

Editor Decision

Thank you for your detailed replies to the the comments of the 2 reviewers. As you have acknowledged, both reviewers raised a couple of interesting points, a few of which are of considerable importance for the underlying interpretation of your results. I very much appreciate that you invested some time to address most comments in-depths. Yet, I feel that some points need a bit more attention in order to clarify your approach for the reader and to avoid misinterpretations:

We appreciated the comments of the Editor and think we could benefit from them. We are sure that they helped us to significantly improve our manuscript. In the following, we present these comments as well as our point-by-point response to all of them.

1. relating to comments of both reviewers, it remains unclear *how* pQ is constructed in your study. More specifically: which pQ does the reader actually get to see? Is it the age distribution of an incoming signal J that leaves a grid cell through *all* exit routes (i.e. Q and ET)? or is it only the age distribution of water leaving the unsaturated zone (Q and ET?) as I seem to understand from your reply No.8 to reviewer 2? In case groundwater storage was considered, how was the hydrologically passive storage accounted for (see e.g. the well known Figure 1 in Zuber, 1986, Journal of Hydrology 86) - this is highly relevant for the age distributions as it exacerbates the dichotomy between celerity and velocity (McDonnell and Beven, 2014)! In addition, as you showed, mHM uses several soil layers. Is pQ a direct reflection of the total outflow from *all* soil layers? A further point is that only 5 years of warm up period were used. As the subsequently used "mean" age can be considerably sensitive to long tails, what is the potential of your estimates being biased towards too young water? In other words, even with 50 years modelling, is it conceivable that we miss some minor volumes of very old water here? These points need to be made much clearer - in the methods but also in results and discussion sections (particularly in the figures!).

This first comment contains several distinct sub-comments. To address them appropriately, we will answer them individually.

- (a) How is pQ determined.

We are sorry for this misunderstanding. Due to the Editor's as well as the reviewers' comments, we are now aware that this crucial aspect was not properly explained in the earlier version of the manuscript. In the revised manuscript, we address these concerns by elaborating on the question what storages and fluxes were used for the analysis. It is now made clear in the Introduction, the Methods section as well as throughout the Results section (plus figure captions) that all our analysis was confined to the soil layer, only. In particular, we present the rationals for this decision both in the Introduction and further elaborate on its ramifications in the Methods section.

- (b) What about the soil layers.

The Editor is correct in his interpretation: we estimated the age distribution (TDDs) of water leaving the unsaturated zone (Q and ET). The groundwater part is not accounted in the analysis (see our elaborations above).

- (c) Warm-up period too short.

The problem of a warm-up period that might be too short is only relevant for backward TTDs. Most of the analysis in our study was performed with respect to forward TTDs. In this case a similar problem exists with respect to a period of time at the end of the estimation period that may have to be discarded. We determined the length of this period by using the partitioning function θ as a measure. This quantity describes the ratio of a water parcel that is entering the CV at a given time and eventually leaves as discharge. Adding this value with the ratio of the water parcel that leaves as ET should always add up to one. We could therefore add up these quantities to determine how much of the time series we could use. Our investigation showed that only close to the last two years needed to be discarded from the analysis. In the revised version of the manuscript, we acknowledge this notion (see beginning of the Results and Discussion section in the revised manuscript).

2. Not everybody may be familiar with the details of mHM and of how the parameters are obtained in that model. It may therefore be important to clarify much earlier in the manuscript,

how the soil moisture storage capacity is determined. Attentive readers will otherwise wonder if some of the storage is unaccounted for if the value is a "normal" calibration parameter (i.e. potential "passive" storage" that within you period of application does not become hydrologically active but may provide a mixing volume; see above and McDonnell and Beven, 2014).

With respect to the way the soil moisture storage is determined, we now address this issue early on in the manuscript (see the Introduction in the revised manuscript) and later we elaborate on it in the Methods section. We also expanded our explanation which parts of mHM are parameters determined during calibration and which are determined as model outputs (see the Methods section in the revised manuscript). In addition, we refer to Zacharias 2007, where important aspects of the calibration process are described in more detail. Finally, we discuss the difference between the water content relevant for outflow generation and for travel-time behavior in the Conclusion section of the revised manuscript.

3. I am missing a clear research question and/or research hypothesis this manuscript is looking at. Please add that at the end of the introduction.

We agree that the manuscript in its previous form did not state concisly the main reserach questions and novel contributions of the study. From our current perspective, we would summarize the main questions as follows: (i) How are spatially distributed quantities, in particular land-cover, precipitation and soil type, impact travel-time behavior, (ii) how do different hydrological regimes impact travel-time behavior and (iii) what is the inter-connection between travel-time behavior and specific conceptualization of different hydrological processes and how may these connections be used for a better model calibration. These points are now better stated in the manuscript (see Introduction in the revised manuscript).

4. Note that figure captions should be stand-alone. In other words, the reader should be able to understand the figure by reading the captions alone. At the present the captions a very vague and uninformative. I would encourage you to be more specific and detailed. Only one example: what are the soil classes 9, 38, etc. in figure 14. what type of soils are we talking about?

We heeded the advise of the Editor and strongly revised the figures caption in the manuscript to make them more self explanatory (see figure captions in the revised manuscript).

In general, I would be glad if you could invest some more time in developing a more detailed and clear description of your methods and a stronger discussion of the limitations of the chosen approach (e.g. what about parameter uncertainty? uncertainty in age distributions? etc. and the effect on the interpretation)

We thank the Editor for the for these suggestions. In the revised manuscript, we have now added more material to address these two main points. First, with regard to the methods, we have significantly expanded our description on how the states and fluxes were generated in mHM and how the TTDs were computed using these states and fluxes. This point was already addressed above. Second, we also added material to the Conclusions where we now critically assess the limitations of our approach. In particular, we discuss the different sources of uncertainty in travel-time estimation. These include uncertainties arising due to input data, simplified model structure and parametric uncertainty. With respect to input data, we refer to prior studies on the validity of the mHM input data. With respect to mHM model structure, we discuss the potential impact of our definition of soil moisture. However, to fully assess this aspect, a model-to-model comparison would be necessary. We are interested to pursue such a comparison with different candidate models being considered. The last point is parameteric uncertainty. Here we refer to the literature that exists on this topic with respect to mHM and discuss the main results from these studies and how they may relate to the estimation of TTDs. This last point is particularly interesting and a full study may be appropriate. These points are all discussed in the Conclusions section of the revised version of the manuscript.

Spatially Distributed Characterization of Soil-Moisture Dynamics Using Travel-Time Distributions

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Abstract.

Travel-time distributions are a comprehensive tool for the characterization of hydrological system dynamics. Unlike streamflow hydrographs, they describe the movement and storage of water inside and through the hydrological system. Until recently, studies using such travel-time distributions have generally either been applied to simple (artificial toy) models or to real-world catchments using available time series, e.g., stable isotopes. Whereas the former are limited in their realism, the latter are limited in their use of available data sets. In our study, we employ a middle ground by using the mesoscale Hydrological Model (mHM) and apply it to a catchment in Central Germany. Being able to draw on multiple large data sets for calibration and verification, we generate a large array of spatially distributed states and fluxes. These hydrological outputs are then used to compute the travel-time distributions for every grid cell in the modeling domain. A statistical analysis shows the general soundness of the upscaling scheme employed in mHM and reveal precipitation, saturated soil moisture and potential evapotranspiration as important predictors for explaining the spatial heterogeneity of mean travel times. In addition, we demonstrate and discuss the high information content of mean travel times for characterization of internal hydrological processes.

Keywords

Travel-time distributions, mesoscale Hydrological Model (mHM), multiscale modeling, catchment hydrology, stochastic hydrology, model calibration

1 Introduction

The description of storage and transport of both water and dissolved contaminants in catchments is a challenging subject due to the high heterogeneity of the subsurface properties that govern their fate (Dagan, 1989). This heterogeneity, combined with a limited knowledge about the subsurface, results in high degrees of uncertainty. As a result, stochastic methods are often applied, where the relevant processes are modeled as being random (Dagan, 1986; Rubin, 2003). Amongst these methods, a powerful tool is the use of travel-time distributions (TTDs), where storage and transport inside the catchment are modeled from a Lagrangian perspective (Rinaldo and Marani, 1987; Rinaldo et al., 1989). This means that the catchment itself

or meaningful parts of it is treated as a control volume (CV). The spatially complex array of different flow paths inside such a CV is consequently ignored and only inlet and outlet fluxes are used for the analysis (Botter *et al.*, 2010; Rinaldo *et al.*, 2011; Botter, 2012). This observation-based description of catchment dynamics makes ~~FFDs~~-TTD's a very robust tool. Although the application of ~~FFDs~~-TTD's goes back many decades (Danckwerts, 1953; Niemi, 1977), recent developments have strongly improved their theoretical foundations turning them into a versatile and coherent tool to characterize catchment dynamics (Bertuzzo *et al.*, 2013; Benettin *et al.*, 2015a; Rinaldo *et al.*, 2015; Porporato and Calabrese, 2015). Owing to this progress, McMillan *et al.* (2012) and McDonnell and Beven (2014) have opined that ~~FFDs~~-TTD's should be used routinely for hydrological model calibration, a notion that has been picked up with tremendous speed (Windhorst *et al.*, 2014; Vereecken *et al.*, 2015; McGuire and McDonnell, 2015). Independently but somewhat parallel to that, Kitanidis (2015) has recently pointed out, that the key to subsurface characterization is to use all available information. From this information-centered perspective, using ~~FFDs~~-TTD's have several advantages. First, the travel-time behavior is controlled by different factors than the hydrograph response. Whereas the latter is relating rainfall–runoff events the former is relating rainfall–runoff water (McDonnell and Beven, 2014; Birkel and Soulsby, 2015). Second, spatially distributed tracer experiments may dramatically increase the information content available for catchment characterization (Birkel and Soulsby, 2015).

This range of advantages have lead to a steady increase in both applied and theoretical studies using ~~FFDs~~-TTD's for the description of catchment dynamics. Applied studies here means that data from real-world sites are used (McGuire *et al.*, 2005; Cardenas, 2007; Broxton *et al.*, 2009; Tetzlaff *et al.*, 2011; Dunn *et al.*, 2012; Hrachowitz *et al.*, 2013, 2015; Harman, 2015). Here the advantage is that the data used for the analysis do not suffer from model errors or other conceptual limitations. However, such data are generally limited in amount (e.g., tracer or isotope time series limited to a few years only, although Hrachowitz *et al.* (2009) used time series of up to 17 years) and variety (only a limited number of data types are available). As a result, such studies might fail to find long-term trends, establish connections between travel-time behavior and specific catchment properties or to investigate the impact of certain hydraulic regimes that are only rarely occurring (e.g., drought or extremely rainy months). In the second category, we find theoretical studies, that either use a very simplified computational model to focus on specific questions (Rinaldo *et al.*, 2006; Duffy, 2010; Botter *et al.*, 2010; van der Velde *et al.*, 2012; Benettin *et al.*, 2015a; Porporato and Calabrese, 2015) or employ more realistic hydrological models that provide a large data set typically not available in real-world sites Sayama and McDonnell (2009); Fenicia *et al.* (2010); McMillan *et al.* (2012). Such theoretical studies allow a more thorough and detailed analysis of the involved processes and their interdependence but may suffer from an oversimplified model setup for in- and outflux generation.

Our study falls into the latter category such that we use a hydrological model, i.e., the mesoscale Hydrological Model (mHM) (Samaniago *et al.*, 2010a; Kumar *et al.*, 2013a), to generate the fluxes and states for the analysis. Using detailed data of precipitation, land cover, morphology and soil type as inputs, mHM is able to provide continuous simulations of spatially distributed fluxes (e.g., groundwater recharge or evapotranspiration) and states (e.g., soil moisture) as outputs. By employing mHM, which is a spatially-distributed hydrological model, we are, however, able to extend these prior studies to a spatially-distributed travel-time analysis. This makes it possible to address several types of investigation. First, it allows for a comprehensive description of the flow and transport dynamics taking place in the catchment. The spatial distribution

of such dynamics can then be related to e.g., land cover and physical properties of the soil as well as to driving forces like precipitation to determine dominant predictive factors. In addition, it allows to investigate how certain parametrizations of the mHM model are related to the travel-time behavior of the catchment. This opens the way for a more robust model calibration of hydrological models using additional datasets (*McDonnell and Beven, 2014; Birkel and Soulsby, 2015; Kitanidis,*
5 2015).

