

RESEARCH ARTICLE

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Key Points:

- A high-resolution distributed groundwater model was used to calculate dynamic travel time distributions and StorAge Selection functions
- Dynamic TTDs were controlled by the activation of shallow flow paths and the intensification of all fluxes when groundwater levels rose
- SAS functions show a preference for the discharge of young water and vary in time as a result of different catchment characteristics

Supporting Information:

- Supporting Information S1
- Supporting Information S2
- Figure S1
- Figure S2
- Figure S3
- Figure S4
- Figure S5

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Transient Groundwater Travel Time Distributions and Age-Ranked Storage-Discharge Relationships of Three Lowland Catchments

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Abstract The contribution of groundwater to streams is controlled by temporally and spatially variable groundwater flow paths with distinctive travel times. The aggregated average travel time distribution (TTD) of all these flow paths functions as a catchment characteristic. Currently, research on TTDs is expanding towards dynamic TTDs and building on this, we present dynamic backward TTDs and residence time distributions using forward particle tracking on a high-resolution spatially distributed groundwater flow model (25*25 m). We show that the dynamic backward TTDs of three Dutch catchments are determined by the interplay between the activation of shallow short flow paths and the intensification of fluxes through all flow paths when groundwater levels rise. In addition, the preference for young water in our lowland catchments appears strongly controlled by drainage density. Variations in catchment mixing with time and between catchments were analyzed using dynamic StorAge Selection (SAS) functions. This showed the effect of differences in geology and topography on the shape of the SAS functions. Additionally, the variability of SAS functions in time was shown to depend on the extent to which new flow paths can be activated. Time-varying SAS functions are required for computation of dynamic TTDs, and this research showed realistic values for the variability in the SAS functions of lowland catchments. The step towards dynamic TTDs is crucial for understanding the temporal and spatial behavior of streams, their chemical composition, and their ecological value.

1. Introduction

Groundwater creates a delay in the precipitation-discharge response of a catchment, distributes water spatially, and influences stream water quality (Kaandorp et al., 2018). In the sandy and intensively drained lowland catchments of the Netherlands, a lack of topography and hard rock promotes infiltration of precipitation into the soil. Therefore, the majority of streamflow originates from groundwater (De Vries, 1994; Wriedt et al., 2007; van der Velde et al., 2011; Hendriks et al., 2014). Consequently, the input of groundwater to lowland streams is a key element that influences stream ecological and chemical functioning. It is especially in these lowland catchments that the groundwater system is characterized by a wide range of groundwater flow paths and travel times (TTs), which control the timing and quality of water that discharges to the surface (Martin et al., 2004; van der Velde et al., 2010; Hamilton, 2012). The multiyear average distribution of these TTs is a complex catchment characteristic that includes information about storage, climate, and flow paths (Gardner et al., 2011; McDonnell et al., 2010; McGuire & McDonnell, 2006; Visser et al., 2009).

While most catchment studies report whole-catchment TT distributions (TTDs), in the current study we specifically focus on the TT of the groundwater contribution to streamflow. A disconnect seems to exist between catchment and groundwater TT communities. Whereas catchment studies using lumped models and stable isotopes often focus on short TTs of days and months (e.g., Birkel et al., 2012; Dunn et al., 2010; Peralta-Tapia et al., 2016) the TTs studied by the groundwater community using groundwater models and tracers such as dissolved gases and radioactive isotopes are generally in the order of years and decades (e.g., Basu et al., 2012; Eberts et al., 2012; Gilmore et al., 2016; Solder et al., 2016; Stewart & Morgenstern, 2016; Visser et al., 2009; Visser et al., 2013). The coupling between groundwater TTDs and stream discharge is especially used in the assessment and prediction of stream discharge from (nitrate) polluted aquifers (e.g., Böhlke & Denver, 1995; Duffy & Lee, 1992; Zhang et al., 2013). The age of groundwater seepage was studied by Broers (2004), who used a model to show that a higher drainage network density leads to larger spatial

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variability in groundwater ages. Gilmore et al. (2016) used a combination of noble and other dissolved gases to show that there is significant variation in the age of seepage within a stream profile, which was also found in an earlier study by Modica et al. (1998). In addition, they hypothesized that TTDs are affected by spatial variation in groundwater recharge and that young water mainly recharges at locations such as ditches and tributaries. Because of the variable recharge and heterogeneity in for example soils and slopes, the contributions of groundwater flow paths vary both in time and space (Engdahl et al., 2016; Gardner et al., 2011; Rozemeijer & Broers, 2007; van der Velde et al., 2009; Visser et al., 2007).

The time variation in groundwater TTs can be represented by dynamic TTDs (Botter et al., 2010; Engdahl et al., 2016; Harman, 2015; Heidebüchel et al., 2012; van der Velde et al., 2012; van der Velde et al., 2010) and occurs due to external variability of the input (i.e., groundwater recharge) as well as internal variability where shallow flow paths become active as the groundwater table rises (Harman et al., 2016; Kim et al., 2016; Rozemeijer & Broers, 2007). Additional information about flow paths combined with the contribution of different ages to streamflow allows direct correlation of, for instance, water chemistry with a specific flow path, which can thus explain variations in water chemistry throughout the year (Benettin et al., 2013; Benettin et al., 2017; Hrachowitz et al., 2016). The effect of flow paths contributing to streams has already been included in some TTs studies using particle tracking approaches (Basu et al., 2012; de Rooij et al., 2013; Gusyev et al., 2014; Modica et al., 1997; Molénat & Gascuel-Oudou, 2002; Visser et al., 2009); however, these studies focused on stationary or summer/winter TTDs. To date, particle tracking approaches have not been used to characterize dynamic TTDs for real catchments.

The TTD of the discharge is related with the ages in the storage of a catchment. The distribution of ages in storage, referred to as the residence time distribution (RTD), is variable in time like the TTD due to changes in recharge to and discharge from storage (Botter et al., 2011; Harman, 2015). The relationship between the ages of discharge (TTDs) with the ages of water in the catchment storage (RTDs) can be described by StorAge Selection (SAS) functions (Benettin et al., 2015; Harman, 2015; Hrachowitz et al., 2016; Rinaldo et al., 2015; van der Velde et al., 2012). SAS functions are spatially and volume integrated and not directly affected by precipitation and evapotranspiration fluxes and storage changes. Therefore, SAS functions purely describe the mixing/selection of the outflows from storage, independent of weather conditions. Based on this, we expect SAS functions to correlate more strongly to landscape properties than TTDs and to be a useful metric to characterize storage-discharge behavior of catchments.

Our aim is to explore both the temporal and spatial groundwater contributions to stream discharge. We attempt to find the processes affecting variable TTs and study their behavior, and try to link this behavior with landscape characteristics. To this end, a high-resolution spatially distributed groundwater flow model with forward particle tracking is used to characterize and compare three head water streams of the Dinkel lowland catchment in the Netherlands by their dynamic TTDs, RTDs, and SAS functions. Building on the stationary TTD approach outlined by Gusyev et al. (2014), for the first time we use a transient model approach to capture flow path variations throughout the year of multiple catchments. According to Hrachowitz et al. (2016), “there is growing evidence that mixing properties of the flow domain may be subject to temporal heterogeneity,” and as such we compare catchment mixing behavior, using dynamic fractional SAS functions (Harman, 2015; van der Velde et al., 2012), in time and between catchments.

2. Materials and Methods

2.1. Study Area

We selected three head water streams of the Dinkel river in the east of the Netherlands that have different land use, hydrological, and morphological characteristics (Figure 1). The area has a temperate marine climate with a mean annual precipitation of 800 to 850 mm, an actual evaporation of 560 mm/year on average and a mean temperature of 9.6 °C. The Dutch part of the Dinkel catchment has substantial height differences, ranging from 80 m above sea level at the top of the moraines to about 18 m above sea level in the river valley. The geohydrology of the study area is characterized by shallow aquifers (1–20 m) located on top of clayey moraines. In addition, the lower parts of the area are also filled with sandy aquifers.

