

5 Simon Hoeg

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## 8 Abstract

9 An analysis of the runoff generation processes in terms of event, recent, and pre-recent runoff  
 10 components is demonstrated for the Swiss pre-Alpine Alp catchment (46.4 km<sup>2</sup>) and two  
 11 smaller tributaries (Erlenbach, 0.7 km<sup>2</sup>, and Vogelbach, 1.6 km<sup>2</sup>), whereas, recent water is  
 12 understood as the contribution of the respective three prior rainfall-runoff events. A four-years  
 13 time series of daily stable water isotope data in stream water and precipitation is used for the  
 14 analysis of seasonal variations. In addition, high-frequency data (10 minutely intervals) are  
 15 used for a detailed visualization of the rapid mobilization of recent water for single events. An  
 16 iterative extension of the standard two-component hydrograph separation method is applied;  
 17 this approach can be interpreted as a discretization of the catchment water and tracer mass  
 18 balance along the event and pre-event time axis. Furthermore, the calculated event, recent,  
 19 and pre-recent runoff components can be used to estimate time-varying backward travel time  
 20 distributions. For the Alp catchment and its tributaries, a nivale runoff regime can be shown,  
 21 in which the event water component generally displays its maximum (>32

**Index terms**— hydrology, runoff generation, stable isotopes, balance equations, hydrograph separation, travel time

## 25 1.1. Introduction

26 In recent years, there has been a noticeable increase in the severity of droughts in central Europe and various  
 27 other regions around the world (Balting et al. (2021), Satoh et al. (2022)). These prolonged periods of water  
 28 scarcity have had significant impacts on the natural water cycle and the ecosystems that rely on it (Trumbore et  
 29 al. (2015), Neumann et al. (2017), Senf et al. (2020)). As a result, addressing this issue requires a comprehensive  
 30 reevaluation of our approach to forest and landscape design (Liu et al. (2022), Gvein et al. (2023)), along with the  
 31 implementation of appropriate hydrological methods. Isotope-based hydrograph separations can be particularly  
 32 helpful in this regard, as they can provide insights into the travel times of water within the catchment.

Over the past few decades, the separation of storm hydrographs using stable isotope tracers has become a standard method for investigating runoff generation processes in catchment hydrology. The early pioneering work was accomplished in the late 1960s and 1970s (Pinder and Jones (1969), Dincer et al. ??1970), Martinec et al. (1974), and Fritz et al. ??1976), Sklash and Farvolden (1979)), and over the years, the methodology has been progressively expanded and adapted to address the challenges and tasks found in the field (Klaus and McDonnell (2013), Jasechko (2019)). Hoeg (2019) recently proposed a method that iteratively extends the standard two-component separation, such that  $n$  time components are separated by using a single stable isotope tracer. This approach can be used to trace the event water over a much longer period after the initial event, hence expanding the space of addressable use cases for catchment hydrologists. Hoeg (2019) applied the new method to an experimental data set of the mountainous Zastler catchment (18.4 km<sup>2</sup>, Southern Black Forest, Germany) and

43 compared the outcome with previous investigations in that area, showing the influence of antecedent moisture  
44 conditions on the event water contributions of subsequent events. This iterative extension uncovered the temporal  
45 structure of the pre-event component, and enabled a closer look at the temporal composition of the pre-event  
46 water, hence determining the extent to which recent events were involved.

47 A catchment response pattern related to antecedent moisture conditions similar to that of the Zastler catchment  
48 was found by Iorgulescu et al. (2007); they used a hydrochemical model based on a parameterization of three  
49 runoff components (direct precipitation, acid soil water, and deep groundwater) to predict conservative tracer  
50 data in the Haute-Mentue catchment (12.5 km<sup>2</sup>, Swiss Plateau). The authors concluded that the soil water  
51 component that corresponds to recent water stored in the upper soil horizons dominates catchment outflow in wet  
52 conditions but is virtually absent in dry conditions. James and Roulet (2009) formulated antecedent moisture  
53 conditions and catchment morphology as controls on the spatial patterns of runoff generation. Based on stable  
54 isotopes, they examined the spatial patterns of storm runoff generation from eight small nested forest catchments  
55 ranging in size from 0.07 to 1.5 km<sup>2</sup> (formerly the glaciated terrain of Mont Saint-Hilaire, Quebec), here as  
56 a function of antecedent moisture conditions and catchment morphology. For the storms observed under dry  
57 conditions, larger magnitudes of new water were generated from the three largest catchments attributable to  
58 basin morphology, while the storms observed under wet conditions exhibited no consistent pattern, with larger  
59 variability among the smaller catchments. The results illustrated the complexity of the influences of antecedent  
60 moisture conditions. For the pre-Alpine Erlenbach tributary (0.7 km<sup>2</sup>) Von Freyberg et al. ??2018) showed  
61 that pre-event water as a fraction of precipitation was strongly correlated with all measures of antecedent wetness  
62 but not with storm characteristics, implying that wet conditions primarily facilitate the mobilization of old (pre-  
63 event) water rather than the fast transmission of new (event) water to streamflow, even at a catchment where  
64 runoff coefficients can be large.