As a case study, we use a ca. 1000 km² catchment in Central Germany for which detailed morphological and climatological data are available to parametrize mHM. In addition, the chosen catchment is the location of the Hainich Critical Zone Exploratory, a comprehensive monitoring network used within the Collaborative Research Center AquaDiva (*Küsel et al., 2016*). AquaDiva seeks to elucidate the critical role of water fluxes connecting surface conditions with biogeochemical functions in
10 the subsurface. One of the goals of this project is to understand how far signal of surface properties, like land cover or land management, can be traced into the subsurface water and solute dynamics. Spatially explicit travel-time distributions are the perfect analytical tool to investigate such questions.

By virtue of using the modeled data from mHM, we are able to address several questions that have not been investigated before. First, how are spatially-distributed quantities, in particular land-cover, precipitation and soil type, impacting travel-time
15 behavior in the soil? Unlike earlier model-based studies, mHM is a spatially-distributed hydrological model. We can therefore add to prior knowledge by investigating the travel-time behavior for every mHM grid cell and relate it to its geo-physical and climatic properties. Next, how do different hydrological regimes (wet vs dry) impact travel-time behavior in the soil? Here we investigate the impact of changing external conditions (meteorological factors) using the long time-series of modeled fluxes and states. Finally, what is the inter-connection between travel-time behavior and specific conceptualization of different
20 hydrological processes, and how may these connections be used for further improvement of model parameterization? Investigating the impact of model-specific conceptualizations on the predicted travel-time behavior can provide a better understanding on how actual measurement may be connected to certain model parameters. For the quantitative analysis, we focus on soil moisture, only, i.e., we exclude groundwater. This was necessary, due to the implementation of groundwater in mHM as a linear reservoir. Although variations, i.e., fluxes, of the groundwater level can be represented well (*Rakovec et al., 2016*) the total amount remains elusive. This is a common feature of hydrological models (*Fan, 2015*) and mHM is no exception. Furthermore, we consider this restriction to be acceptable within the scope of our study, i.e., elucidation of the spatio-temporal dynamics of TTD's. Groundwater by definition is far less impacted by the spatial distribution of precipitation or land cover. In addition, *Benettin et al. (2015b)* recently showed that TTD's show little temporal variability compared to soil moisture.

To present our results on such questions, the rest of the paper is organized as follows: In Section 2 we describe the numerical
30 and analytical tools used in this study. Thus comprises the framework of travel-time distributions, as applied in this study, as well as the relevant features of mHM. In Section 3, we present the results of our study and demonstrate how they relate to the questions raised above. Finally, in Section 4 we summarize our main findings in light these questions and draw some conclusions.

2 Methods

In the following, we provide a short overview of the analytical and numerical tools and methods used in this study. We start by introducing the concept of travel-time distributions. In the following, we use the nomenclature as given by *Benettin et al.* (2015a) and the theoretical framework by *Botter et al.* (2010). In addition to that, we give a short overview of the numerical
 5 model (mHM) which was used for the calculation of the states and fluxes. Finally, we introduce the catchment used in our study.

2.1 Travel-time distributions for a single control volume

Travel-time distributions are a stochastic description of the dynamic of a water parcel moving through a given control volume (CV). The definition of such a control volume for real-world situation is often arbitrary to some extent (see e.g., the schematic
 10 in Figure 1). Within the context of this study, we used a spatially distributed model where the catchment is partitioned in regular grid cells (for more details see Section 2.2 below). Consequently, the boundaries of our CV were given by the grid cells of the model.

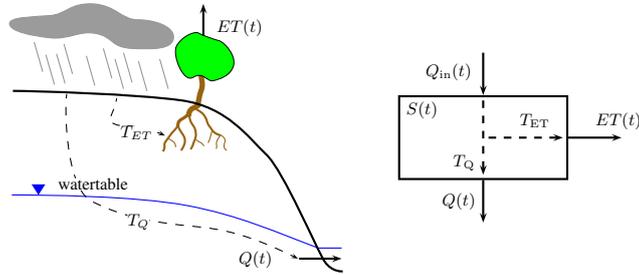


Figure 1. Water movement inside a hill slope (physical schematic on the left and conceptual schematic on the right).

Given that such a CV can be reasonably defined, it is clear that the dynamics of a water parcel is determined by the in- and out-fluxes, that are changing the water content inside it. The time evolution of the water content S inside such a CV is
 15 then given by the following balance equation

$$\frac{d}{dt}S(t) = Q_{in}(t) - Q_{out}(t) = J(t) - (ET(t) + Q(t)). \quad (1)$$

Equation (1) is a simple initial-value problem with the in-flux $Q_{in}(t)$ given by the effective precipitation $J(t)$ whereas the out-flux $Q_{out}(t)$ is given by evapotranspiration $ET(t)$ and runoff per grid cell $Q(t)$.

To denote the different times involved in the dynamic of a water parcel, we followed the notation of *Benettin et al.* (2015a).
 20 Chronological time was accordingly denoted with t , whereas the water parcel entered the CV at t_{in} and left at t_{ex} . At any given time t' in between these two points, any water parcel can therefore be characterized by two different properties; its age T_A as well as its (remaining) life expectancy T_E (see Figure 2).

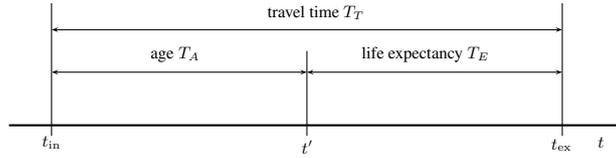


Figure 2. Interpretation Schematic of different times associated with the travel-time dynamics of a water parcel. Age T_A is the time elapsed at t' since the water parcel entered the CV at t_{in} , whereas life expectancy T_E is the time remaining at t' until the water parcel leaves at t_{ex} .

In their paper, Benettin *et al.* (2015a) emphasize the two interpretations that originate from these two points of view. Age is a backwards concept referring to the time passed since the beginning. The associated travel time distribution is therefore called the backward TTD. The concept of backward ~~TTDs~~ TTD's is of particular interest for the characterization of e.g., a water sample, since its composition is determined by the age of the water in the CV. Life expectancy, on the other hand, is a forward concept since it is referring to the time still left until exit from the CV. The associated travel time distribution is therefore called the forward TTD. Such forward ~~TTDs~~ TTD's are relevant e.g., for tracer test, since the concentration of an ideal tracer at the outlet is given by the TTD of its associated water parcel.

In order to derive the ~~TTDs~~ TTD's associated with the forward and backward formulation, Botter *et al.* (2011) presented a derivation using only the states and fluxes inside the CV as well as what they call an age function (for more information on their derivation we also refer to Botter *et al.* (2010) and the references therein). In the following, we assume a uniform age function only. This means that the age distribution of the water leaving the CV is the same as the age distribution of the water inside the CV, i.e., no age preference of the outflow generating processes (discharge and ET) exists. This decision became necessary, since we could not yet draw on any data for the age distribution of water at the outlet of the catchment. As a result, we were not able to compare the predictions of different age functions to any measurements and therefore determine the most adequate description. In absence of such data, the most appropriate choice is the one involving the least amount of information, which is given by the assumption of uniform sampling. Using this assumption, we can state the following for the forward formulation

$$\vec{p}_Q(T_E, t_{in}) = \frac{Q(t)}{\theta(t_{in})S(t)} \exp\left(-\int_{T_E}^t \frac{Q(t') + ET(t')}{S(t')} dt'\right) \quad (2)$$

with $T_E = t - t_{in}$, $t > t_{in}$, i.e., the time from the moment the water parcel entered the reservoir until now. The function θ in Equation (2) is called the partition function (Botter *et al.*, 2010, 2011) and can be derived using the following formula

$$\theta(t_{in}) = \int_{t_{in}}^{\infty} \frac{Q(t)}{S(t)} \exp\left(-\int_{T_E}^t \frac{Q(t') + ET(t')}{S(t')} dt'\right) dt. \quad (3)$$

This partition function describes the portion of the water parcel, entering the CV at t_{in} , that is contributing eventually to discharge as opposed to evapotranspiration. It is consequently a dimensionless number between 0 and 1.

For the backward formulation, we can state the following

$$\overleftarrow{p}_Q(T_A, t_{ex}) = \frac{J(t)}{S(t)} \exp \left(- \int_{T_A} \frac{Q(t') + ET(t')}{S(t')} dt' \right) \quad (4)$$

5 with $T_A = t_{ex} - t$, $t < t_{ex}$, i.e., the time from now until the moment the water parcel leaves the reservoir.

Both these formulations determine the travel time of the water leaving as discharge. The ~~TTD~~ TTD's for the water leaving as evapotranspiration can be determined in an analogous way.

2.2 Numerical model

We used a spatially distributed, grid-based mesoscale Hydrological Model (mHM; *Samaniego et al. (2010a); Kumar et al. (2013a)*) to generate the states and fluxes needed for the TDD analysis described above. The model uses the grid cell as a primary hydrological unit and models the following dominant hydrological process: interception, snow accumulation and melting, root zone soil moisture dynamics, evapotranspiration, surface flow, interflows, recharge and baseflow. The total runoff generated at each grid cell is routed to the neighboring downstream cell following the river network using the Muskingum-Cunge routing algorithm. Interested reader may refer to (*Samaniego et al., 2010a*) for further details on the model components. The model code is open source and can be downloaded from www.ufz.de/mhm. The model has been successfully applied to a number of river basins across Germany, USA and Europe (~~*Samaniego et al., 2010a, b; Kumar et al., 2010, 2013a, b; ?, ?; Thober et al., 2015;*~~

An important and unique feature of mHM is its Multiscale Parameter Regionalization (MPR), that explicitly accounts for subgrid variability of basin physical characteristics such as terrain, soil, vegetation, and geological properties (*Samaniego et al., 2010a; Kumar et al., 2013a*). The model considers different levels of spatial resolution to better account for spatial heterogeneity of inputs, forcings and the modeled hydrological processes (see schematic in Figure 3). The smallest scale (called l_0 within the mHM nomenclature) is representing morphological factors, like elevation, soil type, land cover etc. On the other hand, meteorological inputs can be represented on a larger scale (called l_2 within mHM). The modeling of the hydrology is done on a third scale (called l_1 within mHM) that can vary depending, e.g., on catchment size or computational resources. Based on the MPR technique, morphological inputs are linked to internal model parameters (e.g., through the use of pedo-transfer functions) and a set of regional coefficients (or global parameters, γ). In a second step, the internal parameters are upscaled to the resolution of the hydrological processes, i.e., l_1 , using parameter specific upscaling operators. Thus, MPR takes indirectly subgrid variabilities into account. The global parameters (γ) are space and time invariant and are inferred via a calibration procedure. mHM has 66 global parameters, which is a reasonable number for an optimization problem and is therefore able to avoid overparameterization. Further details on MPR can be found in *Samaniego et al. (2010a); Kumar et al. (2013a)*.

30 Relevant to this study is near-surface and root-zone hydrological process, which are computed using different conceptualizations. In the upmost layer (x_3 in Figure 3) ~~soil moisture~~ water content is estimated using the infiltration excess ap-

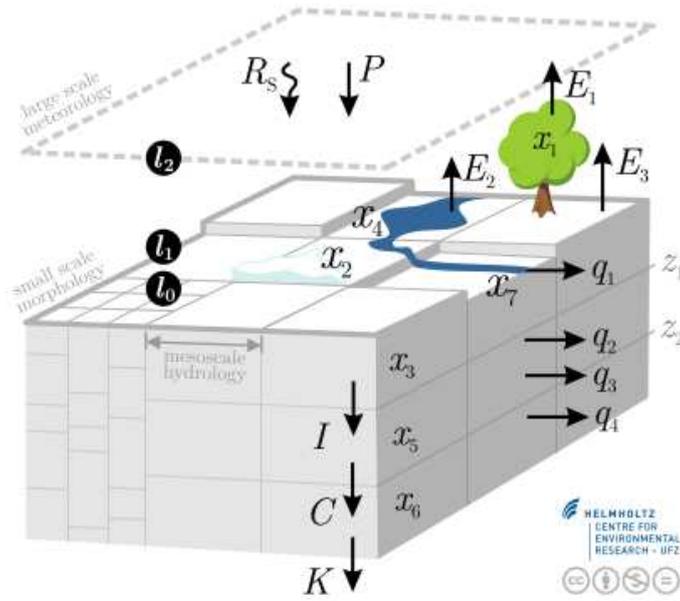


Figure 3. Schematic of the mesoscale hydrological model used in the study, depicting the different scales as well as the states and fluxes represented in a single cell.

proach similar to the HBV model (Bergström, 1995), but enhanced to account for multiple soil layers. Within this layer these layers, the water is either percolating into deeper layers or evapotranspirates to the atmosphere. Therefore, the root zone is characterized by effective parameters for porosity, saturated hydraulic conductivity and the permanent wilting point, which are estimated based on the pedotransfer functions of Zacharias and Wessolek (2007). These effective parameters are estimated due to transfer functions from the global parameters, which are determined during the calibration process. Evapotranspiration is estimated based on potential evapotranspiration, root water uptake and water availability in layer x_3 . In the second layer (x_5 in Figure 3) two different types of interflow take place. Slow interflow q_3 is implemented using a power-law model, whereas fast interflow q_2 is triggered when a threshold value $TV \gamma_{TV}$ is reached, i.e

$$q_3 = \gamma_s x_5^{(1+\alpha)}, \quad (5a)$$

$$q_2 = \begin{cases} \gamma_f (x_5 - TV \gamma_{TV}), & \text{if } x_5 > TV \gamma_{TV} \\ 0, & \text{otherwise} \end{cases}. \quad (5b)$$

In the third level (x_6 in Figure 3) baseflow q_4 is generated using a simple reservoir model, i.e.,

$$q_4 = \gamma_b x_6. \quad (6)$$

However, due to the strong uncertainty typically associated with the estimation of the groundwater storage, we excluded this layer from our study. In the following all analysis is therefore confined to the travel time behavior of the soil only.