The study area is characterized by an extensive system of watercourses (Kuijper et al., 2012). Land use is intensive and consists primarily of agriculture (60–70%) and partly of urban area and nature (forest and natural grasslands). The catchments are intensively drained and the ground and surface water is used for

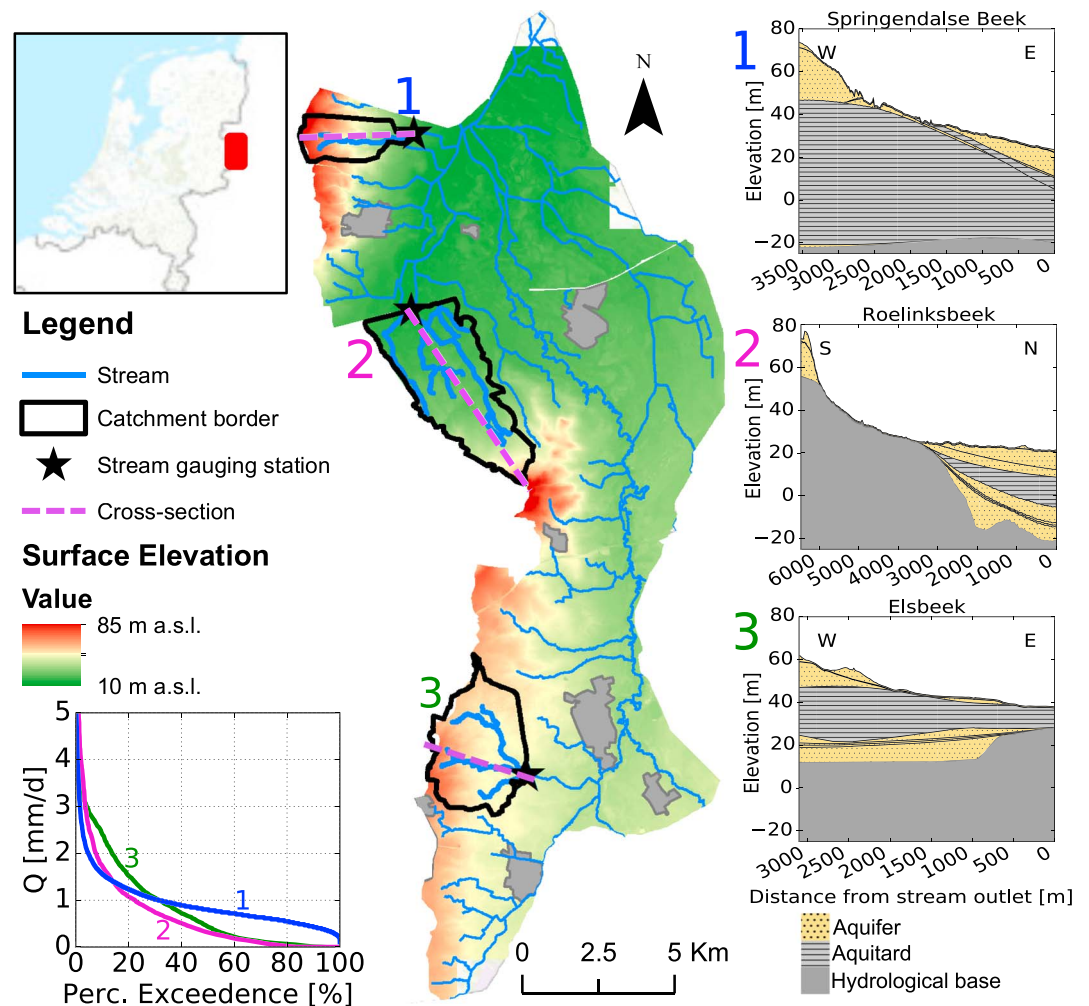


Figure 1. Location of the study catchments in the Dinkel river valley: (1) Springendalse Beek, (2) Roelinksbeek, and (3) Elsbeek. Measured flow duration curves of the catchments are shown in the left bottom corner. The dashed lines in the map represent the cross sections of the geohydrological model buildup of the three catchments, which are shown to the right. Note that the boundaries (1–3) in these figures are not the actual boundaries of the model, which is much larger and encompasses all three catchments.

irrigation, industry and drinking water. Many streams have been altered, mostly to aid agriculture for which weirs have been installed and streams have been straightened and deepened (Kuijper et al., 2012). During the last few decades, measures have been taken to re-naturalize the rivers and streams.

The selected catchments are the Springendalse Beek, Roelinksbeek, and Elsbeek, with sizes of approximately 4, 12, and 11 km², respectively. These streams were chosen based on the available data as well as their distinctive discharge characteristics. The Springendalse Beek has an average discharge of about 0.043 m³/s, the Roelinksbeek of 0.093 m³/s, and the Elsbeek of 0.104 m³/s. The variation of the discharge throughout the year also differs between the streams; the Roelinksbeek has the highest peaks while the Springendalse Beek has a fairly stable discharge and is the only stream that doesn't run dry. Base flow index (Gustard et al., 1992) is 0.8 for the Springendalse Beek and 0.4 for the other two streams. Land use varies: The valley of the Springendalse Beek is mostly forested upstream, while in the Roelinksbeek and Elsbeek catchments agriculture is the predominant land use. In these two catchments the drainage density is therefore around 32% while it is 9% in the Springendalse Beek catchment. Tile drains are located in only 2% of the area in the Springendalse Beek, and in respectively 8% and 13% of the catchments of Roelinksbeek and Elsbeek. Geohydrology is also different: The Springendalse Beek is fed by springs and seepage lakes, while the Roelinksbeek and Elsbeek only have a few distinctive springs.

2.2. Groundwater Model of the Study Area

Using a finite-difference MODFLOW (Harbaugh, 2005; McDonald & Harbaugh, 1988) groundwater model created and calibrated by Kuijper et al. (2012) and described by Hendriks et al. (2014), we simulated groundwater flow on a daily time step in the Dutch part of the Dinkel catchment. The total modeled area was 58 by 45 km, divided into cells of 25*25 m, and contained the three study catchments. The top of the model followed the surface elevation. The vertical model discretisation was based on the Dutch Geohydrological Information System (REGIS II, 2005) and comprised seven layers of variable thickness. A spatially averaged lithological representation is shown in Figure 1 for the three studied subcatchments. Note that these cross-sections represent only parts of the total groundwater flow model and as such the edges of the figures are not model boundaries. Transmissivity in the aquifers was approximately 40 m²/day for the Springendalse Beek, 23–30 for the Roelinksbeek and 10–40 for the Elsbeek. Resistivity of the aquitards in the Roelinksbeek and Elsbeek catchments was 24,000 and 5,200 days, respectively. The ice pushed ridges had some anisotropy (Kuijper et al., 2012) and the porosity of the aquifers was assumed to be 0.3. The entire drainage system including springs, tile drains and ditches was modeled using the DRN and RIV MODFLOW packages. For more information on the functioning of these packages, readers are referred to Harbaugh (2005). The model included an unsaturated zone module, which calculated the groundwater recharge using the precipitation, evaporation, land use, and information about the soil and vegetation (MetaSWAP, van Walsum & Groenendijk, 2008; van Walsum & Veldhuizen, 2011; De Lange et al., 2014).

In order to calculate long groundwater flow paths a spin-up period was needed which was chosen to be approximately 300 years. Therefore, the modeled period was extended to 1700–2010 using continuously repeated climate data from 1965 to 2011 (data from the Royal Netherlands Meteorological Institute KNMI). The model was calibrated on measured groundwater heads in 976 piezometers (Kuijper et al., 2012). A steady-state calibration of the transmissivity and conductances was followed by a transient calibration of river conductance and storage coefficients. After calibration, the average difference between modeled and measured groundwater levels was 0.11, 0.06, and 0.15 m for model layers 1, 2, and 3 respectively. We validated the model for river discharge using the Nash-Sutcliffe efficiency (NSE) coefficient (Nash & Sutcliffe, 1970) to evaluate its functioning. No further calibration and validation was done.