65 Time series of the natural isotopic composition (2H, 18O) of precipitation and streamwater can provide  
66 important insights into ecohydrological phenomena at the catchment scale. However, multi-year, high-frequency  
67 isotope datasets are generally scarce, limiting our ability to study highly dynamic short-term ecohydrological  
68 processes. Von Freyberg et al. ( ??022) recently presented a four years of daily isotope measurements in  
69 streamwater and precipitation at the Alp catchment in Switzerland and two of its tributaries. Therefore, the  
70 current study contributes here in three ways:

71 1. The classic hydrograph separation is embedded in a discretization of the catchment water and tracer mass  
72 balance along the event and preevent time axis. 2. A characteristic seasonal variation of event, recent, and  
73 pre-recent runoff components is shown for the pre-Alpine Alp catchment (46.4 km<sup>2</sup>) and two smaller tributaries  
74 (Erlenbach, 0.7 km<sup>2</sup>, and Vogelbach, 1.6 km<sup>2</sup>). 3. Single rain-runoff events of the Erlenbach catchment are  
75 analyzed in more detail to visualize and quantify the rapid mobilization of recent water.

## 76 2 II. Methods

### 77 3 a) Separation of n Time Components

78 Consider a control volume, for instance, a catchment in a river basin, with the following bulk water balance:  
79  $dS(t)/dt = J(t) - ET(t) - Q(t)$  (1)

80 where S is the time evolution of the water storage, J is the precipitation, ET is the evapotranspiration, and Q  
81 is the total stream discharge. Let C be a conservative isotope tracer with the following bulk mass balance:  
82  $d(CS(t))/dt = C J(t)J(t) - C ET(t)ET(t) - C Q(t)Q(t)$  (2)

83 where C<sub>S</sub>, C<sub>J</sub>, C<sub>ET</sub>, and C<sub>Q</sub> are tracer concentrations of the water storage S, and the volumetric flow  
84 rates J, ET, and Q. In addition, there are the time points t<sub>0</sub>, t<sub>1</sub>, ..., t<sub>n</sub> and time intervals [t<sub>0</sub>, t<sub>1</sub>],  
85 [t<sub>1</sub>, t<sub>2</sub>], ..., [t<sub>n-1</sub>, t<sub>n</sub>] that describe the start and end of n rainfall-runoff events along the time axis.  
86 Furthermore, there is a semantic time measure with the intervals e (event) and p (pre-event) that can be moved  
87 across the rainfall-runoff events, whereas the interval p is the range of all intervals just before interval e. For  
88 instance, the stream discharge Q during event e is composed of water from the current rainfall-runoff event and  
89 the prior rainfall-runoff events, such that  $Q(t) = Q_e(t) + Q_p(t)$  C<sub>Q</sub>(t)Q(t) = C<sub>e</sub>Q(t)Q<sub>e</sub>(t) + C<sub>p</sub>Q(t)Q<sub>p</sub>(t) (3)

90 where C<sub>e</sub>Q and C<sub>p</sub>Q are bulk tracer concentrations in the event and pre-event components, respectively.  
91 The same relations can be applied to the physical variables dS(t)/dt, J(t), and ET(t). Therefore, we can write  
92 generally for each volumetric flow rate V that

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96 are usually the estimated physical quantities.

97 When being applied to all physical variables dS(t)/dt, J(t), ET(t), and Q(t), the linear equation system (6)  
98 can be regarded as a discretization of the ordinary differential equation system (1) and (2) along ? +1 rainfall  
99 events. In the literature, equation (3) is the standard two-component separation model and has been used in  
100 many hydrological investigations (Klaus and McDonnell (2013)). Sklash and Farvolden (1979) and Buttle (1994)  
101 mentioned the following criteria, which also apply to the iterative separation model (6):

102 3. The pre-event component maintains a constant isotopic signature in space and time, and if not, any  
103 variations can be accounted for.

104 I would like to add another criterion (Criterion 4) that is usually implicitly considered and demands that both  
 105 the event water  $V_e$  and pre-event water  $V_p$  cannot be less than zero or larger than the total runoff  $V$  (Liu et  
 106 al. (2004)). Given the equations above, this is the case if the tracer concentration in the volumetric flow  $C_V$  is  
 107 always between that of the event water  $C_{eV}$  and preevent water  $C_{pV}$ . In the context of separation model  
 108 (6), it is required for all the backward iterations that  $C_{eV} < C_V < C_{pV}$ ?  $C_{pV} < C_V < C_{eV}$  (7)

109 Based on the measured knowns  $V$  and  $C_V$  and the estimated tracer concentrations  $C_e V, C_{e-1} V, \dots, C_{e-2} V(t) C_{e-3} V(t) - C_p V(t), \dots, 1$  (11)

111 In addition, from linear equation system (6), we obtain the following total mass balance for each volumetric  
 112 flow rate  $V : V(t) = ? \sum_{i=0}^n V e_i(t) + V p^-(t)$  (12)