These runoff generation processes are represented at every grid cell of mHM. The sum of direct runoff q_1 (not used for the analysis), interflows and baseflow constitutes the grid specific total runoff which is then routed through a river network.

Interested readers may refer to *Samaniego et al. (2010a)* or www.ufz.de/mhm (user manual) for further details on mHM.

As motivated in the Introduction, we followed the concept of e.g., *Botter et al. (2010)* and *Benettin et al. (2015b)* and divided the subsurface into two distinct zones; the soil zone (called root zone by *Botter et al. (2010)* and shallow storage by *Benettin et al. (2015b)*) and the saturated zone (called groundwater region by *Botter et al. (2010)* and deep storage by *Benettin et al. (2015b)*). All analysis in our study was performed with respect to the former. This was seen necessary due to the large uncertainties associated with storage estimation of the deeper regions. Whereas mHM has been demonstrated to provide good estimates for soil moisture (*Rakovec et al., 2016*), storage estimates for groundwater (x_6 in Figure 3) are far less reliable. Focusing on the soil zone, only, was seen justified since the focus of our study was the investigation of spatially distributed factors like precipitation, land cover and soil type, which have comparably little impact on groundwater dynamics. Furthermore, *Benettin et al. (2015b)* demonstrated that travel-time behavior in the deeper zone has comparably little temporal variability, too.

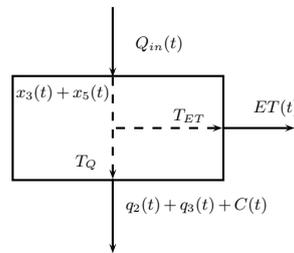


Figure 4. States and fluxes as computed by mHM (see Figure 3) for the derivation of TTD's using Equations 2 to (3) (see Figure 1).

For computation of the TTD's according to Equations (2) to (4), we used for the storage S the combined estimates of layer x_3 and x_5 of mHM (see Figures 3 and 4). For ET we used fluxes the evapotranspiration from x_3 and for Q we used q_2 , q_3 and C . Conceptually the interflow is generated in the unsaturated zone (reservoirs x_3 and x_5) within mHM. Thus, using the interflow as the outflow from the unsaturated zone for deriving the travel times is a valid assumption. Our delineation of shallow and deeper storage was therefore more similar to *Benettin et al. (2015b)* than to *Botter et al. (2010)*.

2.3 Study area and model set-up

In this study, we used a mesoscale catchment in Central Germany with a drainage area of approximately 1000 km^2 to the gauging station at Nagelstedt (see Figure 5). The catchment is the headwaters of the Unstrut river basin, and was selected in this study for its relevance to the Collaborative Research Center AquaDiva (*Kusel et al., 2016*). The terrain elevation within the catchment ranges between 170 m and 520 m with the higher regions in the west and south being the forested hill chain

of the Hainich (see Figure 5). The forested area covers approximately 17% of the catchment, while 78% of the area is covered by crop/grassland. The remaining 5% is urban/build up area. The area is characterized by continental climatic conditions with a mean annual precipitation of approximately 660 *mm* and a mean temperature of approximately 8 °C.

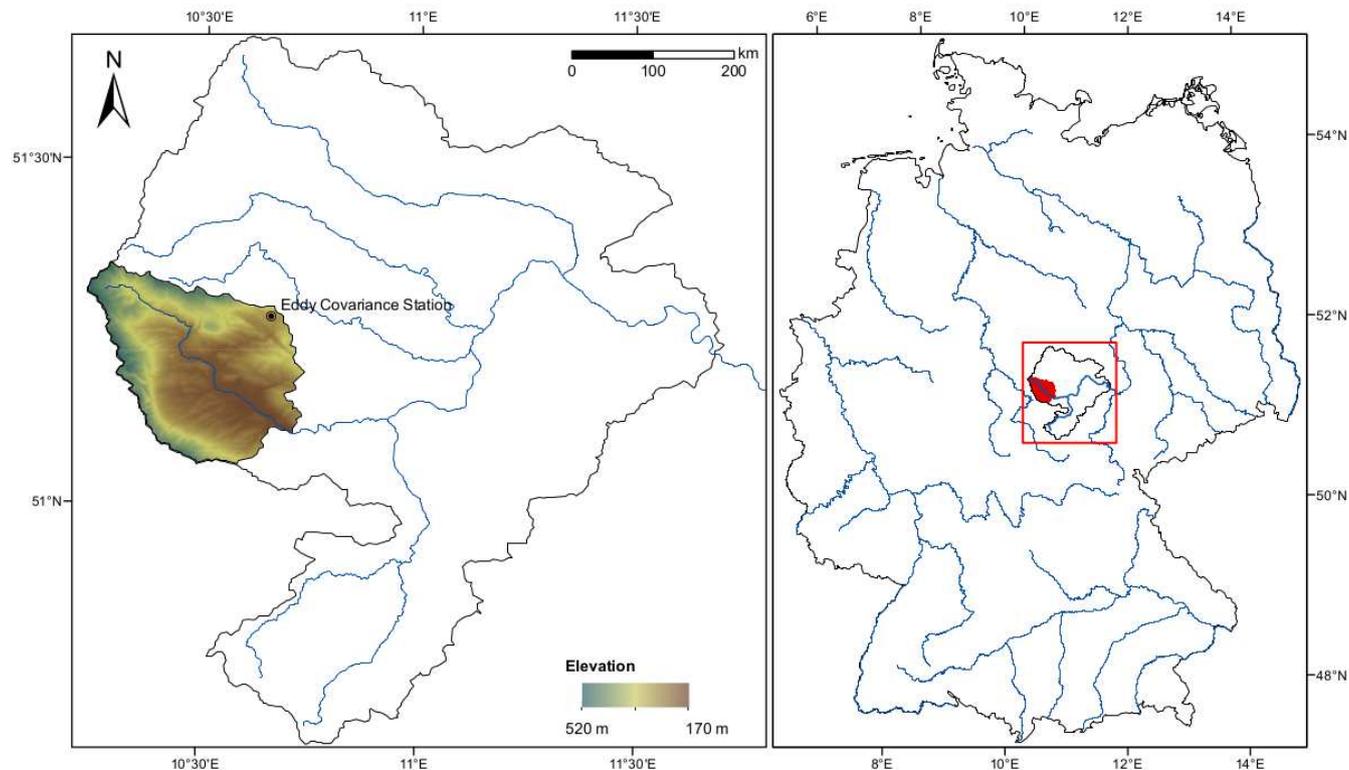


Figure 5. Left: Catchment (highlighted) used in the study shown within the larger confines of the Unstrut catchment (area enclosed by continuous line). The larger rivers of the catchment are shown in blue. The colorbar shows the elevation (in *m*) of the study catchment. Right: Unstrut catchment within the larger confines of Germany. The axis descriptions denote the latitude and longitude values.

We established mHM over the study catchment and performed numerical simulations on several resolutions ranging from 5 200 *m* to 2 *km*. The model was forced using daily gridded fields of precipitation, air temperature and potential evapotranspiration. The point datasets for the precipitation and air temperature at several raingauges and weather stations located in and around the catchment were acquired from the German Meteorological Service (DWD). These point stations were then interpolated on regular grids using an external drift kriging interpolation procedure wherein the terrain elevation was used as an external drift (?)([Samaniego et al., 2013](#)). The potential evapotranspiration was estimated using the *Hargreaves and Samani* 10 (1985) method. Other datasets required to set-up the model include digital elevation model (DEM) and derived terrain properties like slope, aspect, flow direction, catchment boundary; soil and geological maps were provided by the Federal Institute for Geosciences and Natural Resources (BGR) and meta data such as sand and clay contents, bulk density, horizon depths,

dominant hydrogeological classes; CORINE land cover information for the years 1990, 2000 and 2005 available from the European Environment Agency (EEA); and runoff data for the catchment outlet provided by the European Water Agency (EWA) and the Global Runoff

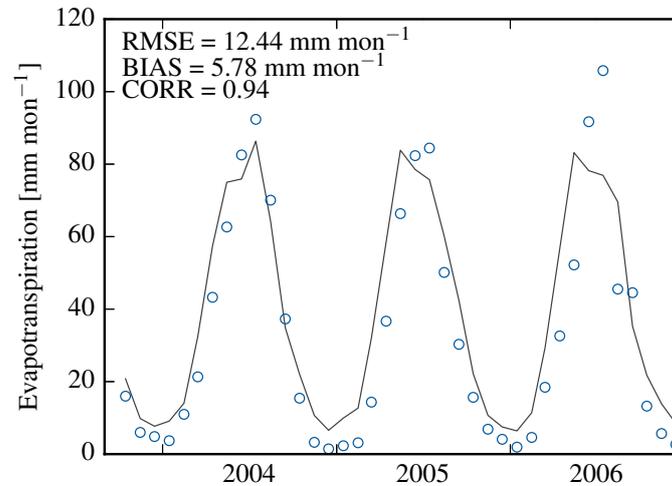


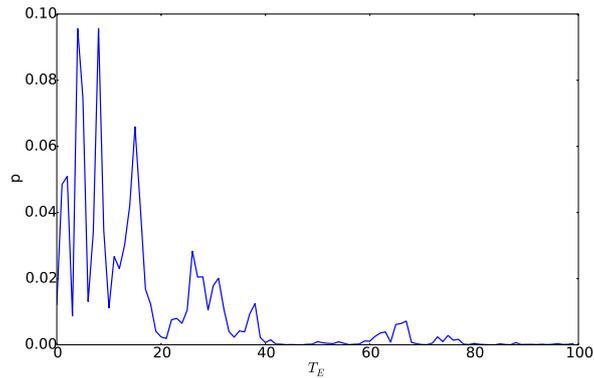
Figure 6. Comparison between monthly measured and modeled evapotranspiration (ET) at the eddy-covariance station Mehrsted (see Figure 5).

The model simulations were performed for the period 1950 to 2005. The first five years of the data were used to warm-up the model to acquire plausible initial conditions. We therefore discarded the first five years of simulations and the further analyses were performed using model outputs for the period 1955 – 2005. The model showed quite good performance with $NSE > 0.8$ for the daily discharge simulations at the Nagelstedt station. Other statistics like bias and correlations were also within a satisfactory range. To further validate our model prediction, we used measurements from a single Eddy Covariance measurement station inside the study area (see Figure 5). This comparison also showed a good agreement between both measurements and model prediction (see Figure 6).

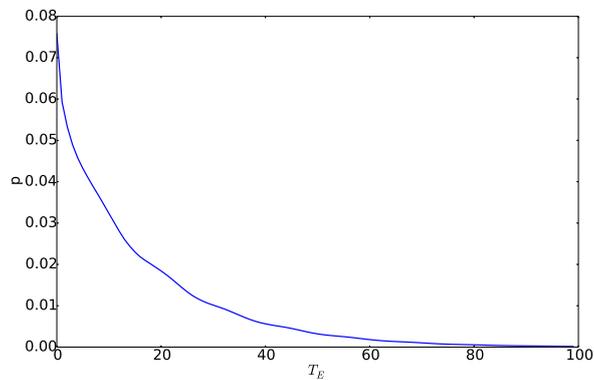
3 Results and discussion

In this section, we present and discuss the results which have been derived using the methods described above. We will begin in the following by demonstrating and exemplify our general research procedure by virtue of a singly yet representative example.