2.3. Travel and RTDs

The water balance of the groundwater in a catchment is given by:

$$\frac{dS}{dt} = R - ET_{GW} - Q \quad (1)$$

where R is groundwater recharge, ET_{GW} represents evapotranspiration and capillary flow directly from the groundwater, dS/dt is the change in groundwater storage, and Q is groundwater discharge (streamflow). For these parameters we use the unit “mm/day,” which we calculated by dividing over the catchment area. This way the different catchments and the values of infiltration and seepage are directly comparable. Apart from R , which has an age of 0 by definition, each component in equation (1) has a specific TTD, as described mathematically by Botter et al. (2011), van der Velde et al. (2012) and Harman (2015). Note that with TTD we mean the backward TTD of the groundwater contribution to streamflow, which indicates the distribution of TTs in stream water, as was nicely described by Benettin et al. (2015). Although unsaturated zones can be important for total catchment TTDs (Sprenger et al., 2016; Green et al., 2018) our goal was to understand functioning of the groundwater storage and groundwater discharge and therefore our focus was on groundwater TTs and time scales. In addition, unsaturated zones are generally thin in the study area (maximum a few meters, but mostly <1 m).

We consider cumulative age distributions which are described by equations (2)–(4):

$$\text{RTD of } S : P_S(T, t) = \frac{s(\tau < T, t)}{S(t)} \quad (2)$$

$$\text{TTD of } Q : P_Q(T, t) = \frac{q(\tau < T, t)}{Q(t)} \quad (3)$$

$$\text{ET}_{GW} : P_{ET_{GW}}(T, t) = \frac{et_{GW}(\tau < T, t)}{ET_{GW}(t)} \quad (4)$$

where $s(\tau < T, t)$ is the storage with TT τ smaller than T at time t , $q(\tau < T, t)$ is the discharge with TT τ smaller than T at time t , and $et_{gw}(\tau < T, t)$ is the evapotranspiration flux from the groundwater with TT τ smaller than T at time t . ET_{GW} is assumed to always sample the most recent rainfall still present in storage S (as in e.g., Heidbüchel et al., 2012).

TTDs and RTDs were constructed using a combination of particle tracking and volume “book keeping” of the evapotranspiration. The TTs and flow paths were calculated transiently using the flow velocities of the MODFLOW groundwater flow model and the particle tracking software MODPATH version 3 (Pollock, 1994). Particles released at the groundwater table at the center of every model grid cell on the first day of every month and given a volume equal to the total groundwater recharge in that cell in the previous month. This was done for a period of 310 years yielding a total of 3,732 particle tracking runs, which depending on the catchment size had between 14 and 58 thousand particles. This monthly setup was chosen for practical reasons to significantly reduce calculation times at the cost that TTs shorter than a month become unreliable. Every month during the period 1990–2010 the particles that end up in the stream network are collected and used to derive the TTD, while the particles that reside in the subsurface are used to derive the RTD. Because a spatially distributed model is used, it will be possible in further analysis to zoom in to subcatchments and construct their TTDs and RTDs.

The groundwater catchments were delineated using the extent of the starting locations of the particles that end up in the streams. In particle tracking using MODPATH, the handling of weak sinks has to be considered (Abrams et al., 2012; Visser et al., 2009) and in our calculations particles were stopped if the fraction of discharge to the sink was larger than 50% of the total inflow to the cell. For comparison, particle tracking was done with weak sink fractions of 0.3, 0.5, and 0.7 which indicated that the overall TTD was not significantly influenced by this choice.

All particles were given a volume, based on the total monthly summed groundwater recharge at their location, which was calculated by the groundwater model.

$$V_{part}(0, t_i) = \sum_{\tau=t_i-dt}^{\tau=t_i} R(\tau) \quad (5)$$

where $V_{part}(0, t_i)$ is the volume of a particle starting at time t_i with TT 0, dt is a monthly time step, and $R(\tau)$ is the groundwater recharge at time τ . To ensure that the volume balance of the particles matches the water balance of the groundwater model water, capillary rise, and evaporation from groundwater need to be accounted for separately. Evapotranspiration fluxes rarely sum up to more than 50% of the storage of a grid cell and consequently particles will not be stopped due to evapotranspiration. Hence, particle volumes were corrected by subtracting the capillary rise flux (negative recharge) from the volume of the most recently started particles while ensuring that particles do not get a negative volume.

All particles have a residence time T , which is the time spent as groundwater, and a maximum TT T_Q , which is the TT at the moment of discharge to the surface water:

$$T(t) = t - t_i \quad (6)$$

$$T_Q(t_Q) = t_Q - t_i \quad (7)$$

where $T(t)$ is the residence time of a particle at time t , t_i is the time of starting of the particle, and $T_Q(t_Q)$ is the TT at the time of discharge t_Q of the particle.

After their release, these particles were tracked forward through the aquifers up to the point of discharge. The results of the separate particle tracking calculations were combined for which all particles contributing to the streamflow were summed for each month.

$$Q(t) = \sum_{\tau=t_i}^{\tau=t_i+dt} V_{part}(T_{max}, \tau) \quad (8)$$

This monthly discharge, determined in addition to the discharge which resulted from the water balance of the groundwater model (sum of RIV and DRN), was used as a check for the TTD model approach: Volumes

discharging from particle tracking should yield approximately the same discharge fluxes as calculated from the groundwater model. To construct the TTD, the particle volumes were used to weight the contribution of TTs to the discharge using equation (3).

The RTD of storage was calculated by summing all groundwater particles that started but had not turned into surface water discharge yet. The sum of their volumes represented the total storage and the RTD was made by volume weighting the particle ages (equation (2)). The 300 years spin up was required to fill up the storage with particles and derive a stable storage volume.

2.4. Age Fractions, Median TT, and MTTs

Because we calculated the full TTD, ranging from the youngest to the oldest water, the contribution of water with specific ages can easily be deduced. However, because of the small grid cells of 25*25 m we do not quantify part of the shortest flow paths. For the TTDs this implies that our results focus on the older and longer flow paths and that the contribution of young water is likely underestimated. Therefore, the youngest discharge fraction we assumed was <1 year and we divided the discharge into three fractions: young (TT < 1 year), medium aged (TT 1–25 years) and old (TT >25 years). In addition to the age fractions, the median TT and mean TTs (MTT) were calculated for each month. Although the MTT is used the most often in literature, the median TT is more descriptive, as it is less influenced by the long tail in TTs, and thus less affected by single particles with very long TTs.

2.5. SAS Functions

Cumulative SAS functions, Ω , describe which water from storage becomes discharge based on the cumulative TTD of discharge as function of the cumulative RTD of storage:

$$\Omega(P_S(\tau < T, t), t) = P_Q(\tau < T, t) \quad (9)$$

The graphical shape of the SAS function is obtained by plotting $P_S(\tau < T, t)$ (equation (2)) on the x-axis against the corresponding $P_Q(\tau < T, t)$ (equation (3)) on the y-axis (Figure 2) (Hrachowitz et al., 2016). For any flux leaving a catchment SAS functions describe the preference for discharging younger water or older water from storage or no preference (uniformly selected; Hrachowitz et al., 2016; van der Velde et al., 2012; van der Velde et al., 2015; Figure 2).

To compare the different catchments and to link landscape characteristics with simple parameters, we described the SAS functions with a two parameter cumulative beta distribution function $I_x(a, b)$ which we then combined into a single parameter. Two parameter cumulative beta distribution functions allow a wide variety of shapes and range from 0 to 1 on both axes (van der Velde et al., 2012). The parameter “a” mostly describes the left side of the shape of the cumulative SAS function, which is the youngest water, while parameter “b” mostly influences the right part of the shape, which is the oldest water. To obtain a single parameter, the cumulative beta distribution “a” was divided by the different values of “b.” Like the cumulative beta distribution “a,” this a/b value indicates whether there is a preference for young or older water, where $a/b < 1$ means a preference for younger water (median TT < median TT uniformly selected catchment), $a/b = 1$ means no preference (uniformly selected), and $a/b > 1$ means a preference for older water (median TT > median TT uniformly selected catchment; Figure 2). More information on the meaning of a/b is given in supporting information S1.

