., 2014), the uncertainty of model could still be reduced significantly by satisfying the output tracer signature (Delavau et al., 2017), especially in cold regions where hydrological processes were more complex. The fact that the model can simultaneously satisfy three calibration objectives over a long period gave confidence in the model realizations (McDonnell and Beven, 2014).

528 Our results indicate that involving the isotope into calibration significantly reduced the uncertainty 529 of quantifying the runoff components. To understand the role of isotope data on reducing the uncertainty, 530 the results of dual-objective calibration variant were analyzed why some of the parameter sets behaved 531 poorly on isotope simulation despite their good performance on discharge and snow simulation. Among 532 the 117 behavioral parameter sets of dual-objective calibration, only 14 of them produced relatively good 533 isotope simulations (MAE<sub>iso</sub> < 1.0). As shown in Fig. 9a and 9b, these 14 isotopic behavioral parameter 534 sets produced the proportion of runoff component within a relatively smaller range (27.5% to 38% for 535 glacier melt, and 58% to 75% for surface runoff), while the 117 behavioral parameter sets produced a 536 much larger variation (24% to 53% for glacier melt, and 40% to 90% for surface runoff). This indicated 537 that involving isotope data for model calibration helped to exclude some unreasonable proportions of 538 runoff component. The distribution of scatter in Fig. 9a and Fig. 9b was similar, and the proportion of 539 surface runoff had a strong correlation with the proportion of glacier melt as shown in Fig. 9c (because 540 of the assumption that glacier melt contributed to surface runoff directly), thus the mechanism that 541 isotope can reject unreasonable proportions were the same for water sources and runoff generation 542 processes. Fig. 9d shows the simulation range of  $\delta^{18}$ O of stream water by calibrated parameters that 543 resulted in glacier melt proportion in the total water input higher than 40%. The simulated isotopic 544 signature showed strong fluctuations due to the high proportion of surface runoff with a larger time 545 variation compared to the relatively steady signature of subsurface runoff. Also, the simulated isotopic 546 values were significantly higher than the observations, which was mainly the result of the excessive 547 isotopic fractionation due to the too much evaporation of surface water (Hindshaw et al., 2011; Wolfe et 548 al., 2007). Fig. 9e shows the simulation range by the parameters with proportion of surface runoff lower 549 than 45%. In contrast to scenarios with too high glacier melt, the simulated isotope signature showed 550 small variation and the mean values were much lower than the observation. Our result also showed that 551 the proportion of surface runoff and glacier melt tended to be higher when the NSE<sub>dis</sub> was higher, 552 indicating that focusing on the simulation of integrated observation of discharge only will likely lead to 553 overestimated surface runoff and glacier melt. These results indicated that the isotope data helped to 554 constrain the quantifications of runoff components by (1) regulating the competition between rapid 555 component with strong variation of isotope signatures (e.g., surface runoff) and slow component with 556 relatively stable isotope signatures (e.g., subsurface runoff) to match the daily fluctuations of observed 557 isotope signature of stream water, and (2) controlling the isotopic fractionation by adjusting the 558 evaporation to satisfy the observed isotopic value.

559

#### [Figure 9]

560 Two model-based methods (MM and FT) were adopted to estimate travel time and residence time 561 in this study, and verified by the result of lumped method (SW). Both MM and FT methods can estimate 562 MTT and MRT, but FT provided more information including TTD and RTD, which was actually more 563 of interest. MM method has been used in several previous studies, including modelling work in snow564 influenced basins (Ala-aho et al., 2017). Consequently, the results of FT and MM were compared in this 565 study, to ensure that the additional information provided by FT method was reasonable. Our result 566 indicated that the two model-based methods produced consistent results, which were also similar with 567 the lumped method, indicating the robustness of MTT and MRT estimation through a tracer-aided model 568 without defining any prior distribution functions.