The starting point for the derivation of soil travel times were the states and fluxes as computed through mHM. Using the time series of soil moisture, evapotranspiration, interflow and recharge, we used Equation (2) to compute the travel time distribution (see Figure 7 a). One of the problems when computing forward TTD's by virtue of Equation (2) is that all the water entering the CV at time t_{in} must leave until the end of the computing period. This means that a certain amount at the end of the available time series could not be used for the analysis. To determine this period, we computed θ_{in} with respect



(a) time-dependent TTD



(b) stationary TTD

Figure 7. Forward TTD of soil moisture with respect to mean travel time (in months) for a single cell ~~within~~ in the Nagelstedt catchment. Panel (a) ~~shows the~~ time-dependent TTD derived using Equation (2) for a given t_{in} . Panel (b) ~~shows the~~ stationary TTD derived using Equation (7).

to discharge as well as to evapotranspiration. Adding up both values for a given t_{in} should add up to one, i.e., all water that entered at t_{in} has left within the available time frame. A value smaller than one therefore indicates that some amount of the water is still inside the CV with possible error inducing effects on the calculation of the TTD's. Analyzing this behavior, we concluded that close to 2 years at the end of the available time series had to be excluded for the calculations of the TTD's (data not shown). The shape of ~~these~~ the resulting time-dependent distributions varied strongly, depending in particular on rainfall events that triggered the mobilization of older water stored within the soil. Another factor, although not apparent from Figure 7, was the water content, i.e., the state of the soil itself. As been demonstrated by Niemi (1977), soil response to rain events is strongly different between wet or dry conditions.

To disentangle this event-driven as well as state-dependent effects from other factors that influence the water movement in the soil, we averaged these time dependent distributions. As a result, we got the stationary ~~FFDs~~-TTD's for every cell

$$\vec{p}_Q(T_E) = \int \frac{Q(t)}{\theta(t_{in})S(t)} \exp\left(-\int_{T_E} \frac{Q(t') + ET(t')}{S(t')} dt'\right) dt_{in} \quad (7)$$

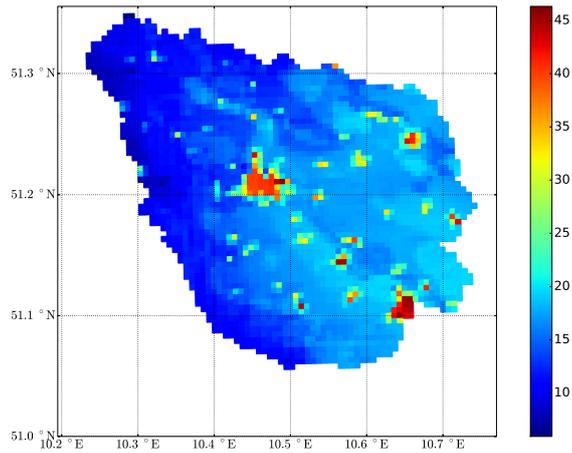
with $T_E = t - t_{in}, t > t_{in}$.

5 In all investigated cases, these stationary ~~FFDs~~-TTD's could be well approximated by an exponential-like behavior (see Figure 7 b). Behavior as seen for stationary ~~FFDs~~-TTD's is often assumed to be valid for ~~FFDs~~-TTD's in general and are consequently modeled using exponential or gamma distributions (*Maloszewski and Zuber, 1982*). Recent works however, have questioned this generalization by emphasizing the time-dependent nature of ~~FFDs~~-TTD's (*Duffy, 2010; Botter et al., 2011*). The examples given in Figure 7 exemplify these concerns by illustrating their respective origins. Consequently, we acknowledged
 10 the inherent differences between these two ~~FFDs~~-TTD's. Furthermore, the study area falls within a humid region with soils being generally wet and rainfall being evenly distributed throughout the year. Under these conditions the assumption of (quasi) stationary TDDs is reasonable (*Tetzlaff et al., 2007; Hrachowitz et al., 2009*). These stationary ~~FFDs~~-TTD's provided the basis for all following analysis, due to allowing the description of the average hydrological response of the catchment. In addition, we also focused on travel-time behavior under specific hydrological regimes, i.e., wet and dry conditions, providing
 15 a more detailed understanding of the catchment.

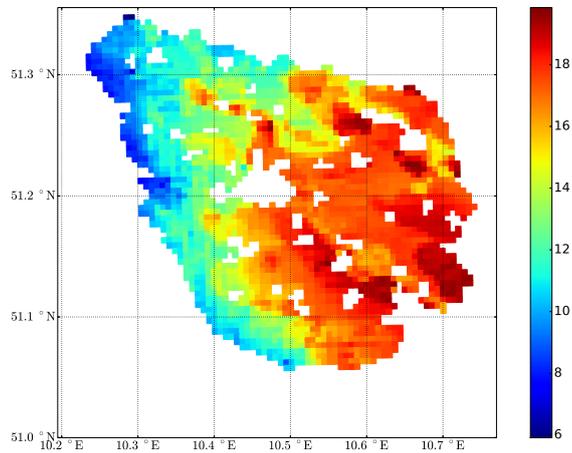
For our statistical analysis, we used these stationary ~~FFDs~~-TTD's, which, due to their exponential-like behavior, can be characterized by its expected value τ . We will call this value mean life expectancy (or mean age respectively) in the following. Estimating this value for every mHM cell provided a single measure for the travel-time behavior in the soil without the otherwise dominating impact of single precipitation events (see Figure 8). One feature that became immediately apparent
 20 were the long travel times in urban areas (see Figure 8 a). This can be explained by the fact that these areas are largely sealed, resulting in low infiltration rates and consequently low turnover rates inside the soil. To disentangle this sealing effect from the soil behavior, we discarded cells inside urban regions from our analysis (see Figure 8 b). This allowed us to investigate the interplay between soil properties and travel-time behavior apart from such artificial influences.

3.1 Impact of modeling resolution

25 Due to its multiscale parameterization, mHM is able to model catchment dynamics at different spatial resolution with the same set of calibration parameters (see e.g., *Samaniego et al. (2010a)* or *Kumar et al. (2013a)*). Within the context of ~~FFDs~~-TTD's, this feature may be used to investigate the potential influence of age-dependent outflow generation. The mathematical theory for including such age dependency has been developed independently by different groups and recently been unified using the umbrella term of StorAge Selection (SAS) functions (*Rinaldo et al., 2015; Harman, 2015*). These functions fully describe
 30 the sampling behavior of the catchment with respect to the age distribution of the stored water when discharge is generated.



(a) all mHM-cells



(b) non-urban mHM-cells

Figure 8. Mean life expectancy (in months) of soil moisture derived by Equation 7 for the Nägelstedt catchment (see also Figure 1 for comparison) once for all mHM cells (left) and for all non-urban cells.

Discharge from a catchment may e.g., be primarily composed of younger or older water or it may show no preference to age whatsoever. SAS functions are therefore a concise mathematical representation of this behavior.

On a physical basis, such preference for different water age should be interpreted as the result of complex mixing processes taking place in the subsurface of the catchment (Botter, 2012; Benettin et al., 2013; van der Velde et al., 2012). In order

to determine the appropriate SAS function for a given catchment, predictions using different functions could be compared with measurements. Alternatively, the form of the SAS function can be determined by using a physically based catchment model (Cornaton and Perrochet, 2006a, b). As already mentioned above, we could not directly infer, which form of a SAS function would be the most appropriate choice for our catchment. Instead we calculated the mean life expectancy for our catchment on different scales using the uniform SAS function. We motivated this choice by the principle of least information (or principle of maximum entropy) stating that amongst different alternatives, the one with the least amount of information should be chosen. Without any additional constraints, a uniform distribution is usually associated with maximum ignorance, therefore, motivating the use of the uniform SAS function.

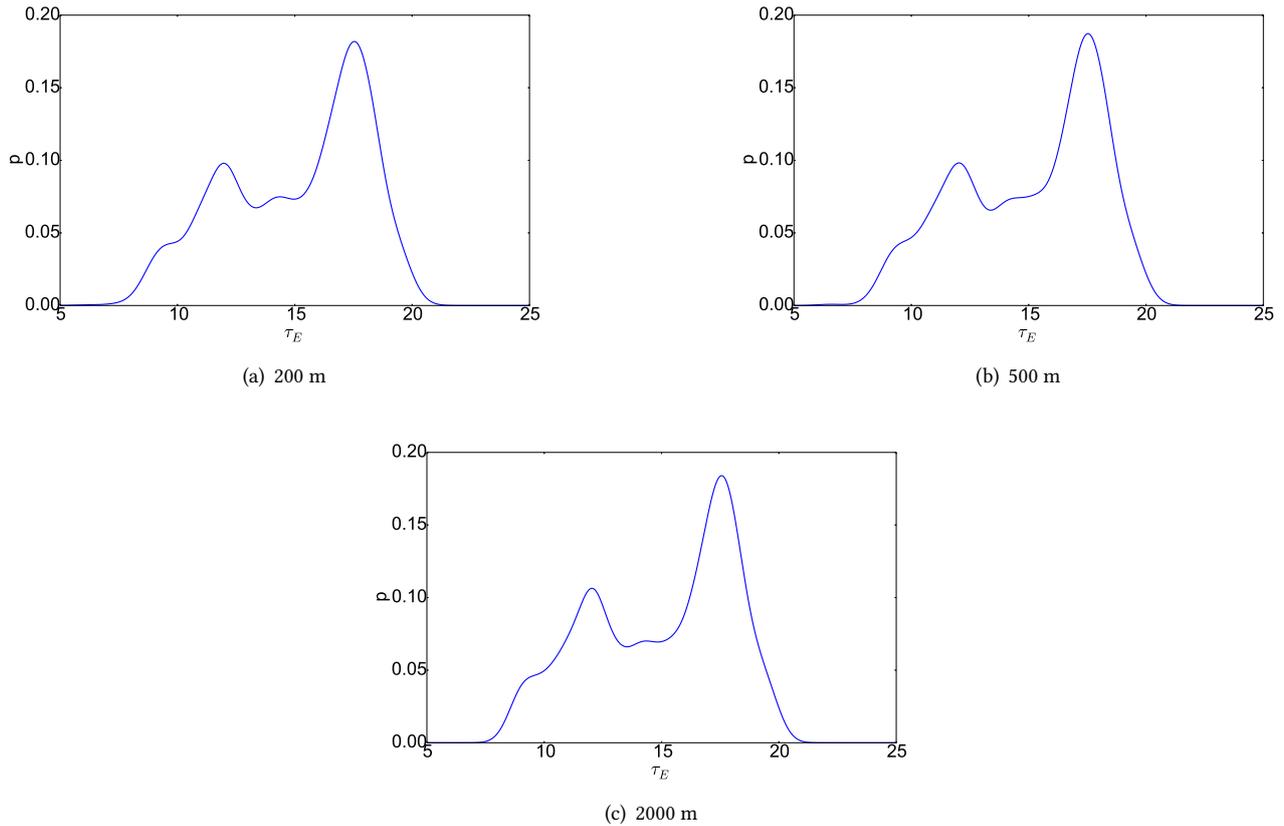


Figure 9. Kernel density estimate of the mean life expectancy (in months) of soil moisture for several grid sizes in the Nängelstedt catchment.

To estimate the possible influence of this decision, we reasoned that a scale-dependent bias in the estimation of travel-time behavior would indicate the existence and possible strength of such an error. This is due to the multi-scale nature of mHM, where subgrid heterogeneity is taken into account by virtue of the Multiscale Parameter Regionalization. Using a smaller grid size would make this heterogeneity explicit and therefore reveal any possible unaccounted subgrid influence. Results from our simulations showed no discernible differences in the statistical distribution of mean life expectancy (see Figure 9). Using

a smaller resolution had positive effect on statistical estimation procedure due to the increase in data points. In addition, we saw more extreme values due to small scale features that were smeared out on coarser resolutions. Other than these two changes, we noted only minor changes in the statistics of mean life expectancy. We therefore concluded that, within the limits of the spatial scales tested here, mixing processes inside our catchment have no major impact on mean life expectancy.

5 We are aware, that this assessment is only covering one possible source of age-dependent outflow behavior and that other unresolved heterogeneity (at even smaller scales or due to other subsurface properties not accounted for in mHM) would influence the outflow generation as well. We therefore regard our conclusions as tentative and open to revision once actual measurements become available.

10 However, our investigation gave us the ability to find a good trade-off between computational costs and data amount for the following statistical analyses. We therefore used a data set from simulations using a grid size of 500 m .