The SAS functions were first described by cumulative beta distributions with a time variable $a < 1$ and a fixed “b” = 1, as was done in studies by van der Velde et al. (2012, 2015) and Danesh-Yazdi et al. (2016). For the Elsbeek and Springendalse Beek this procedure did not lead to an acceptable fit. Therefore, the Springendalse Beek was described with a different fixed value for “b” (1.4) to better describe the contribution of old water (supporting information S1). Due to this, the a/b value of the Springendalse Beek is different than the value of “a”, contrary to that of the Roelinksbeek. For the Elsbeek, a single cumulative beta distribution was not sufficient. Therefore, based on our physical understanding of the catchment, we used a combination of two cumulative beta distributions to represent a shallow and deep aquifer. The shallow reservoir was described in the same way as the Roelinksbeek with a “b” of 1, and the deep reservoir was described as uniformly selected. These reservoirs were combined using an exponential and time-variable mixing factor. For the Elsbeek catchment, we only compare the contribution of the shallow aquifer with the other

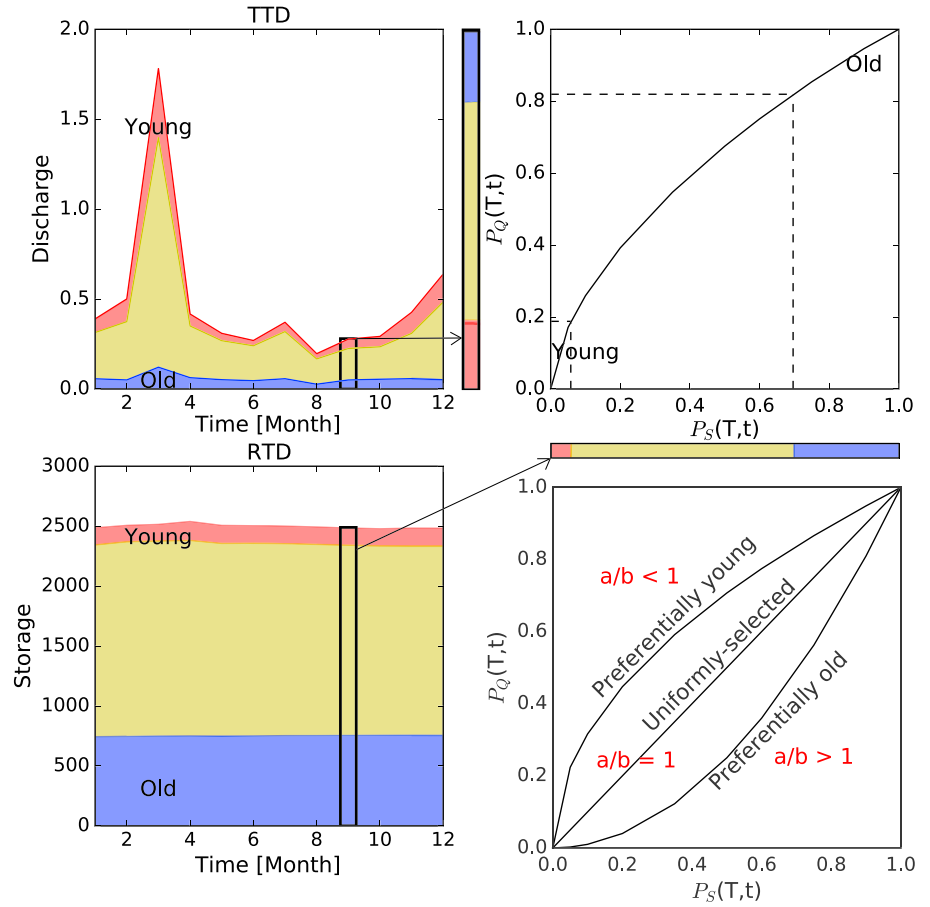


Figure 2. Construction of StorAge Selection functions from discharge travel times and storage residence times. The shape of the SAS function shows the preferential discharge of younger or older water from catchment storage and can be described using cumulative beta distribution $I_x(aQ, bQ)$. This figure contains illustrative data only, not model results. RTD = residence time distribution; TTD = travel time distribution.

catchments, as the combination of the two reservoirs is not directly comparable. A detailed overview of the parameters used in fitting the SAS functions of the different catchments is given in supporting information S1.

In this study, the SAS function of ET_{GW} was represented by a step function (equation (10)), assuming that the youngest water is always used first:

$$\Omega_{ET_{GW}}(P_S, t) = P_{ET_{GW}}(T(P_S, t) \cong H(0)) \quad (10)$$

where $\Omega_{ET_{GW}}$ is the SAS function of the evapotranspiration from groundwater.

3. Results

3.1. Groundwater Flow

The model performance was evaluated by comparing the measured discharge with the computed discharge. With NSE coefficients between 0.50 and 0.92, the model provides a reasonable fit and grasps the variation in runoff characteristics between the catchments due to the differences in catchment shape, drainage network, topography and geology (Figures 3a–3c). We see a small overall underestimation of the discharge in the Springendalse Beek and in all catchments peak flows tend to be underestimated (Figures 3a–3c). This likely relates to surface runoff, macropores, and shallow preferential flow paths that are typically underrepresented by groundwater models (Beven & Germann, 1982). Overall, the model reproduces the dominant hydrological

behavior of the three catchments well, especially when taking into account that this model has been calibrated on groundwater heads only (Kuijper et al., 2012). Stream discharge calculated with the particle tracking approximately matched the discharge fluxes calculated by the groundwater model (see supporting information S2), although slight differences occurred due to difficulties in delineating the fluctuating groundwater catchments.

3.2. Catchment TTDs, RTDs, Fractions, and MTTs

The Springendalse Beek and Roelinksbeek have TTDs with approximately the same shape, although the temporal variation in the TTD is larger in the Roelinksbeek (Figures 3d–3e). The majority of the water is younger than 5 years old and there is a large tail with older water. The TTD of the Springendalse Beek changes between seasons and shows that more young water is discharged in winter than in summer. In the Roelinksbeek it seems that flow in all flow paths increased in equal quantities, leading to a shape of the TTD that is not significantly different between summer and winter (Figure 3e). For the Elsbeek the shape of the TTD differs from the other catchments (Figure 3f). This is because it receives water from a shallow unconfined aquifer and from a deeper second aquifer (Figure 1), which provides relatively young and old water respectively.

A major part of the fluctuation in the discharge volumes of the catchments is caused by the younger fractions, although variation also occurs in the oldest fraction of discharge (Figures 4b, 4d, and 4f). The Springendalse Beek mainly discharges medium aged water (TT 1–25 years) and shows a seasonal pattern with older water in summer and younger water in winter (Figure 5). The young water fraction of discharge (<1 year old) increases in winter to about 25% and decreases in summer to below 20% (Figure 5a), while the relative contribution of older water is about 5% higher in summer than in winter (Figure 5c). Discharge is high in winter and low in summer in these catchments and the contribution of the fractions therefore also varies with discharge (Figures 5d and 5f). The lowest 10% of discharge contains about 15% young and 18% old water, while the upper 10% contains 30% young and 10% old water (Figures 5d and 5f). The Roelinksbeek has a spiky discharge and the stream often dries out during dry summers. The fact that the average TTD shape in summer approximates the average shape in winter (Figures 3d–3f) is also reflected in the TTD fractions, which seem to follow the up and down going pattern of the discharge (Figures 4d and 5). The main difference between summer and winter discharge of the Roelinksbeek is that the variation in contributions is larger during low flows in summer (larger boxes) than during higher flows in winter (Figures 5a and 5d). For the upper 60% of discharge the fractions seem to have a similar trend as the Springendalse Beek with more young and less old water with higher flows (Figures 5d and 5f). The Elsbeek's spiky discharge consists mainly of water younger than 1 and older than 25 years: The medium aged water contributes a minor but stable fraction (Figure 5b), which follows from the specific shape of the TTD (Figure 3f). The discharge of young water (<1-year-old) is just above 20% in summer and increases to 40% in winter (Figure 5a) while the discharge of old water (>25 years old) fluctuates between 50% in summer and 35% in winter (Figure 5c).