569 Although significantly constraining the proportion of runoff component, the uncertainty ranges of 570 simulated MTT and MRT, especially that during baseflow-dominant period (as shown in Fig. 7b) were 571 still rather large, indicating that the estimation of groundwater age had a large uncertainty, which was 572 similar with other model-based age estimation works (e.g., Ala-aho et al., 2017; van Huijgevoort et al., 573 2016). The isotope observations were mainly collected during wet season when precipitation-triggered 574 surface runoff played an important role in runoff generation, thus this process was constrained relatively 575 well by the isotope calibration, showing as the similar fluctuation of MTT during wet season produced 576 by different parameter sets. Although the proportion of subsurface runoff was constrained, the storage 577 volume of groundwater was poorly constrained, because of the relatively simplified structure of the 578 groundwater module of THREW model (Tian et al., 2006), which adopted a two-layer reservoir model 579 to describe the processes of seepage and subsurface flow. Apart from involving more calibration 580 objectives, improving the physical mechanism and the representation of hydrological processes is another 581 important way to constrain the model behavior and reduce uncertainties.

#### 582 **4.2 Insight from MTT and MRT estimation**

583 Based on three MTT estimation methods, this study estimated the MTT and MRT as 1.7 and 1.2 584 years, reflecting the age of stream water and subsurface water storage in the catchment. Excluding the 585 effect of aged glacier meltwater, the MTT of stream water fed by rainfall and snowmelt decreased to 586 around 9 months. Ala-aho et al. (2017) estimated the median water ages of three snow influenced 587 catchments located in the USA, Sweden and Scotland using the MM method as 11 months, 1.5 years and 588 5 years. The water age had a rather wide range among different catchments, which mainly depends on 589 the groundwater storage controlled by the topography and soil characteristics. The process that water 590 travel through the subsurface pathway play comparable role with the snow accumulation process on the 591 water age in snow influenced catchment. The significantly lower MTT and MRT estimated in this study 592 than that of the three catchments in Ala-aho et al. (2017) can be mainly attributed to the large impermeable 593 area on the glacier, leading to a large proportion of surface runoff with very short travel time.

594 The influence factors of the MTT and MRT were analyzed to better understand the hydrological 595 processes in the catchment. The relationship between the daily travel times (including forward MTT, 596 backward MTT and MRT) and the environmental factors (including simulated soil water content and 597 meteorological factors) was analyzed in Fig. 10. The result shows the scatter diagrams between the 598 factors with good correlation. MRT presented a strong negative correlation with the soil water content 599 (Fig. 10a), which was consistent with the finding by Hrachowitz et al. (2013) and Heidbuechel et al. 600 (2012) that the resident water age was sensitive to antecedent wetness and the water stored in subsurface 601 storage. During dry season, the MRT got older as time went on, and the soil water content was decreasing 602 due to the outflow of groundwater. The soil water content increased rapidly during wet season when 603 precipitation occurred and recharged the groundwater, and the MRT got younger due to the recharge of 604 young water. Backward MTT had good relationship with both soil water (Fig. 10b) and precipitation 605 amount (Fig. 10c). During dry season when runoff was mainly baseflow, the age of stream water was 606 similar with the groundwater and consequently controlled by the soil water content. During wet season, 607 the backward MTT was largely dependent on the precipitation amount, because a large proportion of 608 young precipitation event water contributed to stream water quickly through surface runoff pathway, 609 leading to small MTT. This was also consistent with the result reported by Hrachowitz et al. (2013) and 610 McMillan et al. (2012) that the water flux age was controlled by fast and slow processes under wetting-611 up and drying-up conditions. The forward MTT reflected the time that the input water took to travel 612 through the catchment, showing good relationship with temperature (Fig. 10d). The negative correlation 613 between forward MTT and temperature was mainly related to two processes, i.e., the snow accumulation, 614 and the evaporation. When the temperature was low, the precipitation was mainly in the form of snowfall, 615 which cannot contribute to the stream quickly, resulting in a long travel time. On the contrary, the water 616 would be exposed to intense evaporation when the temperature was high. Large proportion of 617 precipitation event water left the catchment quickly by evaporation before infiltrating into groundwater 618 and going through the long subsurface pathway, resulting in a short travel time. The negative relation 619 between forward MTT and temperature indicated an accelerated water cycling process as a result of 620 climate warming.