3.2 Statistical analysis of mean life expectancy

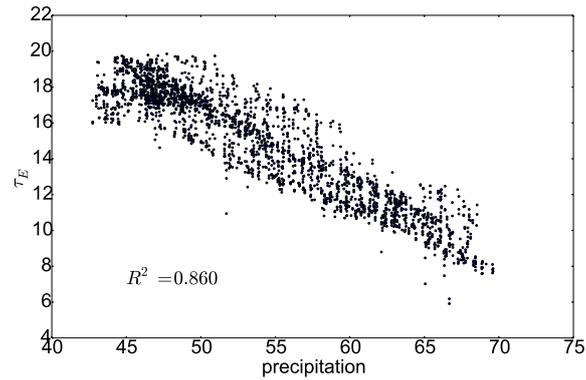
The mean life expectancy τ of a water parcel inside a catchment is the result of a complex interplay of morphological and climatological factors. Several recent studies have therefore tried to determine their relative importance under varying conditions (McGuire *et al.*, 2005; Cardenas, 2007; Broxton *et al.*, 2009; Tetzlaff *et al.*, 2009, 2011). Contrary to these studies where field
15 measurements were used, we used results from computational simulations only. This gave us a much larger dataset, both in time and space, from which we could infer the relative impact of different factors, in particular meteorological (precipitation), land surface (land cover, leaf-area index) and subsurface (soil) properties. Notably, our approach differs from Hrachowitz *et al.* (2009) such that our analysis is based on model-derived gridded simulations of TDDs as compared to the observation-based basin-wise quantification of TDDs.

20 In the first step, we determined for every cell the statistical relationship between the mean life expectancy τ and a number of potential predictors like average precipitation, soil depth, soil type or leaf-area index (LAI). Similar to Hrachowitz *et al.* (2009), we used the coefficient of determination R^2 to quantify the strength of the statistical relationship. This quantity equals to one minus the ratio of the remaining variance vs. the total variance of the data themselves. It is therefore a measure of the variance explained by the model (which was always assumed to be linear in our study).

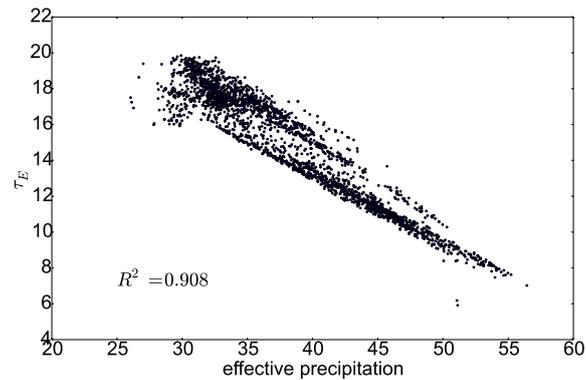
25 3.2.1 Precipitation

The analysis above showed the strong impact of precipitation on the event-based ~~TTD's~~-TTD's (see Figure 7). It is therefore to be expected to exert strong control on the steady-state ~~TTD's~~-TTD's as well. In our model two different quantities can be distinguished: first, the precipitation itself as well as, second, the effective precipitation. The latter value is here defined as
30 the water flux that is actually entering the soil, i.e., corrected by surface runoff (through sealing), canopy interception and snowmelt. While the precipitation can be measured with high accuracy, it is the effective precipitation that directly impacts the soil-moisture dynamics.

The scatterplot of both data sets against the mean life expectancy show a significant negative correlation between them (see Figure 10). This negative relationship can be explained such that precipitation events apply pressure to the water already



(a) precipitation



(b) effective precipitation

Figure 10. Scatterplot of mean life expectancy (in months) of soil moisture vs. monthly values (in mm) for precipitation (Panel (a)) and effective precipitation (Panel (b))~~(in mm) vs. mean life expectancy (in months).~~

stored in the soil. Instead of immediately traveling through the soil, the water from these events rather pushes older water out. Strong precipitation events therefore lead to a 'flushing out' of the soil and cause a shorter life expectancy.

3.2.2 Terrain elevation

In our next analysis, we used the physical elevation as a variable for our regression model. The height can simply be derived from the digital elevation model (DEM), which, in mHM, is represented using data obtained from the Shuttle Radar Topography Mission.

Using a scatter plot for visualizing the statistical relationship between mean life expectancy and the DEM showed a negative correlation (see Figure 11 a), i.e., longer life expectancy correlated with lower heights of the terrain, and with a linear coefficient of determination of $R^2 = 0.668$. Since no direct causal connection can be drawn between physical elevation and

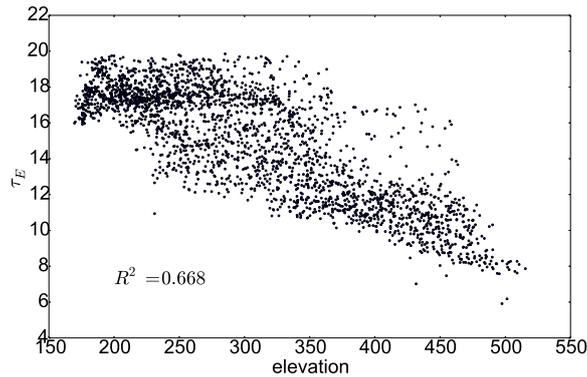


Figure 11. Scatterplot of elevation-mean life expectancy (in months) of soil moisture vs. mean life expectancy-elevation (in months).

travel-time behavior, such a high value is indicative of underlying mechanisms. One of these is the aforementioned precipitation, since higher altitudes are correlated with stronger mean precipitation levels (linear coefficient of determination of $R^2 = 0.812$). Performing a multiple linear regression, including precipitation and saturated soil moisture (discussed below), showed strong correlation between these variables (data not shown). It therefore stands to reason to attribute potential causal effects to these covariates, only.

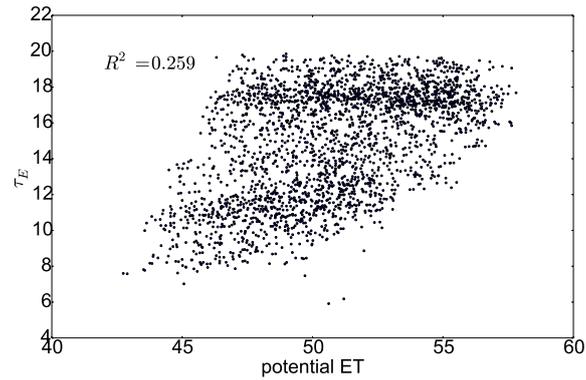
3.2.3 Evapotranspiration

Evapotranspiration is directly influencing the form of a TTD (see e.g., Equation 2). Consequently, we anticipated a strong correlation between mean evapotranspiration rates and mean life expectancy.

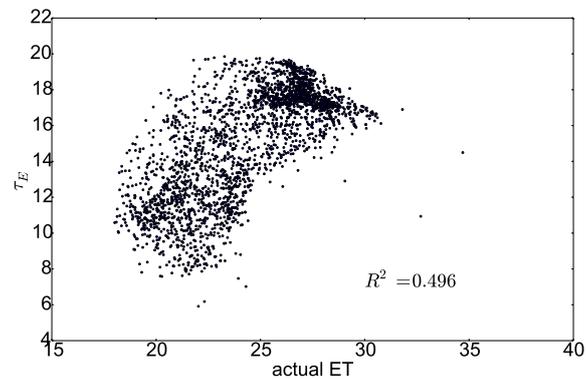
With respect to evapotranspiration, two different definitions are typically distinguished: potential evapotranspiration (PET) and actual evapotranspiration (AET). As implied by its name, PET describes the maximum possible rate of evapotranspiration at a given site. This value is dependent on quantities like solar radiation and temperature that can generally be measured with good accuracy (Samani, 2000). Using theoretical models, good estimates can therefore be provided for PET at a given site (Almorox et al., 2015). On the other side, AET is a real quantity that can be measured. In principle, in situ measurements can therefore provide good estimates (e.g., the eddy-covariance method). In practice, however, exact measurements are hampered by a series of factors (Wang and Dickinson, 2012). As a consequence, PET can often be estimated with higher accuracy than AET.

Scatterplots of both PET and AET show a positive correlation between evapotranspiration and mean life expectancy in general (see Figure 12). This correlation is more pronounced for AET with a coefficient of determination of $R^2 = 0.496$ vs. only $R^2 = 0.259$ for PET.

Contrary to precipitation, which is an inflow mechanism, ET is an outflow mechanism. It is not pushing but rather sucking the water out of the CV, which explains the difference in behavior of precipitation and ET. The lower relative strength of the correlation (compared to precipitation) can be explained such that ET is only one of the two outflow mechanisms (the



(a) PET



(b) AET

Figure 12. Scatterplot of mean life expectancy (in months) of soil moisture vs. monthly ~~potential as well as actual~~ evapotranspiration values (in *mm*). Displayed are both potential evapotranspiration (Panel (a)) and actual evapotranspiration (Panel (b)).

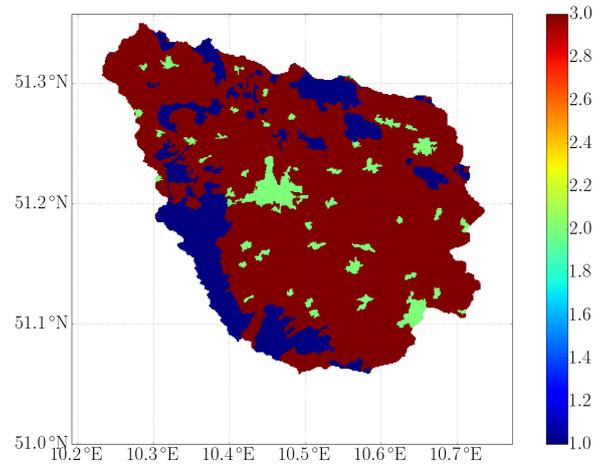
other being discharge). The relative stronger impact of AET compared to PET was also anticipated. AET is directly used in Equation (2) for the calculation of ~~TTD~~TTD's, whereas PET is only coupled by virtue of an additional function.

As explained above, for real-world situations, better estimates can often be provided for PET. The higher explanatory power of AET has therefore to be balanced with its often less accurate estimate. Depending on the accuracy of measurements of
 5 AET, PET estimates may be a better predictor of mean life expectancy.

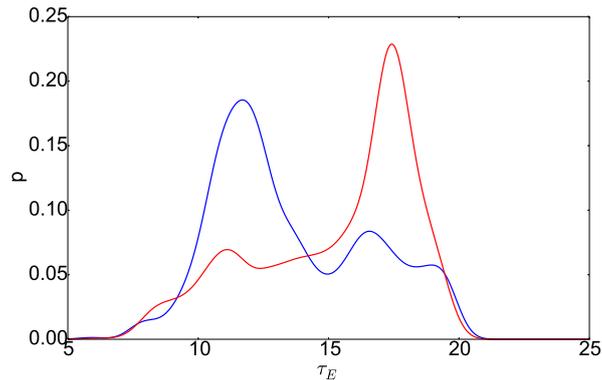
3.2.4 Land cover properties

Land cover is an important interface controlling the strength of incoming fluxes through artificial and natural sealing. In mHM, three different land-cover types are distinguished: forest, crop/grassland and urban area. As explained above, we excluded mHM cells inside urban area from our analysis in order to better focus on the soil properties themselves. To further

elucidate possible influence of the remaining land cover types, we separated the catchment into forest and crop/grassland and calculated the mean travel times separately.



(a) spatial distribution



(b) kernel density estimate

Figure 13. Land cover in the Nängelstedt catchment (1-blue = forest, 2-green = urban, 3-red = crop/grassland). Spatial Panel (a) shows the spatial distribution of land cover in the highest resolution l_0 and Panel (b) shows the kernel density estimate estimates of the mean life expectancy (in months). The blue curve represents forest and of soil moisture for the red curve represents grassland land cover types.

Estimating the PDF of the mean life expectancy for both land cover types separately, revealed strong differences between them both in shape of the respective PDF and the range of values (see Figure 13). As shown above, results for the combined data set showed a distinct bimodal behavior (see Figure 9). In contrast to that, the PDF for both land cover types were almost

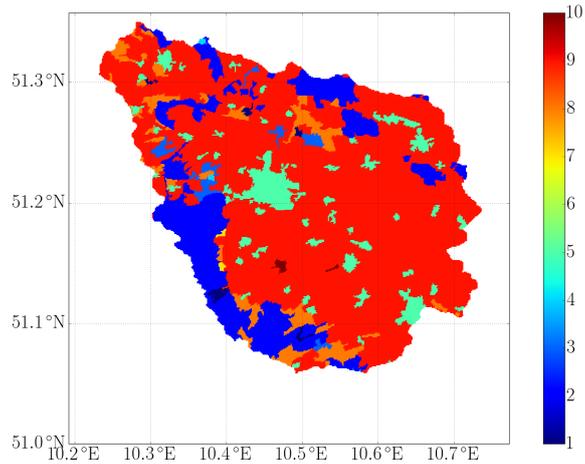
unimodal. The most dominant peaks of every singly PDF coincided with the two peaks of the combined PDF. The behavior of the latter can therefore – to some degree – be considered to be a superposition of the former.