In all catchments the storage of the older water is less variable than the younger water, as both the seasonal and longer term trends are more profound in the young water fractions (Figures 4a, 4c, and 4e). The storage of the Springendalse Beek that contributes to discharge of the stream was determined at 2.5 m, with slightly higher storage in winter than in summer (Figure 4a). The storage of the Roelinksbeek is approximately 2.3 m and looks similar to the RTD of the Springendalse Beek (Figure 4d). Although there is potentially tens of meters of saturated zone in the catchments (Figure 1), the particle tracking showed that the deeper flow routes discharge outside the catchment and do not contribute to storage. The Elsbeek catchment on the other hand has a calculated storage of 6.6 m on average, which is much higher than the other streams as a result of the contributing deeper aquifer (Figure 1) and consequently the storage consists for a large part of water older than 25 years (Figure 4e). It must be noted that calculation of the amount of water storage of catchments is generally difficult because of the uncertainty in the thickness of aquifers and the part of aquifers that contribute to discharge (Ali et al., 2014; Soulsby et al., 2009), and to this point, the results presented here are approximations based on the particle tracking calculations.

3.3. Dynamic SAS Functions

The calculated RTDs and TTDs were combined into dynamic SAS functions (equation (9)) and described using cumulative beta distributions as illustrated in section 2.5. The fitted a/b values of the cumulative beta

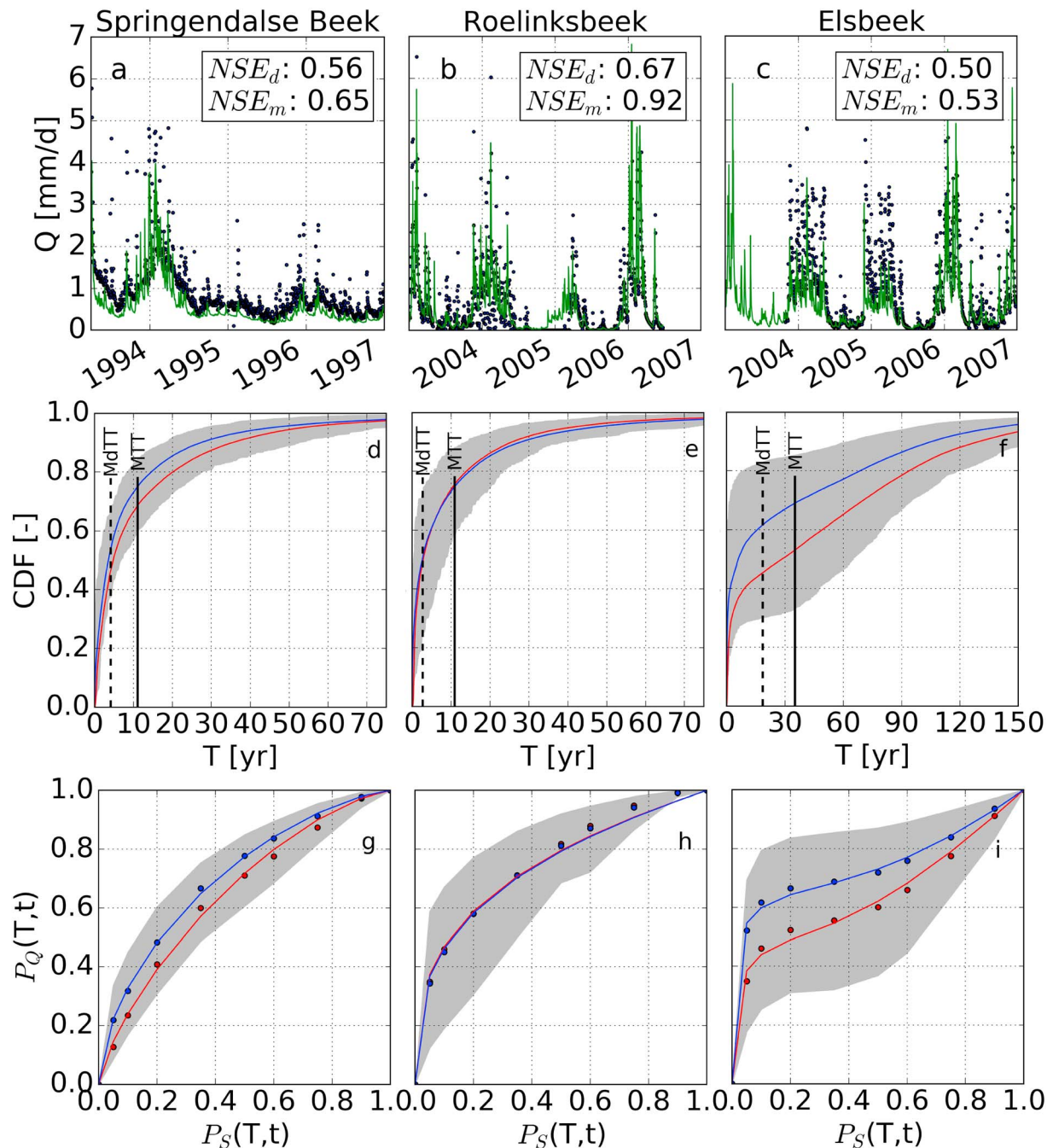


Figure 3. (a–c) Measured (dots) versus calculated (lines) daily discharges and Nash-Sutcliffe efficiency (NSE) coefficients calculated on daily (NSE_d) and monthly (NSE_m) mean data of shown model periods. (d–f) Time-weighted averaged CDF of travel time distributions (TTDs; similar to MHRFs in Heidbüchel et al., 2012) of the catchments in summer (red) and winter (blue). The gray area indicates the variance of the TTDs. The average median (MdTT) and mean (MTT) travel times are indicated with a dashed and solid line respectively. (g–i) Calculated (points) and fitted (line) average SAS functions for summer (red) and winter (blue) of the three catchments. The gray area indicates the minimal and maximal SAS functions.

distributions were averaged per month using the 20 year time frame they were calculated for (Figure 6). Only the shallow aquifer of the Elsbeek (fitted with the cumulative beta distribution with $b = 1$) was included here, as it is more comparable with the other catchments than the combination of the shallow and deep aquifer. On average, the a/b values of the Springendalse Beek appear to be higher than those of the Roelinksbeek and Elsbeek (Figure 6), indicating a lower preference for younger water and a more uniform-selection. The Elsbeek and Springendalse Beek show a clear seasonal pattern with lower preference for young water in