113 Resolving the system of balance equations being introduced in section 2.1 for the mentioned unknowns (  $V_e$  ,  
 114  $V_{e-1}$  . . .  $V_{e-?}$  and  $V_p$  ,  $V_{p-1}$  . . .  $V_{p-?}$  ), also called end members, requires appropriate estimators  
 115 for the end member concentrations ( $C_{eV}$  ,  $C_{e-1V}$  , . . . ,  $C_{e-?V}$  and  $C_{pV}$  ,  $C_{p-1V}$  , . . . ,  $C_{p-?V}$  ). For instance, when referring to the use case of hydrograph separations, which is addressed by the volumetric  
 116 flow rate variable  $Q$ , the isotope signature  $C_{eJ}$  of precipitation  $J$  can be taken as an estimator for the tracer  
 117 concentration  $C_{eQ}$  regarding end member  $Q_e$  , whereby changes in the isotope composition of precipitation  
 118 as a result of evapotranspiration  $ET$  or changes in the water storage  $S$  (e.g. snow pack with sublimation and  
 119 re-sublimation processes) must be considered. This context becomes visible if balance equations (1) and ( 2) are  
 120 restricted to the event water fraction, such that

122 5 b) Determination of End Member Concentrations and Error  
123 Estimation

124 In a similar way, the isotope composition  $C_Q$  in the discharge  $Q$ , which occurs right before a rainfall-runoff event,  
 125 can be taken as an estimator for the tracer concentration  $C_p Q$  of the end member  $Q_p$ , whereby changes in the  
 126 isotope composition of the bulk pre-event isotope composition because of interception  $J$ , evapotranspiration  $ET$   
 127 , or changes in the water storage  $S$  (rapid mobilization of pre-event water) must be considered, which becomes  
 128 obvious, if balance equations ( 1 ) and ( 2 ) are restricted to the pre-event water  $Q_e(t) = J_e(t)Q_p(t) = J_p$   
 129  $(t) - ET_p(t) - dS_p(t)/dt C_p Q(t)Q_p(t) = C_p J(t)J_p(t) - C_p ET(t)ET_p(t) - d(C_p S(t)S_p(t))/dt$  (14)

The above equations show that the quality of the estimators  $C e_Q$ ,  $C e-1_Q$ , ...,  $C e-?_Q$  and  $C p_Q$ ,  $C$

Q can be improved by increasing the observation rate in all volume flow rates  $dS(t)/dt$ ,  $J(t)$ ,  $ET(t)$ , and  $Q(t)$ , and in fact, there are various approaches in the literature that have directly or indirectly addressed the time variant transformation of tracer mass precipitation via evapotranspiration losses or water storage changes. For instance, by solving the above set of ordinary differential equations ??Laudon et In a natural hydrological system, the accurate determination of tracer concentrations  $C_e Q$ ,  $C_e-1 Q$ ,  $\dots$ ,  $C_e-? Q$  and  $C_p Q$ ,  $C_p-1 Q$ ,  $\dots$ ,  $C_p-? Q$

138 remains a difficult task, and this is where the error estimators come into play. For the analysis of the field data,  
 139 the sensitivity of the model plus the input errors of the known variables are usually included in one measure.  
 140 For instance, Genereux (1998) and Uhlenbrook and Hoeg (2003) have demonstrated this based on analytic  
 141 expressions for the case of uncorrelated known variables and assumed uncertainties, that is, a classical Gaussian  
 142 error propagation. Others, for example, ??uczera Gaussian error propagation means that the uncertainty  $u_j$  of  
 143 the unknown variable  $y_j$  ( $j = 1..n$ ) is related to the uncertainties  $u_i$  ( $i = 1..m$ ) of the known

## 144 6 Investigating the Seasonal Variations of Event, Recent, and 145 Pre-Recent Runoff Components in a Pre-Alpine Catchment 146 using Stable Isotopes and an Iterative Hydrograph Separation 147 Approach

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151 variables x i (i = 1..m) (assumed to be independent from each other) in the following way: u j = ?y j ?x 1 u 1  
 152 2 + ?y j ?x 2 u 2 2 + .. + ?y j ?x m u m 2(15)

153 The first-order partial derivatives  $y_j \frac{\partial}{\partial x_i}$  can be collected in the following Jacobian  $n \times m$  matrix:  $J(y_1 \dots y_n) = \begin{bmatrix} \frac{\partial y_1}{\partial x_1} & \dots & \frac{\partial y_1}{\partial x_m} \\ \vdots & \ddots & \vdots \\ \frac{\partial y_n}{\partial x_1} & \dots & \frac{\partial y_n}{\partial x_m} \end{bmatrix}$  (16)

155 For instance, in case of  $\gamma = 0$  for the linear equation system ( ??) with the unknown variables  $V_e$  and  $V_p$   
 156 and known (respectively estimated) variables  $V$ ,  $C_V$ ,  $C_e V$ , and  $C_p V$ , we get the following:  
 157  $J(V_e, V_p, t) = \dots$  (17)

## 8 THE BACKWARD TRAVEL TIME DISTRIBUTION,

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When looking at the denominators of the single Jacobian entries, we can consider the uncertainties  $u_e V(t)$  and  $u_p V(t)$  as functions of  $1/(C_e V(t) - C_p V(t))$ . Beyond the uncertainties of the known variables, a high model-driven uncertainty is expected for event water isotope concentrations that are close to the corresponding pre-event water isotope concentration.

An iterative separation model (??) can be used to trace event water over a much longer period after the initial event being studied. For the obtained result, I use the term separated event water response to emphasize the fact that the traced response relates to the water of exactly one rainfall event. Conceptually, it can be related to the more commonly used term travel time distribution. Basically, it can be a time-varying approximation for this.