The relationship between these two land cover types was such that forest resulted in much shorter mean travel times compared to crop/grassland. This pronounced difference may be partially due to a correlation with precipitation patterns that have already been shown to exert a strong influence on travel-time behavior. Forest in the study catchment (as well as in Germany in general) is disproportionately found in hilly and mountainous regions. These regions in turn show stronger precipitation values. The tendency depicted in Figure 13 may therefore be also caused by this covariate. However, this correlation between forested and high-precipitation area would not explain the distinct differences between both land-cover types. Another factor, overlapping with the former, may be due to the differences in water uptake. Trees are rooted into deeper soil layers compared to crop and grass and are therefore able to access a larger part of the subsurface water body. This larger access combined with the higher precipitation values as well as other factors would explain the almost non-overlapping travel-time behavior demonstrated in Figure 13.

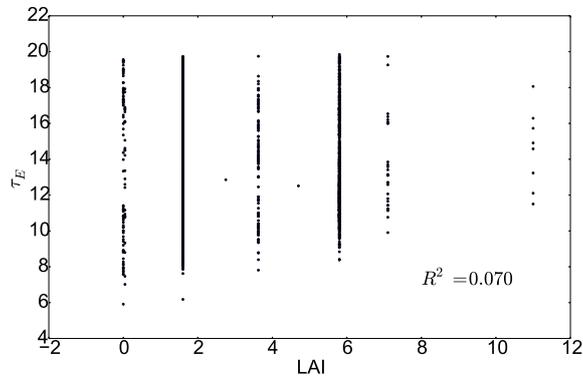
In addition to this classification scheme, mHM uses the leaf area index (LAI) to describe land cover properties. The LAI describes the ratio of the cell that is effectively covered by plant canopy. Due to the already established influence on evapotranspiration (see above), it stands to reason to expect an influence on the mean life expectancy as well. Comparing LAI class and land cover reveals a strong overlap between both (see Figure 13 (a) and Figure 14 (a)). Roughly, forest land cover corresponds with LAI class 1 – 4, urban land cover corresponds with LAI class 5 and grassland corresponds with LAI class 6 – 10.

Using the same approach as above, i.e., investigating the mean life expectancy for every LAI class independently, consequently revealed the same overall tendency for LAI classes compared to land cover types (data not shown). This was anticipated due to the aforementioned overlap between the two classification schemes. In addition, we saw little diversity for LAI classes within the same land cover class (data not shown).

However, this tendency was not present when using the actual leaf-area values associated with every LAI class. These values could be constant over the year e.g., in case of coniferous forest or vary strongly e.g., in case of deciduous forest. To make values from different LAI classes comparable, we averaged the respective values year-wise. A scatter plot of leaf area index vs. mean life expectancy does not show any strong correlation between the two with similar ranges of values being found for almost all LAI values (see Figure 14 (b)). This discrepancy can be explained by the implementation of the LAI in mHM. In contrast to the land cover type, that is used for the determination of ET processes in the upmost soil layer, LAI values are only used for interception and do consequently not directly influence travel-time behavior. As a result, any possible relationship between LAI and ~~TTD~~TTD's is therefore biased and conclusions from our results must take into account this limitation critically.



(a) spatial distribution



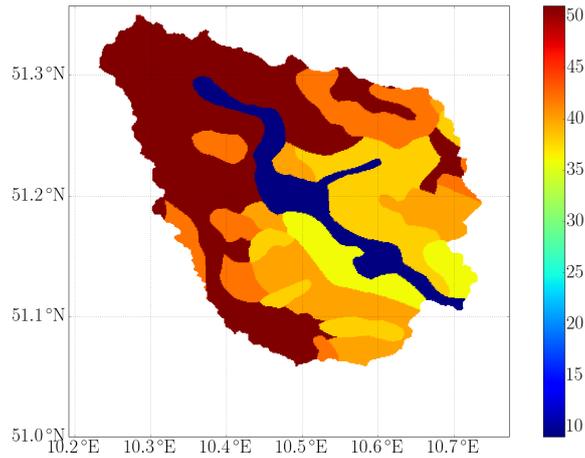
(b) scatter plot

Figure 14. Leaf area index (LAI) in the Nängelstedt catchment. Spatial-Panel (a) shows the spatial distribution and Panel (b) shows the scatter plot of LAI vs. mean life expectancy (in months) of soil moisture vs. LAI.

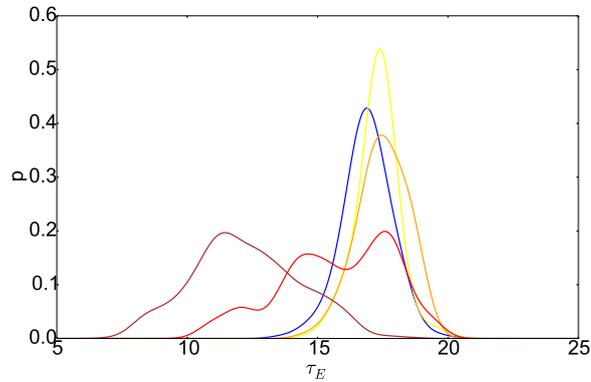
3.2.5 Soil properties

An important input parameter in mHM is the soil type inside every cell. This property is implemented in mHM using the German soil data base Bodenübersichtskarte 1:1.000.000 (BÜK 1000) (*Federal Institute for Geosciences and Natural Resources (BGR), 1998*).

- 5 Due to this relevance in the model, we anticipated a strong impact of the soil type in a cell on the resulting mean life expectancy. Estimating the PDF of mean travel times for every soil type individually, did indeed show significant differences between them (see Figure 15). Soil classes found in the geographically lower regions of the catchment generally show longer



(a) spatial distribution



(b) kernel density estimate

Figure 15. Soil classes in the Nängelstedt catchment. **Spatial** Panel (a) shows the spatial distribution and Panel (b) shows the kernel density estimate of the mean life expectancy (in months) of soil moisture for selected soil classes. Blue curve represents soil class 9 (36% sand and 10% clay), yellow curve represents soil class 38 (12% sand and 15% clay), orange curve represents soil class 40 (10% sand and 19% clay), red curve represents soil class 42 (7% sand and 39% clay) and brown curve represents soil class 51 (19% sand and 70% clay).

mean travel times with a unimodal distribution shape, whereas soil types in the geographically higher regions correspond with generally shorter mean travel times with the shape of the distributions being less regular. This qualitative analysis reveals some overlap with the land cover distributions as well as mean precipitation rates. It is consequently not possible to directly infer causal correlation from statistical correlation.

In addition, the soil class is a symbolic variable, i.e., its values only indicate a certain type of soil but does not directly relate to any numerical quantity associated with this soil type. Consequently, we could not infer any quantitative connection between soil types and resulting travel-time behavior.

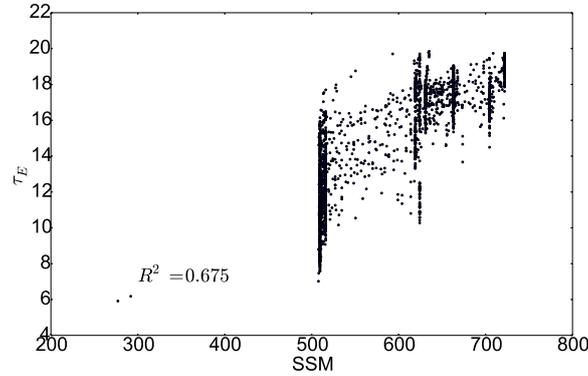


Figure 16. ~~Statistical relationship between~~ Scatter plot of mean life expectancy (in months) and of soil moisture vs. saturated soil moisture (in mm).

To address this problem, we used the saturated soil moisture of the soil. This quantity is the amount of pore space per cell that can be potentially filled with water (porosity times the depth of root-zone soil layer). Its value is determined in mHM through pedo-transfer functions using the soil textural information on percentage of sand, clay and bulk density. Comparing these values in every single cell with the mean life expectancy shows a very strong statistical relationship with a coefficient of determination $R^2 = 0.675$.

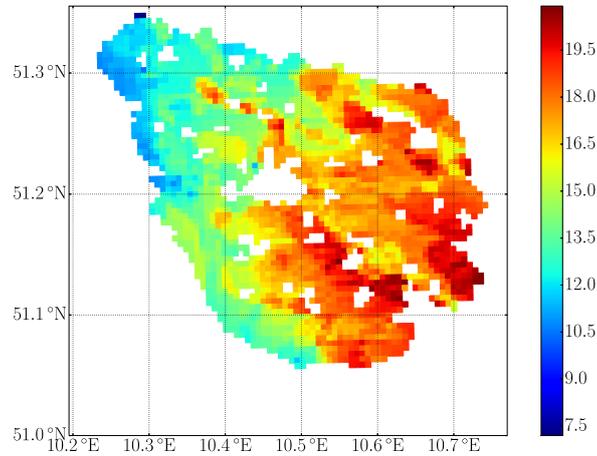
The high correlation values of the saturated soil moisture can be explained by a mixture of causal and statistical factors. On one hand, it is reasonable to expect the total amount of storage to be filled with water to have a significant effect on the resulting travel-time behavior. On the other hand, the soil types show a strong overlap with other factors like precipitation levels and land cover types that have already been discussed above.

3.3 Statistical analysis of mean age

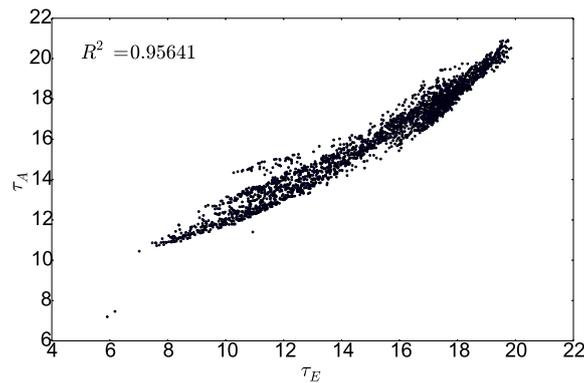
As described above, the difference between the forward and backward formulations of travel time has long been acknowledged (Niemi, 1977) and many studies have investigated their relationship (Cornaton and Perrochet, 2006a; Botter, 2012; Benettin et al., 2013; Harman, 2015; Benettin et al., 2015a). Both these formulations are linked by virtue of the so called Niemi relation

$$J(t_{in})\theta(t_{in})\vec{p}(t - t_{in}|t_{in}) = Q(t)\overleftarrow{p}(t - t_{in}|t), \quad (8)$$

which can be derived by considering a water parcel entering the CV at t_{in} and leaving at t . Consequently, mean life expectancy and age only coincide in case of steady-state conditions. As a result, we also investigated the behavior of mean age to elucidate connections and differences between forward and backward formulations for our catchment.



(a) spatial distribution



(b) scatter plot

Figure 17. Mean age of soil moisture in the Nängelstedt catchment. **Spatial** Panel (a) shows spatial distribution and Panel (b) shows scatter plot of mean age vs. mean life expectancy (see Equation 8) in months.

Visually comparing mean age (see Figure 17) and mean life expectancy (see Figure 8 b) in the Nängelstedt catchment showed strong qualitative and quantitative similarities. Accordingly, we also got a very strong statistical relationship between these two quantities with a coefficient of determination of $R^2 = 0.956$. Overall, the relationship was very linear with mean age values falling short of mean life expectancy for both small and large values.

	Pre	PET	SSM
mean life expectancy	0.860	0.260	0.675
mean age	0.728	0.143	0.711

Table 1. R^2 values for several predictors of mean travel time

Due to the mathematical and physical similarities, such a strong connection was anticipated. To further investigate possible origins of their respective differences, we performed the same statistical analysis for mean age.

To that end, we considered proxy variables that have already been shown to have a considerable impact on travel-time behavior. As demonstrated by the analysis above, these were precipitation (Pre), potential evapotranspiration (PET) and saturated soil moisture (SSM) as proxies for influx, outflux and state respectively. Results showed overall the same trend for mean age and life expectancy with respect to these predictors (see Table 1). Precipitation was the most dominant factor for both quantities with the saturated soil moisture being a close second. This is in contrast to e.g., *Benettin et al. (2015a)*, who emphasized the role of the outfluxes for the time evolution of both age and life expectancy. In our analysis, we saw that proxy variables for influx and state showed strongest correlations with mean travel-time behavior. On the other hand, PET, which is a good proxy for one of the two outfluxes, showed only moderately strong correlations with said behavior. In case of mean age, this relationship was even weaker compared to the other two (precipitation and saturated soil moisture). Since we could not provide a proxy variable for the other outflux, i.e., discharge, we excluded this quantity from our analysis.