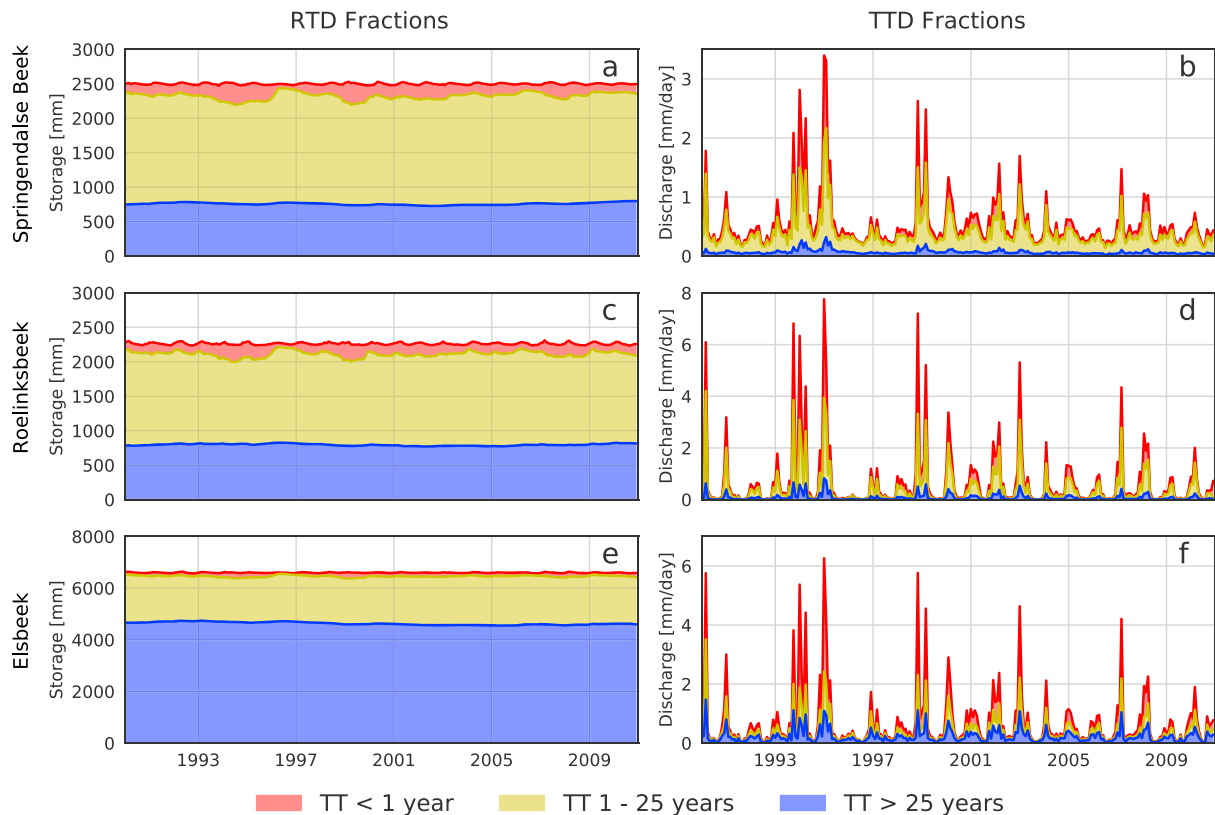


Figure 4. Three age classes of the residence time distributions and travel time distributions of the Springendalse Beek (a, b), Roelinksbeek (c, d), and Elsbeek (e, f) catchments.

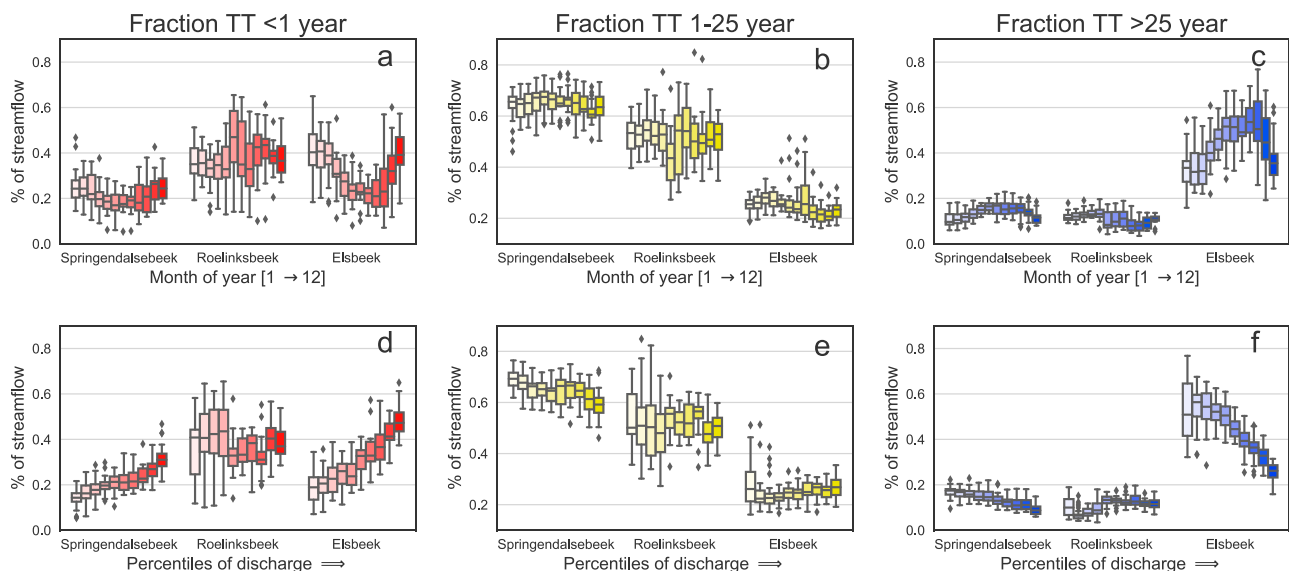


Figure 5. Fluctuations of discharge fractions provided by young ($TT < 1$ year), medium ($TT 1\text{--}25$ years), and old water ($TT > 25$ years) between months (a–c) and with discharge percentiles (d–f) in the catchments of the Elsbeek, Roelinksbeek, and Springendalse Beek. Boxplots are the result of the grouping and averaging of the modeling results of 1990–2010 by month and discharge class. TT = travel time.

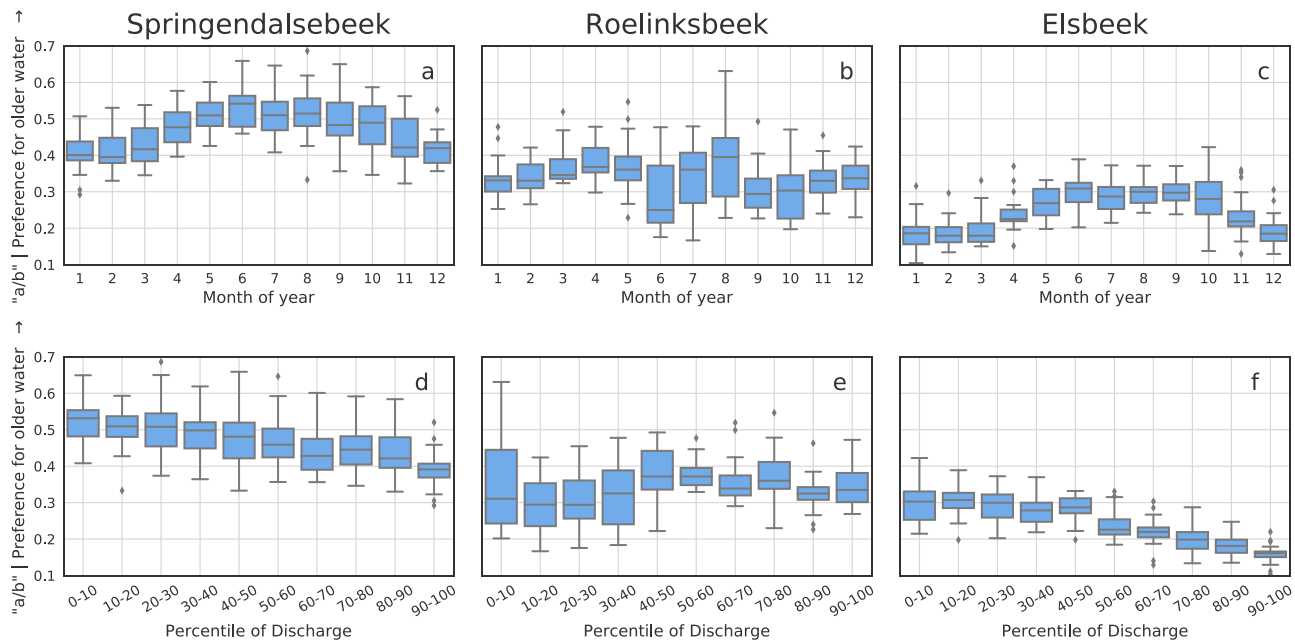


Figure 6. Variation in time (a–c) and with discharge (d–f) of the preference for the discharge of older water in the three catchments. Note that for the Elsbeek catchment (c, f), this only describes the shallower aquifers while the catchment also included a uniformly selected deep aquifer delivering old water. Boxplots are the result of the grouping and averaging of the modeling results of 1990–2010 by month and discharge class.

summer and higher preference in winter (Figures 6a and 6c); in other words, water in summer is more uniformly selected than in winter. As discharge is lower in summer and higher in winter, this means that the preference for younger water in the Springendalse Beek and Elsbeek catchments increases with discharge (Figures 6d and 6f). This pattern is not clear in the Roelinksbeek, which shows a large variation in young water preference during the summer months (Figures 6b and 6e). Despite the large variation, there appears to be a decrease in the preference for older water in spring, followed by an increase in summer when flows are low (Figure 6b). Late summer to autumn again shows a higher preference for younger water. During low discharge conditions (dry) in the Roelinksbeek the preference for younger water is highest (Figure 6e), while in higher discharge conditions (wet) the preference is lower but seems to slightly increase with discharge.