The travel time distribution is the response or breakthrough of an instantaneous, conservative tracer addition over the entire catchment area. It is the probability distribution that can be derived analytically based on the physical assumptions of the system under investigation. By applying a convolution integral, it can balance the tracer inputs and outputs of equation (2), as follows (Niemi (1977)): For non-stationary systems, it makes sense to distinguish between two types of probability density functions: the forward travel time distribution and backward travel time distribution (Niemi (1977)). The forward travel time distribution,  $h(t, t_{in})$ , is the probability distribution of the travel times  $t$  conditional on the injection time  $t_{in}$  of a volume flow (e.g., precipitation).  $C_Q(t) = \int_{t_{in}}^t h(t, t_{in}) dt$  (18)

## 8 The backward travel time distribution,

? $h(t, t_{in})$ , is the probability distribution of the travel times  $t$  conditional on the exit time  $t$  of a volume flow (e.g., discharge). When the system is in a steady state (constant input/output fluxes), then the forward and backward travel time distributions collapse into a single probability density function (Niemi (1977))  $h(t - t_{in}, t) Q(t) = J(t_{in}) (t_{in} - t) h(t - t_{in}, t_{in})$  (19)

where  $J(t_{in})$  is a partition function describing the fraction of rainfall  $J(t_{in})$  that ends up as runoff  $Q$ . In addition, an age function  $Q$  can be defined that describes the ratio between the number of water particles with an age in the interval  $[t, t + dt]$  sampled by  $Q$  at time  $t$  and the amount of particles with the same age stored in the control volume at that time:  $Q(t, t_{in}) = (t - t_{in}) h(t, t_{in})$  (20)

where  $h(t, t_{in})$  is the probability distribution of the residence times  $t$  of the water particles stored within the control volume at time  $t$ .

The age function  $Q(t, t_{in})$  is an interesting quantity in the sense that the tracer concentrations  $C_e Q, C_e -1 Q, \dots, C_e -k Q$  and  $C_p Q, C_p -1 Q, \dots, C_p -k Q$

$Q$  from equation system (6) can be represented as a function of the same. For instance, the pre-event concentration  $C_p Q$  for the event  $e$  can be calculated as follows:  $C_p Q(e) = e-1 - k Q(t - t_{in}, t) h(t - t_{in}, t) dt$  (21)

The basic procedure to reconstruct the event water response has already been demonstrated in Hoeg (2019), where the single components  $Q_e, Q_{e-1}, \dots, Q_{e-k}$  were arranged in the following way: Assume we are interested in the event water contribution of event 1 during events 2, 3, and 4. In this case, I can arrange one after the other, in which we have the event water  $Q_e$  of event 1, the last event water  $Q_{e-1}$  of event 2, the second-to-last event water  $Q_{e-2}$  of event 3, and the third-to-last event water  $Q_{e-3}$  of event 4, as illustrated in Figure ??.

When referring to the rainfall events  $J_1, J_2, \dots, J_{e-1}$  and the related catchment responses  $Q_1, Q_2, \dots, Q_{e-1}$ , I can define the separated event water response as follows:  $H[t_1, t_2, \dots, t_{e-1}] = \int_{t_1}^{t_{e-1}} Q_k(t) dt$  (22)

This is the volume flow of the water that entered the catchment at interval  $[t_1, t_2]$  with rainfall  $J_1$ , which appears in the stream discharge  $Q$  during interval  $[t_1, t_2]$ , here as a result of the rainfall events  $J_1, J_2, \dots, J_{e-1}$  and the related catchment responses  $Q_1, Q_2, \dots, Q_{e-1}$ .

Furthermore, I postulate that the volume-weighted function of the time-varying separated event water response  $H[t_1, t_2, \dots, t_{e-1}]$  can be considered an approximation of the time-varying backward travel time distribution,  $h(t, t_{in})$ , on the time interval  $t \in [t_1, t_2, \dots, t_{e-1}]$  when it comes to all water molecules that entered the catchment (the control volume of system (1)) at  $t_{in} \in [t_1, t_2, \dots, t_{e-1}]$   $h(t, t_{in}) = H[t_1, t_2, \dots, t_{e-1}] / (t - t_{in}) dt$  (23)

, respectively.  $h(t, t_{in}) = H[t_1, t_2, \dots, t_{e-1}] / (t - t_{in}) dt$  (24)

regarding the semantic time intervals  $e, e+1, \dots, e+k$  on  $k$  backward iterations. ??018) mention that the Alp tributaries are wet throughout most of the year. This is due to the high clay content, the low drainable porosity and shallow soils. The water table is generally close to the soil surface, especially in hollows and flatter areas, where the hydraulic gradient is low or at the bottom of hillslopes because of the large amount of water coming from upslope areas. Surface soil moisture measurements show that soil moisture is lowest in the forested ridge sites and highest in the flatter meadow and wetland sites. Total annual precipitation in the Alp valley is strongly controlled by elevation, averaging 1791 mm/year in the flat northern part near the outlet, and roughly 30% more in the mountainous headwaters of the catchment (2300 mm/year). Snowfall comprises up to one-third of the total precipitation in the headwaters of the Alp, although snowfall is frequently interrupted by rainfall during mild periods in winter, with a corresponding occurrence of rain-on-snow events (Rücker et al. (??019)).