621

#### [Figure 10]

#### 622 4.3 Limitation and uncertainty

623 Multiple water sources brought difficulties to hydrological modelling in glacierized basins 624 (Zongxing et al., 2019). Focusing on the tracer transportation processes, the model developed in this 625 study made some simplifications on the processes related to snow and glacier to make the model structure 626 parsimonious. First, the snow accumulation and melting processes were simulated by a simple 627 temperature-based method, which was relatively lack of physical mechanism compared to the energy-628 based methods (e.g., Pomeroy et al., 2007). Nonetheless, this method had an acceptable behavior and 629 was widely used in studies of snow simulation (e.g., He et al., 2014), and the simulated SCA was 630 validated by the MODIS data during ablation period in this study. Second, the evolutions of glacier 631 thickness and area were not simulated in the model. Simplification of a constant glacier area likely led 632 to an overestimation of the contribution of glacier melt to runoff, as the glacier cover area should get 633 smaller due to the climate warming. However, this simplification should have minor influence on the 634 result because the changes of glacier area was rather small in a short simulation period of seven years.

635 The influence of calibration objective function was inadequately assessed in this study. Although 636 the measurement units of NSE<sub>dis</sub> was different from the MAE<sub>SCA</sub> and MAE<sub>iso</sub>, their values were in the 637 same order of magnitude when the model performance were acceptable (similarly with He et al., 2019). 638 Consequently, they were combined directly to reflect the simultaneous performance on the three 639 objectives. Plenty of studies have developed methods to solve the problem of multiple objective 640 calibration by introducing an integrated evaluation metric (e.g., Gupta et al., 2009; Shafii and Tolson, 641 2015) or giving weights to each objective (e.g., Tong et al., 2021). This study mainly aimed to evaluate 642 the value of isotope data on improving the model behavior, rather than developing a general calibration 643 strategy, thus the three evaluation metrics were added together with equal weights for simplification, to 644 represent the condition that all the three objectives were simulated well. Our main findings indicated that 645 the results calibrated by discharge and isotope were more behavioral in many aspects than the results 646 calibrated only by discharge. The potential influences of calibration metrics and methods on the finally 647 optimized result still needed further exploration.

648 The lack of source water sampling made it difficult to fully validate the modelling result. Although 649 the isotope signature of stream water was reproduced well, it cannot guarantee that the isotopic variations 650 of groundwater, snowmelt were simulated correctly. The quantification of runoff components was also 651 hard to verified. The end-member method cannot be applied as a reference due to the lack of water source 652 tracer data. A previous study of snow cover and runoff modelling work in the same basin (Zhang et al., 653 2015) provided a potential reference. That work indicated that the contribution of rainfall, snowmelt and 654 glacier melt in 2006 were 30%, 10% and 60%, respectively, which was markedly different from the result 655 of this study. The runoff simulation in Zhang et al. (2015) was conducted by a simplified conceptual 656 model with limited physical mechanism, which did not consider the processes of subsurface runoff and 657 evaporation. And the glacier melt runoff coefficient (the ratio of glacier melt runoff to the total glacier 658 melt) estimated by that study was very small (0.182), indicating that a large proportion of glacier melt 659 did not contribute to the surface runoff directly, which is inconsistent with the common assumption in 660 previous studies (e.g., Seibert et al., 2018; Schaefli et al., 2005). The extremely low glacier melt runoff 661 coefficient might lead to overestimation of the contribution of glacier melt. The significant differences 662 between the two studies mainly resulted from the difference of model structure. Intensive source water 663 sampling together with systematic glacier observation might improve the behavior of hydrological model 664 in glacierized basins and help us better understand the runoff processes.