Because of the use of a spatially distributed model, it was possible to explore the differences within catchments. We constructed separate SAS functions for the upstream and downstream parts of the Springendalse Beek and Roelinksbeek and composed the monthly time-averaged a/b values, which are reported in supporting information S3. In the Springendalse Beek the upstream and downstream parts had the same seasonal pattern as the whole catchment, but the upstream part has a higher preference for older water while the downstream part has a lower preference. In the Roelinksbeek the upstream part seems to have the same seasonal patterns as the other catchments with a higher preference for older water in summer. The downstream part however seems to have no seasonal pattern.

The preference for young or old water expressed by a/b was plotted against the modeled monthly mean depth of the groundwater level and against the fraction of the model cells that drain water in the specific month (Figure 7). As the groundwater levels and drainage networks differ greatly in the upstream and downstream parts of the Springendalse Beek, they were plotted separately in Figure 7. While for the Elsbeek and downstream part of the Springendalse Beek there is an increase in the preference for older water with dropping groundwater levels, for the upstream part of the Springendalse beek this preferences seems to decrease (Figure 7a). Likewise the Elsbeek and downstream part of the Springendalse Beek show an increase in the preference for old water when there are fewer actively draining cells. The Roelinksbeek shows no clear relation with either the depth of the groundwater or the fraction of draining cells (Figure 7b).

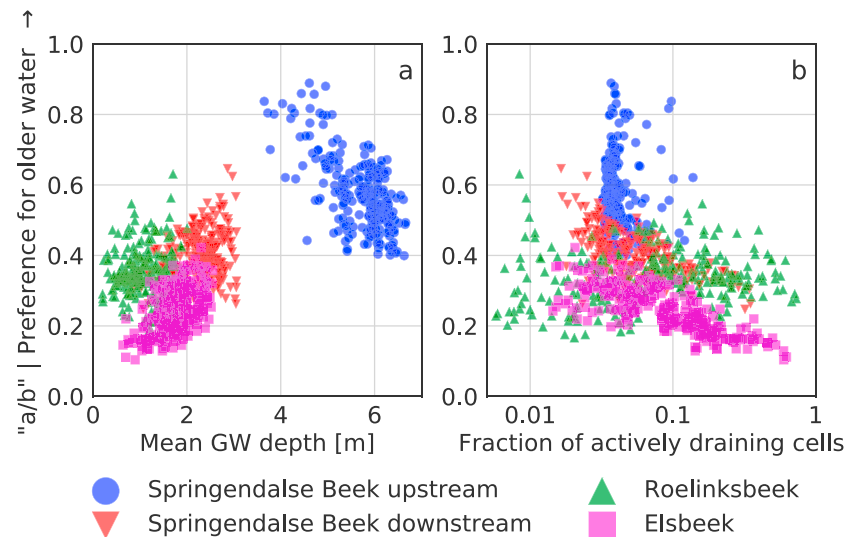


Figure 7. The StorAge Selection a/b plotted against the modeled monthly mean groundwater depths (a) and the fractions of the cells actively draining (b).

4. Discussion

4.1. Travel and RTDs

The catchment RTDs showed seasonal variation, particularly in the younger water fraction. Changes in storage only occur near the surface because changes in groundwater levels and the RTD of the storage are the result of (a) the addition of new water from groundwater recharge with a TT of 0, (b) the aging of water already in storage, and (c) the preferential discharge of water with a certain age from the storage.

The increased fraction of young water in the wet season found for both the Springendalse Beek and Elsbeek (Figure 5) is in agreement with earlier research (e.g., Morgenstern et al., 2010), where it was found that streamflow under baseflow conditions contains water with longer TTs than during high flows. This variation can be explained by changes in flow paths: high groundwater levels in winter lead to the discharge of more young water through shallow flow paths that are tapped by tile drains or shallow ditches. In addition, research has shown that fluxes of old groundwater are more stable throughout the year than those of young groundwater (e.g., Rinaldo et al., 2011; van der Velde et al., 2012; van der Velde et al., 2015). Our results from the Springendalse Beek and Elsbeek catchments show that the amount of younger water contributing to discharge indeed varies by a larger amount than the old water contribution (Figure 5). However, the Elsbeek has a more variable discharge than the Springendalse Beek, despite the fact that it contains a larger amount of old water. An explanation is that the groundwater levels in the Elsbeek are shallower and thus oscillate around the depth of the drainage network, leading to more variation in active stream network (Figures 7b and 8) and consequently into more variation in discharge.

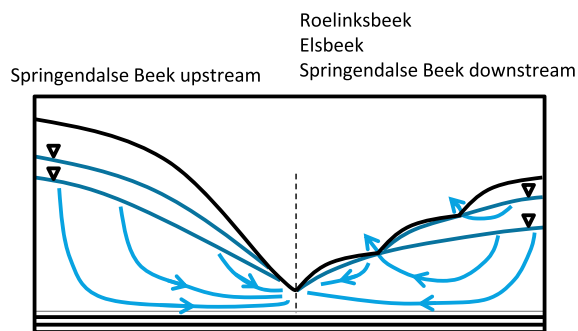


Figure 8. Conceptual drawings of the main mechanism acting in the study catchments. Rising groundwater levels control catchment travel times in two ways: They increase the speed of fluxes in the full range of flow paths (left), and they (re) activate shallow flow paths (right).

4.2. Catchment SAS Functions

The dynamical SAS functions of our study catchments always show a/b values below 1. This means that these catchments have a preference for the discharge of young water (Figure 6), just like previous studies have shown for relatively flat catchments with thick aquifers (van der Velde et al., 2012; Benettin et al., 2017). The stronger preference for the discharge of young water in wet periods with high storage that was found for the Elsbeek and Springendalse Beek (Figures 6d and 6f) is called the “inverse storage effect,” and was also reported by Harman (2015); Benettin, Soulsby, et al. (2017); and Pangle et al. (2017). On the contrary we did not find this effect for the

Roelinksbeek, where the variability of the SAS functions showed no clear correlation with seasons, storage or discharge (Figure 6). This behavior will be further discussed in section 4.3. While the Springendalse Beek and Roelinksbeek were described directly by one SAS function, we needed a combination of two SAS functions to describe the Elsbeek, in a similar way as the multi-RS approach reported by Benettin, Soulsby, et al. (2017).

4.3. Catchment Processes and Functioning

Conceptually, the difference in functioning between the catchments can be explained by considering two interacting mechanisms that activate flow paths as a result of rising groundwater levels and increasing catchment storage (Figure 8). In the first mechanism, rising groundwater levels activate shallow flow paths which only function seasonally (Hrachowitz et al., 2013; Rodhe et al., 1996; Rozemeijer & Broers, 2007; van der Velde et al., 2012; van der Velde et al., 2015). This way, the extent of the stream network increases with rising groundwater tables, as more shallow streams, ditches and drains start discharging water (de Vries, 1995; Rozemeijer & Broers, 2007). Groundwater age tends to increase with depth (Vogel, 1967; Broers & van Geer, 2005) and therefore the shallow flow paths that are activated lead to an increased young fraction as well as a change in the shape of the SAS function. In the second mechanism, rising groundwater levels intensify the fluxes through the full range of flow paths. This process pushes water down into the aquifer in infiltration areas and up to the surface from storage in exfiltration areas (Botter et al., 2010; Cartwright & Morgenstern, 2015; Duvert et al., 2016). As such, fluxes through all flow paths are intensified more or less equally and therefore the contributions of different flow paths to discharge are not changed and SAS functions remain the same. These two mechanisms can be classified as a kind of internal and external variability, as defined in Harman et al. (2016) and Kim et al. (2016). Internal variability includes changes of internal flow paths which affect SAS functions (e.g., activation of shallow flow paths) and external variability includes the effect of input/climate variability which does not change SAS functions (e.g., flux intensification).