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## 217 9 Investigating the Seasonal

218 The ratio of current to potential evapotranspiration (ET<sub>a</sub>/ET<sub>p</sub> ratio) serves as a valuable indicator of water  
219 availability for plants. When this ratio falls below 0.8, it suggests an increased likelihood of drought-related  
220 impairments (Allgaier Leuch et al. ( ??017)). In the Alp catchment, we can expect ET<sub>a</sub>/ET<sub>p</sub> values to range  
221 from 0.61 to 0.9 in the valley bottoms, and from 0.81 to 1.00 in the upslope areas. These figures are based on  
222 the long-term average spanning from 1981 to 2010.

223 In the present study, a four-years time series (June 2015-May 2019) of daily stable water isotope data in  
224 stream water and precipitation is used for the analysis of seasonal variations. The data set is described by Von  
225 Freyberg et al. ( ??022), who provided detailed information on the precipitation and streamwater sampling,  
226 sample handling and isotope analysis. Streamwater isotopes were measured in the Alp main stream and in two of  
227 its tributaries (Erlenbach and Vogelbach). Precipitation isotopes were measured at two grassland locations in the  
228 Alp catchment: in the headwaters at 1228 m a.s.l. and near the outlet at 910 m a.s.l. The data set also includes  
229 the daily time series of key hydrologic and meteorologic variables, such as daily streamwater and precipitation  
230 fluxes, air temperature, relative humidity, and snow depth.

231 To better classify single rainfall runoff events and seasonal relationships, the hydrological data were  
232 supplemented with meteorological data (e.g., amount of snowfall, evapotranspiration, soil moisture) from ERA5.  
233 The reanalysis product ERA5 has recently been released by European Centre for Medium-Range Weather  
234 Forecasts (ECMWF) as part of Copernicus Climate Change Services (Hersbach et al. (2019)). This product  
235 covers the period from 1979 to present.

## 236 10 3.

237 4.

## 238 11 5.

239 Q The calculated event, recent, and pre-recent runoff components can be used to estimate time-varying backward  
240 travel time distributions. In the following, I demonstrate this on the basis of a simple example. Five rainfall  
241 runoff events, taken from a field study in the Erlenbach catchment between September 2016 and October 2017  
242 (Von Freyberg et al. ( ??018)), serve as the examples. The analysis is based on the separation model ( ??)  
243 applying three backward iterations ( ? = 3). To determine the end member concentrations ??3) and ( 14) are  
244 not explicitly solved; instead, the pre-event water concentrations are taken directly from the measured isotope  
245 compositions in the discharge right before the hydrograph rises. The event water concentrations refer to the  
246 volume-weighted isotope composition in the respective precipitation event.  $Q_e$  no  $C_e$   $Q$  ,  $C_{e-1}$   $Q$  , . . . ,  $C_{e-3}$   
247  $Q$  and  $C_{pQ}$  ,  $C_{p-1}Q$  , . . . ,  $C_{p-3}Q$  , differential equations (

248 For calculating the Gaussian standard errors, an uncertainty at the scale of the discharge measurements in the  
249 field,  $u(Q) = 0.001$  [m<sup>3</sup> /s], and of the isotope analysis in the laboratory is adopted, that is,  $u(C_Q) = 0.09$  [?],  
250  $u(C_{eQ}) = 0.09$  [?],  $u(C_{pQ}) = 0.09$  [?]

251 , and continuously added for each backward iteration: ??018) reported that pre-event water is more efficiently  
252 mobilized under wetter conditions, showing  $u(C_{e-1}Q) = 0.18$  [?],  $u(C_{p-1}Q) = 0.18$  [?],  $u(C_{e-2}Q) = 0.27$   
253 [?],  $u(C_{p-2}Q) = 0.27$  [?],  $u(C_{e-3}Q) = 0.36$  [?], and  $u(C_{p-3}Q) = 0.36$  [?].

254 Figure ?? shows the separated event water response according to equation (22) and Figure ?? for five rainfall  
255 runoff events between September 19, 2017 and October 21, 2017. The volume-weighted version of this time-  
256 varying response can be considered an approximation of the time-varying backward travel time distribution, as  
257 shown in equation (23). In addition, the so-called rapid mobilization of pre-event water can be detected. For  
258 instance, in the that the rapid activation of the pre-event water at Erlenbach (even during small storms) can be  
259 explained by generally shallow perched groundwater tables in the aquifer overlying the low permeability bedrock.  
260 The median Gaussian standard error for the calculated event, recent, and pre-recent runoff components are  $u(Q_e) = 81.9\% / 0.01$  mm 10min ,  $u(Q_{e-1}) = 123.4\% / 0.10$  mm 10min ,  $u(Q_{e-2}) = 205.2\% / 0.18$  mm 10min  
261 ,  $u(Q_{e-3}) = 231.6\% / 0.07$  mm 10min , and  $u(Q_{p-3}) = 32.3\% / 0.18$  mm 10min . For better illustration,  
262 Figure 8 shows the separated event water response for a single rainfall runoff event (number 3) together with the  
263 Gaussian standard error bounds according to equation (15).  
264