665 In glacierized basins where glacier meltwater played an important role on runoff generation, the 666 object of the three MTT estimation methods were different. The total runoff could be divided into 667 precipitation-triggered runoff (including rainfall-runoff and snowfall-snowmelt-runoff) and glacier melt 668 runoff. Considering that glacier was also formed by the precipitation over past years, the lumped SW 669 method should have reflected both runoff processes, because it was based on the tracer data of 670 precipitation and total runoff. The two model-based methods mainly focused on the precipitation-671 triggered runoff, because the glacier revolution process was simplified in the model. The MTT estimation 672 of total runoff should be based on the assumed MTT of glacier melt water. In this study, assuming the 673 MTT of glacier melt as 1000 days, the model-based results were similar with SW method, indicating that 674 the assumption of glacier melt MTT was appropriate, which was actually misleading. The time scale of 675 glacier update was much longer than this assumed value, because glacier generally took decades to 676 hundreds of years to move from accumulation zone to ablation zone (Soncini et al., 2014; Yao et al., 677 2012). The good agreement among the three methods indicate that the SW method significantly 678 underestimated the age of glacier. This was mainly due to the limited applicable time scale of stable 679 isotope in water. It was reported that seasonal cycle of stable isotope in precipitation were most useful 680 for inferring relatively short travel time of 2-4 years (McGuire and McDonnell, 2006; Sprenger et al., 681 2019; Stewart et al., 2010). The assumed glacier melt MTT of 1000 days was within this range, thus the 682 similar result of three methods could verify the model representation of the precipitation-triggered runoff 683 process, and the cross validation between MM and FT methods further enhanced the robustness of the 684 travel time estimation. Consequently, we can expect that if a tracer suitable for longer travel time (e.g., 685 <sup>14</sup>C) was used to estimate the proper MTT of total runoff, we could better infer the age of water according 686 to the model-based estimation of precipitation-triggered runoff MTT.

#### 687 5. Conclusions

688 A tracer module was integrated into the THREW hydrological model to constrain the various runoff 689 processes, and was tested in a glacierized catchment on the Tibetan Plateau. Measurements of oxygen 690 stable isotopes of the stream water were used to calibrate the model parameters, in addition to the 691 observations of discharge and MODIS SCA. The behaviors of the model, especially the quantifications 692 of runoff components were compared among the calibration variants with different objective, to test the 693 value of isotope data on constraining the model parameters. A lumped method (SW) and two model-694 based methods (MM and FT) were applied to estimate the water travel time in the study basin. Our main 695 findings are: (1) The THREW-t model performed well on simultaneously reproducing the variations of 696 discharge, snow cover area, and the isotopic composition of stream water, despite of a small water sample 697 number of precipitation was available to provide isotope input data. (2) The contributions of rainfall, 698 snowmelt and glacier melt to the annual runoff were quantified as 47.4%, 23.4% and 29.2%. Surface 699 runoff (contributing around 64.9%) was more dominant than subsurface flow in the annual runoff. 700 Calibration with isotope data significantly reduced the uncertainties by regulating the competition 701 between rapid and slow runoff components to fit the variation of observed isotope signature, and resulted 702 in more plausible quantifications of contributions of runoff components to seasonal runoff. (3) The 703 estimated MTT of model-based methods MM and FT met well with that of a sin-wave lumped parameter 704 method, indicating the robustness of travel time estimation benefiting from the use of water isotope data. 705 The precipitation-triggered MTT was significantly shorter than the MTT of total runoff, indicating the 706 effect of old glacier meltwater. The MRT was longer than precipitation-triggered MTT, indicating the 707 catchment scale incomplete mixing processes, and the affinity for young water due to the rapid flow 708 pathways such as runoff on impermeable glacier surface. The temporal variation of MTT and MRT was 709 dependent on the catchment wetness conditions and meteorological factors.

#### 710 Code/Data availability

711 The isotope data and the code of THREW-t model used in this study are available by contacting the 712 authors.

#### 713 Author contribution

YN, ZH and FT conceived the idea; LT provided the observation data; YN, FT, LT and ZH conducted
 analysis; LS provided comments on the analysis; all the authors contributed to writing and revisions.

#### 716 **Competing interests**

717 The authors declare that they have no conflict of interest.

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975		List of figures				
976	1.	Figure 1. Location and topography of the study area				
977 978 979	2.	<b>Figure 2.</b> Uncertainty ranges of simulations in the calibration period produced by the behavioral parameter sets of the single-objective (subfigure a to c), dual-objective (subfigure d to f) and triple-objective (subfigure g to i) calibration variants.				
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993 994 995 996 997 998	9.	Figure 9. The role of isotope calibration on constraining the proportion of runoff components. (a) The relationship between $MAE_{iso}$ and proportion of glacier melt. (b) The relationship between $MAE_{iso}$ and proportion of surface runoff. (c) The relationship between proportion of surface runoff and that of glacier melt. (d) The simulated isotope in stream water produced by the parameter sets estimating proportion of glacier melt higher than 40%. (e) The simulated isotope in stream water produced by the parameter sets estimating proportion of surface runoff lower than 45%.				
999 1000 1001	10.	<b>Figure 10.</b> The scatter diagrams between (a) MRT and soil water content, (b) backward MTT and soil water content, (c) backward MTT and rainfall, and (d) forward MTT and temperature.				