3.4 Joint impact of multiple variables on mean travel times

In the analysis above, the statistical relationship between mean travel-time behavior and a number of variables was presented and discussed. This was done for every variable individually to elucidate its possible impact on mean travel times. In addition to this simple analysis, we also investigated the joint impact of several variables. Such results can be of relevance for prediction, i.e., using a set of variables to predict travel times in a given CV.

To that end, we used the variables that had been shown to have the highest impact individually, i.e., precipitation, saturated soil moisture and potential evapotranspiration, and performed a multiple linear regression. Simple linear regression had already demonstrated that both precipitation and saturated soil moisture could explain a significant amount of the variability contained in the dataset. Combining these factors could therefore improve the predictability even further. We therefore applied Forward Stepwise Selection to generate a series of models with increasing complexity. The first single-variable model consequently used only precipitation as the variable with the highest single R^2 value. Next, the double-variable model used both precipitation and saturated soil moisture and the most complex three-variable model used precipitation, saturated soil moisture and potential evapotranspiration jointly.

Results for the default case, showed that, compared to using only one variable (precipitation), using two variables for the regression (precipitation and saturated soil moisture) improved the predictability of mean travel times (see Table 2). This was expected since both variables alone provided already high R^2 values. In addition, precipitation and saturated soil moisture did

	Pre	Pre + SSM	Pre + SSM + PET
mean life expectancy	0.860	0.911	0.913

Table 2. R^2 values of for several regression models of increasing complexity.

only show moderate correlation ($R^2 = 0.451$), so adding the latter variable added new information to the prediction model. The correlation that existed between precipitation and saturated soil moisture is explained by the an orographic effect, i.e., hilly regions in the catchment, with typically lower values of saturated soil moisture, also show higher precipitation values. In contrast, using three variables (precipitation, saturated soil moisture and potential evapotranspiration) resulted in almost negligible improvement (see Table 2). This is due to the already lower impact of PET compared to precipitation and saturated soil moisture. In addition, PET showed comparably stronger correlation with both precipitation and saturated soil moisture (data not shown), therefore adding only little new information compared to the other two variables. Such low impact of outgoing fluxes compared to precipitation has already been reported before, for the case of synthetic toy models (*Daly and Porporato, 2006*). Moreover our results agree with the findings of *Hrachowitz et al. (2009)*, who also reported similarly strong explanatory power of climatic variables like precipitation as well as soil and land surface properties.

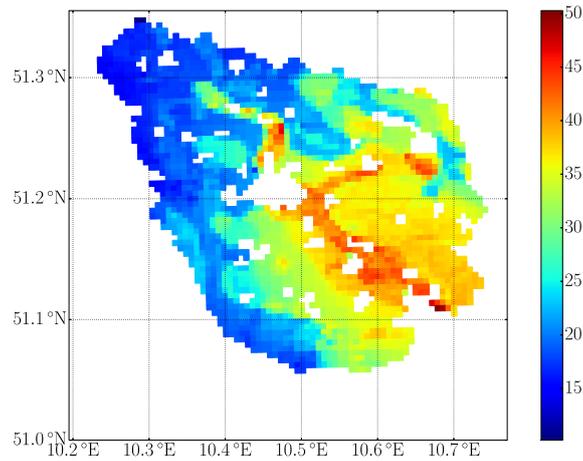
3.5 Impact of hydrological regime on travel-time behavior

The analysis above revealed the strong impact of the influx (i.e., precipitation) as well as the state variable (i.e., saturated soil moisture) on the travel-time behavior. To further elucidate their impact, we investigated travel-time behavior independently for different hydrological regimes during the considered period of time, i.e., from 1955 - 2005. To that end we partitioned the available time series into regimes based on soil moisture (state variable) and precipitation events (influx).

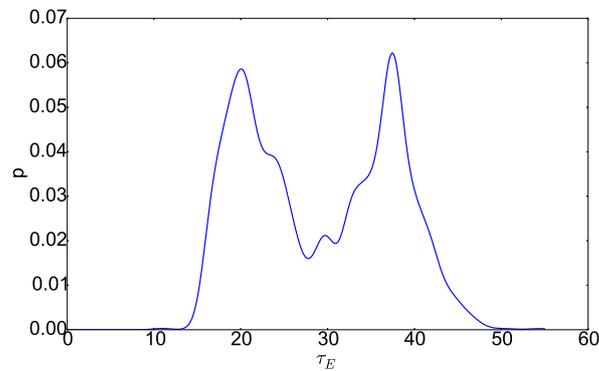
In the first case, we averaged the time series of mean saturated soil moisture in the whole Nagelstedt catchment for every year, i.e., 50 years in total. Next, we divided the resulting time series such that years with an average soil moisture content above 85th percentile of the time series were labeled as wet years. In contrast, years with an average soil moisture content below 15th percentile of the time series were labels as dry years. This annual partitioning was seen necessary due to the strong annual fluctuations of this variable. Finally, we performed the same analysis as describe above for both – now smaller – datasets.

Using results from dry years only (see Figure 18), showed a similar qualitative travel-time behavior but strong quantitative contrast compared to the mean travel-time behavior discussed above (see Figure 8). Compared to the general case, mean life expectancy was much larger in dry years. In addition, dry years exhibit a wider range of possible values with the largest one (over 50 months) being almost 4 times a large as the smallest one (approximately 12 months).

Wet years on the other hand, exhibit a very small range with the smallest value (approximately 5 months) being roughly only half as large as the largest value (approximately 11 months) (see Figure 19). Compared to the general case, where the largest value (approximately 20 months) were roughly 3 times as large as the smallest value (approximately 6 months), these



(a) spatial distribution

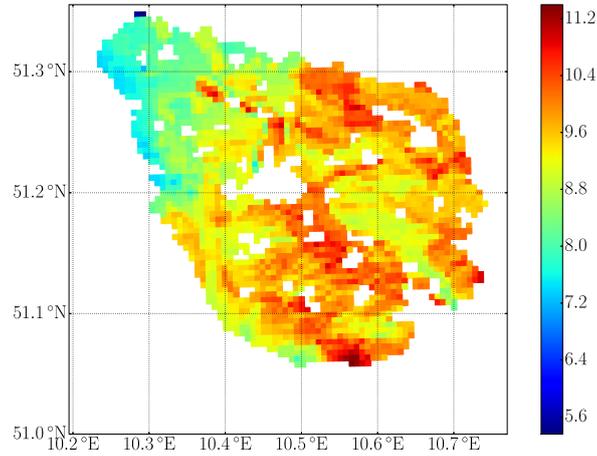


(b) kernel density estimate

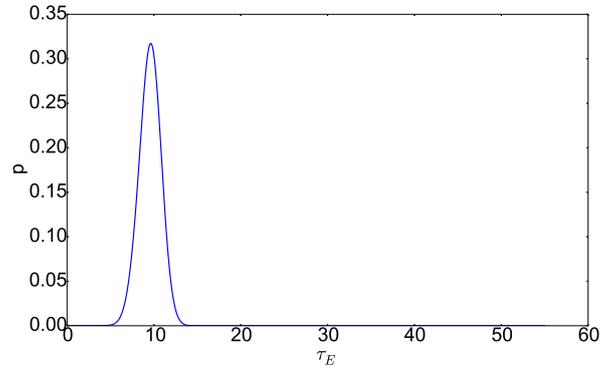
Figure 18. Mean life expectancy ~~(in months)~~ of soil moisture in the Nängelstedt catchment in dry years. Panel (a) shows the spatial distribution and Panel (b) shows the kernel density of mean life expectancy (in months).

two scenarios fall on either side of this spectrum. This stark discrepancy demonstrates again the strong impact of the state variable (soil moisture) on travel-time behavior. Another difference between the mean travel-time behavior in wet years and the general case is the unimodal distribution of the former. The analysis above revealed how the bimodal behavior is mostly due to the different soil types and therefore reflects the strong impact on this property on the overall soil-moisture dynamics.

- 5 The disappearance of this bimodal behavior is therefore reflective of how the soil becomes 'forced into line' when being filled up with water leveling prior differences.



(a) spatial distribution



(b) kernel density estimate

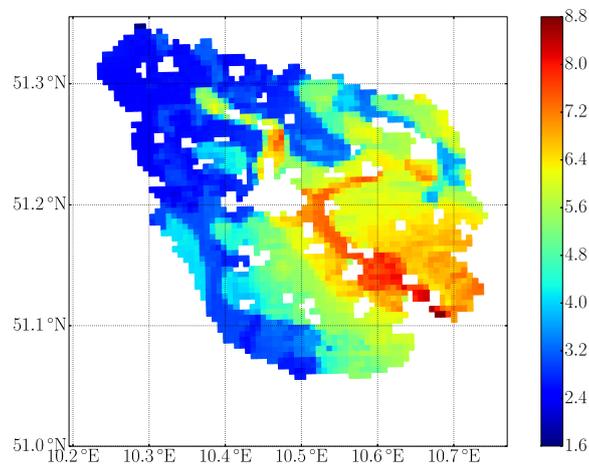
Figure 19. Mean life expectancy ~~(in months)~~ of soil moisture in the Nängelstedt catchment in wet years. Panel (a) shows the spatial distribution and Panel (b) shows the kernel density of mean life expectancy (in months).

	Pre	PET	SSM
wet years	0.374	0.084	0.388
dry years	0.781	0.223	0.834

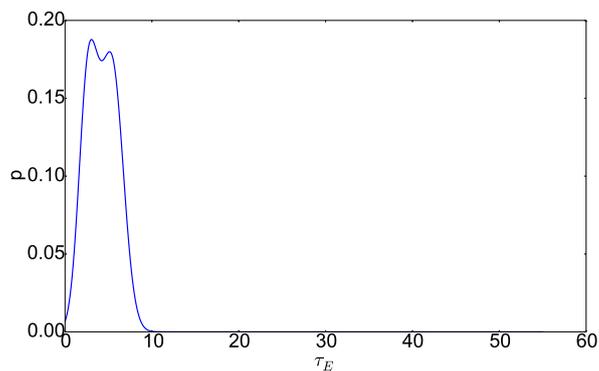
Table 3. R^2 values for several predictors of mean travel time (as caused by wet and dry years).

In addition, results showed different statistical dependency of travel-time behavior with respect to precipitation, PET and SSM (see Table 3). Dry years showed very similar correlation values compared to the general case (see Table 1 a). On the other side, correlation values for wet years were remarkably smaller.

In the second case, we also investigated travel-time behavior depending on influx, i.e., in case of months having above average precipitation values (rainy months). To that end, we constrained our analysis to forward travel-time distributions which were triggered by heavy rain events. This means that, in analogy to the analysis above we only used months with precipitation values above the 97th percentile and performed again the same analysis for the reduced dataset.



(a) spatial distribution



(b) kernel density estimate

Figure 20. Mean life expectancy τ_E (in months) of soil moisture in the Nägelestedt catchment caused by rainy months. Panel (a) shows the spatial distribution and Panel (b) shows the kernel density of mean life expectancy (in months).

- 5 Results showed strong differences in mean life expectancy during rainy months compared to the scenarios discussed above (compare Figure 20 with Figures 8 and 18). Compared to wet years, we saw even lower mean life expectancy. This can be explained by the strong impact of the rain on soil moisture leading to a flushing of the soil. We also saw a similarly small variance and a nearly unimodal distribution of mean travel-time values.

	Pre	PET	SSM
mean life expectancy	0.736	0.221	0.857

Table 4. R^2 values for several predictors for mean travel time (as caused by rainy months).

	all years	dry years	wet years
QI_f	0.150	0.061	0.173
QI_s	0.298	0.239	0.317
QB	0.552	0.700	0.512

Table 5. Relative contribution of the different fluxes to runoff generation.

In addition to that, we saw differences for the statistical correlation of mean life expectancy with precipitation, potential evapotranspiration and saturated soil moisture (see Table 4). Compared to the standard travel-time behavior, precipitation was slightly less explanatory with mean life expectancy. This was caused by lower overall variation in precipitation values, due to constraining our analysis to large values therefore excluding low and medium range rain events. In contrast to that, R^2 values for PET and SSM increased.