## 265 12 Alp

266 In the following, an analysis of the runoff generation processes in terms of event  $Q_e$  , recent  $Q_{r-3}$  , and  
267 pre-recent  $Q_{p-3}$  runoff components is demonstrated for the pre-Alpine Alp catchment (46.4 km<sup>2</sup> ) and two  
268 smaller tributaries (Erlenbach, 0.7 km<sup>2</sup> , and Vogelbach, 1.6 km<sup>2</sup> ), whereas recent water is understood as the  
269 contribution of the respective three prior rainfall-runoff events, that is To compensate for altitude effects on the  
270 catchment scale, the daily isotope concentrations in precipitation considers the available isotopic samples of the  
271 two measuring stations and are area weighted based on to the hypsometric curve of the Alp catchment. The  
272 analysis is based on the separation model (6) that applies three backward iterations ( ? = 3), whereas the entire  
273 data set is being processed; that is, 147 rainfall runoff events are being investigated for the Alp catchment, 113  
274 rainfall runoff events for the Erlenbach catchment, and 120 rainfall runoff events for the Vogelbach catchment.  
275 Again, the end member concentrations Alp discharge and precipitation. In addition, where necessary, the end

## 13 V. CONCLUSIONS

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276 member concentrations are partially adjusted to ensure that Criterion 4 (equation ( 7)) is continuously fulfilled.  
277 This approach does not reduce or increase the size of the relative error, but it does stabilize the solution overall  
278 (Hoeg (2021)). $Q_{r-3} = Q_{e-1} + Q_{e-2} + Q_{e-3}$ (25) $C_e Q, C_{e-1} Q, \dots, C_{e-3} Q$  and  $C_{p-1} Q, C_{p-2} Q, C_{p-3} Q, \dots$

279 Although the absolute Gaussian standard error bounds are moderately low with values between 0.09 mm/d  
280 and 0.61 mm/d (see Table 1), the relative errors associated with this hydrograph separation are relatively  
281 high, with values between 38.5% and 622.4%. Therefore, the results may not lead to accurate quantitative  
282 conclusions. Nevertheless, qualitative statements about the seasonal runoff formation in the Alpine catchment  
283 area and its tributaries Erlenbach and Vogelbach are possible, as I will show below. To visualize the seasonal  
284 variations, I calculate the Pardé coefficients for each runoff component, which is the quotient of the long-term  
285 (four years) average monthly discharge and long-term (four years) average annual discharge. The precipitation  
286 and evapotranspiration regimes are added to better evaluate and classify the monthly and seasonal changes of the  
287 runoff components, which are: stream discharge  $Q$ , event water  $Q_e$ , recent water  $Q_{r-3}$ , and pre-recent water  
288  $Q_{p-3}$ . Evapotranspiration data are taken from the reanalysis product ERA5 of the Copernicus Climate Change  
289 Service. For the Alp catchment and its tributaries, a nivale runoff and precipitation regime can [-] Alp be shown  
290 in Figure 9. In relation to the stream discharge  $Q$ , the event water component  $Q_e$  has its maximum (>32%)  
291 during August in general, as shown in Figure 10 and Figure 11. The highest recent water fractions  $Q_{r-3}$  (50-59%)  
292 can be expected at the beginning of winter (December, January), whereas the lowest fractions (7-14%) are found  
293 at the end of autumn (November). In return, higher pre-recent water fractions  $Q_{p-3}$  can be found in the summer  
294 (July, 60-82%) and in the middle of autumn (October, 64-74%). The two tributaries expose an additional peak  
295 of pre-recent water in the snowmelt season (April, 47-62%), during which the lowest event water fractions (6-8%)  
296 can be expected. For the Alp catchment, the event water component (29-32%) and recent water component  
297 (56%) clearly exceed the pre-recent component (17-21%) in August and September after the month July, which  
298 is characterized by relatively low precipitation, high evapotranspiration, and low soil moisture. A sensitivity  
299 analysis regarding the evapotranspiration rates ET from ERA5 confirms the calculated seasonal variations of  
300 the Pardé coefficients both for the Alp catchment and its tributaries, Erlenbach and Vogelbach. The analysis,  
301 shown in Figure 12, indicates a shift in proportions towards Added monthly Pardé coefficients (1: January -12:  
302 December) of event water  $Q_e$ , recent water  $Q_{r-3} = Q_{e-1} + Q_{e-2} + Q_{e-3}$ , and pre-recent water  $Q_{p-3}$  for  
303 the Erlenbach, Vogelbach, and Alp catchments in relation to the monthly Pardé coefficients of stream discharge  
304 ( $Q$ ). Monthly Pardé coefficients (1: January -12: December) of precipitation ( $J$ ), and evapotranspiration (ET),  
305 based on the four-year investigation between June 2015 and May 2019, and an iterative extension of the standard  
306 two-component hydrograph separation method. Sensitivity analysis to assess the impact of ET on the daily end  
307 member concentrations ( $C_e Q, C_{e-1} Q, \dots, C_{e-3} Q$ ). This analysis involves considering a rate factor  $f_{ET}$ ,  
308 which can take values of 0.25 or 0.5. To first-order approximate the daily end member concentrations, the  
309 daily precipitation rates ( $J$ ) are directly reduced by 25% or 50% of the daily evapotranspiration rates, similar  
310 to equation (13). The evapotranspiration data used in this analysis are obtained from the reanalysis product  
311 ERA5. ??022)) is unlikely to lead to a greater mobilization and availability of pre-recent runoff components  
312 in this area. Consequently, the Median of the relative and absolute Gaussian standard errors (in % and mm d  
313 ) for event water  $Q_e$ , recent water  $Q_{r-3}$ , and pre-recent water  $Q_{p-3}$  in the Erlenbach, Vogelbach and Alp  
314 catchment between June 2015 and May 2019, here based on an iterative extension of the standard two-component  
315 hydrograph separation method with the uncertainties of stream discharge,  $u(Q) = 0.001$  [m<sup>3</sup> /s], and of the  
316 isotope analysis  $u(C_e Q) = 0.09$  [?],  $u(C_{e-1} Q) = 0.09$  [?],  $u(C_{e-2} Q) = 0.27$  [?],  $u(C_{e-3} Q) = 0$ .  
317