**Figure 1.** Location and topography of the study area



Figure 2. Uncertainty ranges of simulations in the calibration period produced by the behavioral
parameter sets of the single-objective (subfigure a to c), dual-objective (subfigure d to f) and tripleobjective (subfigure g to i) calibration variants.



Figure 3. Daily simulations of (a) each water source and (b) the corresponding isotopic compositions.
The black points and red/blue lines in subfigure (b) represent the isotope composition of the water sources
as represented by the corresponding color in subfigure (a).



1016 Figure 4. Average proportion of different sources in the annual water input for runoff generation



Figure 5. Seasonal contributions of rainfall, snowmelt and glacier melt to total water input estimated by
the (a) single-objective, (b) dual-objective and (c) triple-objective calibration variants. The error bars
indicate the uncertainty ranges simulated by the corresponding behavior parameter sets.



Figure 6. Comparison between the MM and FT methods: scatterplots for daily (a) MTT and (d) MRT;
time series of the daily (b) MTT and (e) MRT; and probability density functions of the daily (c) MTT and
(f) MRT.



1029 Figure 7. Uncertainty ranges of time series (a) MTT and (b) MRT simulated by MM method.1030





1032 Figure 8. The weighted average probability distributions of (a) forward TTD, (b) backward TTD, and (c)

1033 RTD estimated by FT method



**Figure 9.** The role of isotope calibration on constraining the proportion of runoff components. (a) The relationship between MAE<sub>iso</sub> and proportion of glacier melt. (b) The relationship between MAE<sub>iso</sub> and proportion of surface runoff. (c) The relationship between proportion of surface runoff and that of glacier melt. (d) The simulated isotope in stream water produced by the parameter sets estimating proportion of glacier melt higher than 40%. (e) The simulated isotope in stream water produced by the parameter sets estimating proportion of surface runoff lower than 45%.



Figure 10. The scatter diagrams between (a) MRT and soil water content, (b) backward MTT and soil
water content, (c) backward MTT and rainfall, and (d) forward MTT and temperature.

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Year	Period	Precipitation sample number	Stream sample number
2006	April 6 <sup>th</sup> to November 11 <sup>th</sup>	24	31
2007	April 23 <sup>rd</sup> to October 9 <sup>th</sup>	39	25
2010	May 5 <sup>th</sup> to October 18 <sup>th</sup>	63	23
2011	March 28 <sup>th</sup> to November 6 <sup>th</sup>	69	32
2012	June 16 <sup>th</sup> to September 22 <sup>nd</sup>	42	14

**Table 1.** Characteristics of precipitation and stream samples

**Table 2.** Calibrated parameters of the THREW-t model

Symbol	Unit	Physical descriptions	Range
nt	-	Manning roughness coefficient for hillslope	0-0.2
WM	cm	Tension water storage capacity, used in Xinanjiang model (Zhao,	0-10
		1992) to calculate saturation area	
В	-	Shape coefficient used in Xinanjiang model to calculate saturation	0-1
		area	
KKA	- Coefficient to calculate subsurface runoff in $Rg=KKD$		0-6
		$(y_S/Z)^{KKA}$ , where S is the topographic slope, $K^{S_S}$ is the saturated	
		hydraulic conductivity, $y_s$ is the depth of saturated groundwater, Z	
		is the total soil depth	
KKD	-	See description for KKA	0-0.5
$T_0$	°C	Melting threshold temperature used in Eqs. (1) and (2)	-5-5
$DDF_s$	mm/°C/day	Degree day factor for snow	0-10
$DDF_G$	mm/°C/day	Degree day factor for glacier	0-10
CI	-	Coefficient to calculate the runoff concentration process using	0-1
		Muskingum method: $O_2 = C_1 \cdot I_1 + C_2 \cdot I_2 + C_3 \cdot O_1 + C_4 \cdot Q_{lat}$ , where $I_1$	
		and $O_1$ is the inflow and outflow at prior step, $I_2$ and $O_2$ is the	
		inflow and outflow at current step, $Q_{lat}$ is lateral flow of the river	
		channel, $C_3 = I - C_1 - C_2$ , $C_4 = C_1 + C_2$	
<i>C2</i>	-	See description for <i>C1</i>	0-1