3.6 Relevance of ~~TTD's~~ TTD's for hydrological inference

The above results demonstrated the impact of certain soil properties, as implemented in mHM, on mean travel times using the R^2 metric as a measure. In addition to that statistical analysis, their relationship can further be elucidated by analyzing Equation (2) or (3). Assuming for example a very simple linear relationship for both Q and ET with respect to S we get for Equation (2) the following

$$\vec{p}_Q(T_E, t_{in}) = \frac{\alpha_Q}{\theta(t_{in})} \exp(-\alpha_{ET}T_E) \exp(-\alpha_Q T_E). \quad (9)$$

Equation (9) shows how under such simplified assumptions, the TTD of such a CV would follow an exponential distribution with its mean travel time being related to the recession constants α_Q and α_{ET} . As shown above, such an exponential behavior is visible in the mean behavior (see Figure 7 right), whereas non-stationary ~~TTD's~~ TTD's show this exponential behavior to be superimposed by the event-based nature of the governing fluxes (see Figure 7 left).

In addition to these differences, we also saw different mean travel-time behavior for different regimes (see above). These differences can be explained by the actual implementation of Q and ET in mHM, which is generally non linear (see Section 2.2). In order to assess the different roles of each soil process on discharge generation, we calculated the relative contribution of each outflow mechanism for each regime. The data in Table 5 show how much of the water that entered the soil during a given time and left eventually as discharge was leaving as baseflow QB , slow interflow QI_s or fast interflow QI_f . On

	all years	dry years	wet years
R^2	0.6059	0.6954	0.3619

Table 6. R^2 values for recharge vs. mean travel times for different regimes.

average, baseflow contributed the most to discharge with fast interflow having the smallest share. This overall distribution became stronger pronounced during dry years with baseflow taking the largest share of outflow generation and fast interflow becoming negligible. For wet years this trend is reversed, with water entering the soil during rainy months having an almost equal distribution. These different weights show the relative impact and therefore the relative information content that travel-time distributions could contain, i.e., travel times in dry years are mostly the results of the successive processes leading eventually to base flow (see Figure 3), whereas travel times during storm events contain information on all discharge processes combined.

To further elucidate the relationship between the resulting mean travel times and certain model parameters, we performed a regression analysis comparing the recession constant for recharge with the mean travel times for different regimes. Results confirmed the relationship described above with mean travel times during dry years showing the strongest correlation (see Table 6).

Such a high interdependency between certain model parameters and data from different flow regimes is not unique for ~~TTDs~~TTD's. Using discharge alone would reveal similar overall tendencies, e.g., discharge data from droughts is more informative for calibrating baseflow recession constants. What is, however, new is the additional information content, which is not contained in discharge data alone. Not only can this improve calibration efforts, it allows the inference of additional system states. This is in particular relevant, but not confined to, the total amount of stored groundwater. Discharge data are not sensitive to, and therefore not informative for, groundwater levels, but only to its relative changes. ~~TTDs~~TTD's on the other hand, strongly depend on the total amount of water stored in every CV. Using both data types for inference would therefore allow to provide reasonable estimates of this quantity. Similarly, the estimation of water in the root and vadose zone can be improved.

In addition *Birkel and Soulsby (2015)* highlight the temporal aspects of travel times on model calibration. They point out, how the sampling frequency of the time series should match the expected travel times of the underlying process. Our results above revealed different time scales for different hydrological regimes, that differed by almost an order of magnitude. Despite this heterogeneity, all travel times in our study remained in the range of months. Under such circumstances, a high resolution measurement campaign with daily or even hourly intervals would not be necessary.

Although the above explanations provide only a limited perspective on the relationship between ~~TTDs~~TTD's and model parameters, it can be said that the strong interlink between the travel-time behavior and outflow generation indicates the high information content of the former with respect to the latter. As a result, travel-time distributions should be regarded as highly informative for the calibration of hydrological models. As mentioned in the Introduction, *McDonnell and Beven (2014)*

have made the case for the usefulness of ~~TTDs~~-TTD's for the parametrization of such models. The above presentations provide empirical support for this notion.

4 Conclusions

In this study, we investigated ~~soil behavior~~ the spatially-distributed soil-moisture dynamics in the Nägelstedt catchment by virtue of travel-time distributions. The states and fluxes, needed for the derivation of the travel times, were numerically computed using the mesoscale Hydrological Model (mHM), which was calibrated against 55 years of discharge data as well as using detailed data on soil properties, land cover and precipitation. We performed a statistical analysis of mean travel times to describe the soil response decoupled from the event-driven impact of precipitation.

Comparing the derived mean travel times for several modeling scales (spanning over one order of magnitude), we did not see any significant difference in their distribution. This indicates ~~the~~ a general soundness of the parametrization scheme of mHM used for the calculation of the states and fluxes on the different modeling scales. Our analysis shows that precipitation, saturated soil moisture and potential evapotranspiration are strong statistical predictors of mean travel time behavior. We also note that on average shorter mean travel times correspond to forested area and larger ones to crop/grassland, an observation that we linked to both correlations between forested and high-precipitation areas as well as the different water uptake mechanisms of trees vs. crop/grass.

We also investigated the travel-time behavior for different hydrological regimes, i.e., for dry and wet conditions (using soil moisture and precipitation as indicators). Our analysis revealed significantly different travel-time behavior for each of these regimes. Despite the strong heterogeneity of soil properties as well as (to a lesser extent) precipitation values, we could discriminate these regimes also in the resulting distribution of mean travel times.

Under dry conditions, we saw mean travel times having a pronounced bimodal distribution with long mean travel times and large variance. Such long travel times reveal the strong impact of baseflow on the generated outflow, whereas the large variance shows the variety of soil responses under dry conditions. Such conditions are therefore suited for inferring soil properties relating to baseflow generation. In addition, due to the large variance of soil responses, such conditions would allow to infer the spatial origin of solutes found in discharge streams. Such inferences are however, hampered by the long travel times involved. Not only are long time series needed, measurements must also being performed during such dry conditions.

Under wet conditions, we saw mean travel times having a unimodal distribution with shorter mean travel times and a smaller variance. This shorter travel times are caused by a larger influence of the slow and fast interflows on the total discharge behavior. As a result, ~~TTDs~~-TTD's derived under such conditions may be suited for inferring the parameters relating to these hydrological processes.

In case of rainy months, which overlap with wet conditions to a significant degree, we saw a similar distribution of travel times, but with even shorter mean values. This indicates a stronger impact of fast interflow on the total discharge behavior. Such information can therefore be valuable for improving the parametrization of the fast interflow related processes.

It is important to emphasize that our results have been derived with respect to a single hydrological model, i.e., mHM, only. ~~However, due~~ As a result, we also need to critically assess the limitations of this approach and its impact on the reliability of our conclusions. First, mHM treats the hydrological storage in every compartment as fully mixed. In the absence of additional information, we consequently assumed a uniform sampling scheme for the discharge generation from every mHM cell. This may have introduced errors in the age distribution of fluxes and therefore the travel-time behavior as discussed in Section 2.1. Due to the well established ability of mHM to take subgrid heterogeneity into account, we have ~~good~~ confidence in the physical plausibility of the spatially explicit soil moisture states and fluxes. ~~The framework we used here is general and can be used in conjunction with other hydrological/land surface models applied in other catchments. Moreover, future studies may incorporate other explanatory variables for the spatial heterogeneity of~~ TTDs. In the absence of, say solute data, we have, however, to consider these assumptions as tentative and open to revision. The other limitation of our approach stems from the computational nature of our study introducing a number of uncertainties. Like any hydrological model, mHM may suffer from three different sources of uncertainty; input uncertainty, structural uncertainty and parametric uncertainty. We would therefore like to assess their nature and potential impact on our results and conclusions. First, input uncertainty is referring to the uncertainties inherit in the forcing of the model, i.e., precipitation. Our results have shown the strong impact of precipitation on travel-time behavior. It would therefore stand to reason to expect a strong impact of any uncertainty from precipitation to propagate to the resulting travel-time behavior. However, we investigated mean behavior only, where time series from many months were averaged. We therefore consider possible contributions to our results to be minor. Next, structural uncertainty depend on the conceptual implementation of subsurface processes within mHM and our choices of different mHM compartments for our analysis. In Section 2.2, we discussed this issue by providing the rationals for, e.g., including the interflow components into our analysis. Finally, parameter analysis is probably the largest total source of uncertainty and several studies have recently investigated its impact on mHM output generation (Samaniego et al., 2013; Cuntz et al., 2015; Livneh et al., 2015). The studies show that, while the fluxes are typically well represented in mHM (Livneh et al., 2015), the overall soil moisture storage showed less accuracy, in particular during droughts Samaniego et al. (2013). For droughts, our results showed in general long travel-times and pronounced soil specific behavior with comparably lesser impact of precipitation. While we do not expect a major impact on the qualitative nature of these results, we should consider the quantitative aspect, i.e., the specific values for mean travel times to be inconclusive. In general, we consider the uncertainty stemming from the storage estimate to be the most relevant due to having both comparably lower accuracy and the strong impact on overall travel-time behavior demonstrated above. This is exacerbated since the water content relevant for outflow generation may not be the same as the one relevant for travel-time behavior. Immobile water due to, e.g., dead-end pores, affects the latter but not the former. It is, however, this connection between the total water content and the resulting travel-time behavior that makes the use of TTD's an important tool for a better calibration of hydrological models.

~~Having~~

As an outlook, we can say that, having established a comprehensive description for the storage and release of water in the investigated catchment, the natural next step is the integration of reactive solute transport. As demonstrated by e.g.,

Botter et al. (2010), the concept of travel-time distributions can directly be adapted to account for the transport of both conservative and reactive solutes. This extension would facilitate to compare our predictions with the wealth of data that has been and continues to be collected within the AquaDiva center at the Hainich Critical Zone Exploratory (*Küsel et al.*, 2016). Thereby, we will be able to test our predictions by virtue of a large data set as well as initiate the collection of additional new data.

Appendix A: Forward and backward formulation of travel times

Both the forward and backward formulations for ~~FFDs~~ TTD's can be derived from Equation (1) by additionally associating each term with its distribution, so

$$\frac{d}{dt}[S(t)p_S(T,t)] = J(t)p_J(T,t) - ET(t)p_{ET}(T,t) - Q(t)p_Q(T,t). \quad (\text{A1})$$

Here T is a placeholder for either the age or life expectancy of the water parcel. The total derivative in Equation (A1) can be reformulated using the material derivative, so

$$\left(\frac{\partial}{\partial t} + \frac{d}{dt}T \frac{\partial}{\partial T} \right) [S(t)p_S(t,T)] = J(t)p_J(t,T) - ET(t)p_{ET}(t,T) - Q(t)p_Q(t,T). \quad (\text{A2})$$

Equation (A2) is the general PDE describing the time evolution of the age of the water in the CV. It is worthwhile to note that there is a significant inconsistency in the literature with respect to this equation. *Botter et al.* (2011) discuss the backward formulation of Equation (A2), while referring to it as the Master Equation (ME). This is certainly justified given that the ME is describing the time evolution of the PDF of any Markov process, i.e., a stochastic process that is local in (chronological) time. This condition is true for Equation (A2). In addition, Equation (A2) is not only local with respect to chronological time t but also with respect to the travel time T . Interpreting T as x , it becomes obvious that Equation (A2) is analogous to the much simpler Fokker-Planck equation or, since there is no 'diffusion', the even simpler advection-reaction equation. On the other side, *Porporato and Calabrese* (2015) are careful to trace this equation back to the seminal work of both *McKendrick* (1925) and *von Förster* (1959) in population dynamics. Consequently, they call this equation the McKendrick-von Förster (MKVF) equation.

One problem with Equations A1 and A2 is the lack of closure, i.e., $p_S(T,t)$, $p_J(T,t)$, $p_{ET}(T,t)$ and $p_Q(T,t)$ are different variables. The solution to this problem is the formulation and/or derivation of a dependency, i.e., closure, between the storage and the fluxes through

$$p_F(T,t) = \omega(T,t)p_S(T,t). \quad (\text{A3})$$

This closure function $\omega(T,t)$ must follow some properties to ensure the normality of both $p_S(T,t)$ and $p_F(T,t)$, with the latter being the PDF of a flux, i.e., effective precipitation, evapotranspiration or discharge. These closure functions are

called StorAge Selection (SAS) functions in the literature (*Rinaldo et al.*, 2015). Several different formulations exist with the one given above being based on the work of *Botter* (2012).

The shape of the SAS function is determining the preference of the fluxes, e.g., discharge, for several ages of the water stored in the CV. In the backward formulation a flat function would correspond to no preference with respect to age, a
5 monotonously decreasing function would correspond to a preference for younger water and a monotonously increasing function would correspond to a preference for older water

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10 Federal Institute for Geosciences and Natural Resources (BGR), the European Water Archive, the Global Runoff data Centre, the project CarboEuropeIP (EU-FP6) as well as Axel Don for the provision of eddy covariance data. .

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