## 318 13 V. Conclusions

319 In the present study, I have shown that the classic two-component hydrograph separation can be iteratively  
320 embedded in a discretization of the catchment water and tracer mass balance along the event and pre-event  
321 time axis. With this method, it is possible, for instance, to analyze and quantify the rapid mobilization of  
322 recent water for single (high-frequency measured) rainfall-runoff events, and to estimate time-varying backward  
323 travel time distributions. When applied to longer time series of daily stable water isotope data in stream water  
324 and precipitation, the method can be used to analyze seasonal variations of event, recent, and pre-recent runoff  
325 components.

326 In relation to the Alp catchment, the relatively high fractions of event water and recent water in stream  
327 discharge during August and September represents a rather unexpected result that certainly requires further  
328 investigation, but shows that fundamental assumptions, used for instance in runoff recession analysis, need to be  
329 questioned and that the sensitivity of the catchment water balance in response to drought situations could be  
330 greater than expected.

331 The author would like to thank Jana von Freyberg (ETH -Swiss Federal Institute of Technology Zurich,  
332 Department of Environmental Systems Science) for providing the additional excerpt of the high-frequency  
333 measurement series from the Erlenbach catchment. My special thanks goes to chief editor Marie V. Carlsen,  
334 assistant editor Marian C. Miller, and two anonymous reviewers, who contributed to this work with numerous  
335 improvement proposals. potential to incorporate this approach into hydrological models to improve their accuracy  
336 and predictive capabilities. Moreover, there are opportunities to apply the method in more complex and detailed  
337 ways, potentially uncovering new insights in catchment hydrology. This could include, for instance, more detailed



Figure 1:

342

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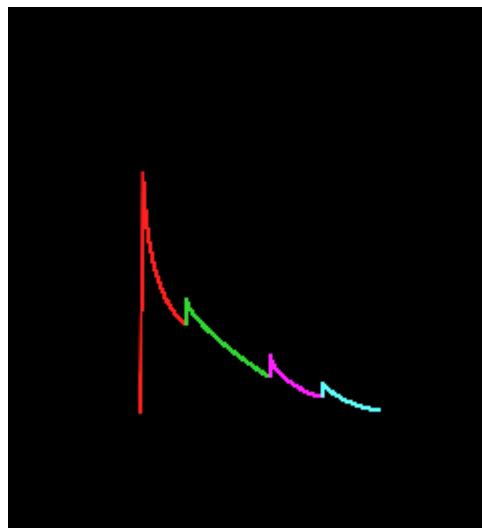
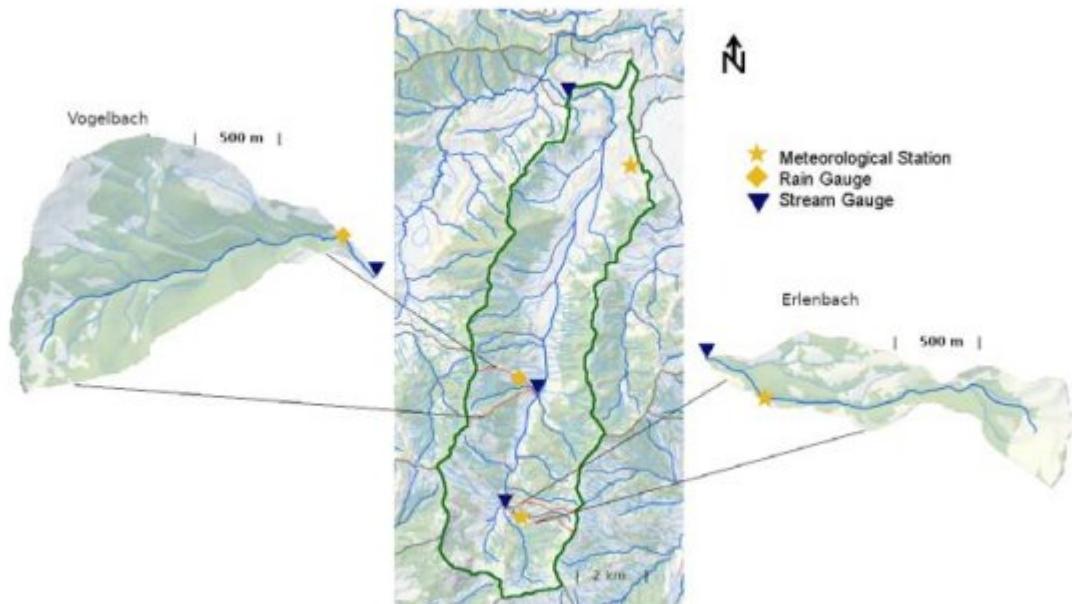