calibration variant	Number of behavior parameter sets	period	NSE <sub>dis</sub> <sup>a</sup>	MAEsca	MAEiso
Single-	126	calibration	0.79	0.25	2.21
objective			(0.75-0.84)	(0.08-0.78)	(0.66-4.10)
		validation	0.79	0.24	2.53
			(0.71-0.84)	(0.07-0.79)	(0.77-4.88)
Dual-	117	calibration	0.79	0.10	2.18
objective			(0.75-0.85)	(0.08-0.18)	(0.73-4.71)
		validation	0.80	0.08	2.38
			(0.73-0.84)	(0.06-0.19)	(0.84-4.96)
Triple-	19	calibration	0.74	0.13	0.68
objective			(0.70-0.81)	(0.08-0.18)	(0.48-0.83)
		validation	0.79	0.11	0.93
			(0.73-0.84)	(0.06-0.18)	(0.72-1.19)

**Table 3.** Comparisons of the model performance produced by three calibration variants.

a: Bracketed values represent the minimal and maximal values produced by the behavioral parametersets.

Season	Water source <sup>a</sup>	Single-objective	Dual-objective	Triple-objective
Annual	Rainfall	48.0	44.2	47.4
	Snow melt	19.7	22.0	23.4
	Glacier melt	32.2	33.8	29.2
	Uncertainty	12.4	9.4	6.2
Spring	Rainfall	5.4	4.1	4.5
	Snow melt	55.5	56.3	61.6
	Glacier melt	39.1	39.5	33.9
	Uncertainty	24.2	13.7	14.2
Summer	Rainfall	55.6	53.5	56.6
	Snow melt	13.0	14.0	15.2
	Glacier melt	31.4	32.4	28.2
	Uncertainty	10.7	9.7	5.1
Autumn	Rainfall	38.7	30.9	35.0
	Snow melt	29.2	33.9	35.3
	Glacier melt	33.6	35.1	29.7
	Uncertainty	18.5	11.2	11.0
Winter	Rainfall	0	0	0
	Snow melt	66.4	55.3	63.3
	Glacier melt	33.6	44.7	36.7
	Uncertainty	26.6	22.3	31.5

1065 Table 4. Average percentages of water sources in the annual water input for runoff1066 generation.

1067 a: The uncertainty of the contribution is defined as  $E = \sqrt{E_R^2 + E_S^2 + E_G^2}$ , where  $E_R$ ,  $E_N$  and  $E_G$ 1068 represent the standard deviations of the contributions of the water sources produced by the corresponding 1069 behavioral parameter sets. Subscripts of *R*, *S* and *G* represent rainfall, snow meltwater and glacier 1070 meltwater, respectively. 1071

Season	Runoff path	Single-objective	Dual-objective	Triple-objective
Annual	Surface	65.9	66.4	64.9
	Subsurface	34.1	33.6	35.1
	Uncertainty	11.8	12.1	4.1
Spring	Surface	64.3	64.0	68.0
	Subsurface	35.7	36.0	32.0
	Uncertainty	10.4	9.2	7.2
Summer	Surface	79.9	79.0	79.1
	Subsurface	20.1	21.0	20.9
	Uncertainty	10.0	10.7	4.4
Autumn	Surface	40.1	43.1	36.6
	Subsurface	59.9	56.9	63.4
	Uncertainty	15.4	16.0	6.7
Winter	Surface	5.1	5.0	5.0
	Subsurface	94.9	95.0	95.0
	Uncertainty	9.9	4.5	2.7

**Table 5.** Simulated contributions of runoff components to annual runoff