Figure 2:



712

Figure 3: 7 ©Figure 1 :Figure 2 :

$$C_e Q_e(t) = C_e J_e(t) - C_e ET_e(t) - d(C_e S_e(t)) / dt \quad (13)$$

fraction, such that

Figure 4:

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Year 2023

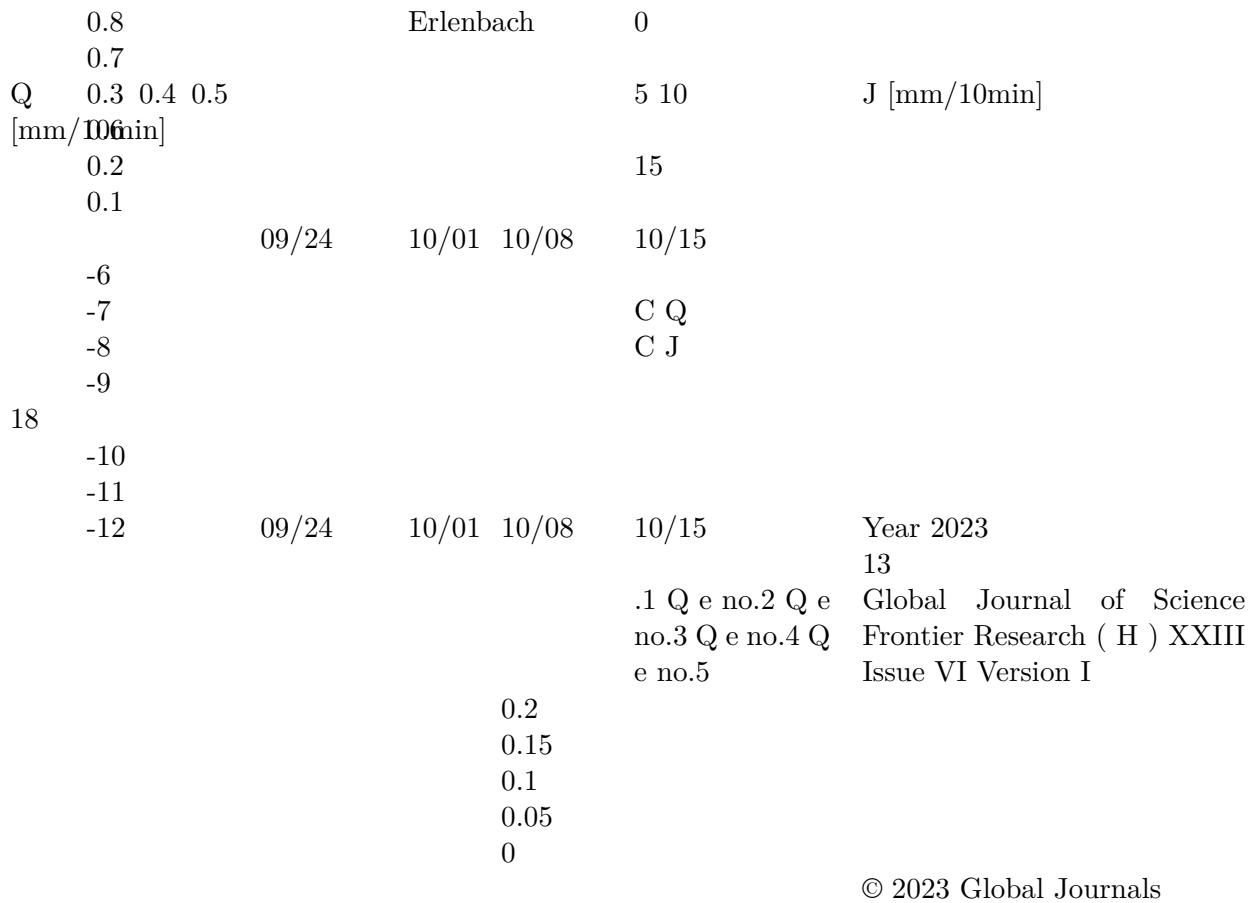
12

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Figure 5: Investigating the Seasonal Variations of Event, Recent, and Pre-Recent Runoff Components in a Pre-Alpine Catchment using Stable Isotopes and an Iterative Hydrograph Separation Approach

#### IV. Results and Discussion



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Figure 7: Investigating the Seasonal Variations of Event, Recent, and Pre-Recent Runoff Components in a Pre-Alpine Catchment using Stable Isotopes and an Iterative Hydrograph Separation Approach

1

36 [?], and  $u(C p-3 Q) = 0.36$  [?].

(Scherler et al. (2016)).

Figure 8: Table 1 :

? ? ? 10. Appendix A.2. Greek Symbols partition function backward iteration travel time ? age function

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Figure 9: 1) Investigating the Seasonal Variations of Event, Recent, and Pre-Recent Runoff Components in a Pre-Alpine Catchment using Stable Isotopes and an Iterative Hydrograph Separation Approach

### 343 .1 Acknowledgements

### 344 .2 Appendix A.1. Latin Symbols

345 The data used for this study can be retrieved via Von Freyberg et al. ??2018)

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