In the Springendalse Beek and Elsbeek catchments both mechanisms are clearly present: due to the activation of shallow flow paths the SAS functions vary with discharge and show the “inverse storage effect” (Figure 6), while due to flux intensification in the wet season both the amount of young and old water vary (Figure 4) but SAS functions are not affected. Another interesting finding is that the Roelinksbeek does not seem to show the “inverse storage effect” on the time scale we are considering. The fact that there is no systematic variation in the different TT fractions with discharge in the Roelinksbeek catchment (Figures 4 and 5) suggests that variation in groundwater levels here does not activate new shallow flow paths. However, the extent of active drainage varies especially strong in this catchment, which suggests a dynamic drainage system (Figure 8). Comparing the up- and downstream part of the Roelinksbeek revealed that the upstream part, which includes an aquifer on top of the moraine, functions similar to the other catchments and does show the “inverse storage effect” with a higher preference for younger water in the wet season (see supporting information S3). The unclear seasonal pattern shown in Figure 6b is thus the result of the downstream part of the Roelinksbeek catchment, where the preference for young water shows no seasonal pattern. An explanation can be found in the fact that the summer discharge in the Roelinksbeek catchment is very low and that streamflow regularly ceases during dry periods. During the transition from wet to dry, groundwater levels drop and the drainage network shrinks. However, part of the Roelinksbeek catchment only has a thin aquifer layer (Figure 1) where groundwater levels drop less deep and the formation of old water is not possible. At these locations, relatively young water will remain being discharged during the first part of the dry period and as a result the preference for younger water in the catchment actually increases. When groundwater levels drop further this preference for younger water ceases and only deep flow paths remain. As the catchment wets up following the dry period the reverse happens and a higher preference for younger water precedes the higher preference for older water in the wet period. These analyses show that both flow path activation and flux intensification may occur in a catchment even though stable SAS functions may suggest only the occurrence of flux intensification.

4.4. Landscape Characteristics Controlling Catchment Functioning

Our study did not include enough catchments to systematically analyze relationships between landscape properties and SAS function, but we can link the shape of the obtained SAS functions and its variability to several dominant catchment characteristics.

For the activation of young flow paths to occur in a catchment, there needs to be a drainage network that falls dry during dry periods and is reactivated during wet periods. This way, stream geometry as well as artificial drainage control the shape of catchment TTDs and SAS functions. By activating shallow flow paths, artificial drainage shifts the complete TTD towards younger ages, decreasing the fluxes of old water and thus increasing the age of this old fraction (e.g., Danesh-Yazdi et al., 2016; Rozemeijer et al., 2016). As all three catchments showed at least some variation in the SAS functions (Figures 3g–3i), flow path activation must be taking place.

The upstream part of the Springendalse Beek catchment has a higher preference for the discharge of older water than the downstream part (see supporting information S3). These subcatchments have contrasting drainage network and aquifer thickness. While the upstream part has a relatively stable discharge due to the many springs draining the thick aquifer, the downstream part has a more spiky discharge with many agricultural drains and ditches, a shallower groundwater level and fewer slopes, leading a higher preference for the discharge of younger water. Despite this, the seasonal pattern in a/b (inverse storage effect) is similar. It is therefore surprising that the preference for younger water seems to decrease with higher groundwater levels (Figure 7a). We argue that this correlation is a spatial artifact, caused by the fact that at the start of the drying period groundwater levels on the stream valley flank remain high, while the first parts of the drainage network are already deactivated. The Elsbeek catchment also shows an increase in the preference for old water in the dry season when there are deeper groundwater levels and fewer cells draining water. This supports the idea that these catchments function in similar ways (Figure 8) and are highly influenced by the drainage network. The Roelinksbeek, however, shows no clear relation with either the depth of the groundwater or the fraction of draining cells (Figure 7b), which results from the mixing of spatially contrasting storage-discharge behavior (as described earlier). This is similar to the spatial effect of groundwater levels on a/b reported for the upstream part of the Springendalse Beek.

The SAS functions of the Elsbeek, with a geology consisting of two aquifers, had a more complex shape which could not be described by a single cumulative beta distribution (Figure 3i). Instead, a mix of two cumulative beta distributions was used, representing a shallow reservoir with a preference for young water and a deep uniformly selected reservoir. This was also described by Gusyev et al. (2013) for one of their study catchments, where they showed how, depending on the hydrogeological conditions in the aquifers, a river contained a mixture of young water from a shallow layer and old water from a deep layer. Similar observations were made for groundwater wells, for instance by Eberts et al. (2012) who showed the importance of major hydrogeological features for the TTD of wells. This result shows that the specific subsurface configuration exerts a strong control over the shape of the SAS functions that cannot always be captured by a simple cumulative beta distribution. This leads us to conclude that while drainage processes seem to control the preference for young water as well as the variation in storage selection in time (Figure 7), geological constraints control the overall shape of the SAS function.

In our study, evapotranspiration from groundwater was taken from the youngest water, and because of that, the average age of the groundwater increases when evapotranspiration increases. On the other hand, evapotranspiration can be considered to counteract piston-flow mobilization of flow paths, as it reduces the pressure on the system and consequently the flows of older water. This is similar to Cartwright and Morgenstern (2015), who argued that higher evapotranspiration decreases the volume of discharge of older water. It can therefore be argued that evapotranspiration can both increase and decrease catchment TTDs and the SAS a/b .

4.5. Time Variability of SAS Functions in Lumped Catchment Reservoir Approaches

SAS functions are applied in literature to describe the coupling between S and Q and to create TTDs for catchments for which not a detailed spatial distributed model is available. For this, a SAS-functional form is chosen or fitted, which is then combined with water balances to construct the TTDs (Figure 2). Our study catchments show a preference for the discharge of young water and the “inverse storage effect” where the preference for younger water increases during the wet season. This seasonality was already used by both van der Velde et al. (2015) and Harman (2015), who specified storage-dependent selection (SAS) functions. Recently Benettin, Soulsby, et al. (2017) showed TTDs that were also time variable with catchment wetness (storage) and Danesh-Yazdi et al. (2016) included time-variable selection by choosing SAS functions based on seasons. Extending on these studies, our research shows variation in catchment SAS functions that were actually calculated from a transient groundwater model. The SAS functions of the Springendalse Beek and Elsbeek

showed clear variation with wetness, supporting the practice of varying SAS functions with storage. However, the SAS function of the Roelinksbeek catchment did not have a clear correlation with storage, discharge or seasons.

4.6. Limitations and Implications

This paper studied TTs of only the groundwater as it delivered most of the discharge in our study catchments due to the thin unsaturated zones and thick aquifers. For the full TTD of catchments with thick unsaturated zones and shallow aquifers it is of importance to take TTs through the unsaturated zones into account. Both the effect of unsaturated zone and preferential flow paths on catchment TTs are captured in tracer studies and it should be an aim to include these more in catchment modeling studies. In addition, for simplicity we used step functions to represent evapotranspiration (equation (10)). Although evapotranspiration may have been oversimplified in our study, we also did not take TTs and mixing in the unsaturated zone into account which is where most of the evapotranspiration processes take place. In this study we computed monthly varying TTDs and SAS functions, while shorter term variability is also important especially when TTDs are used to compare with grab-samples of water chemistry (e.g., Benettin, Soulsby, et al., 2017). This short time variability, however, was not the focus of our study as we aimed to understand the effects of geology and catchment structure on TTDs. Future research should focus on the effects of preferential flow paths and the resulting short time variability in catchment mixing/selection. In addition, recent studies focused on SAS functions of ET (e.g., Quéloz et al., 2015), which can be combined with groundwater SAS functions.

The TTDs in this paper are the result of forward modeling and therefore provide conceptual understanding of the different catchment systems, given the underlying model. The advantages of calculating TTDs using a distributed groundwater model as opposed to lumped catchment reservoir models are that effects of heterogeneity are better captured and the possibility to model TTDs in a forward way instead of fitting them on measured concentration or discharge time series. This opens the opportunity to calculate the effect of measures and scenarios on catchment TTDs (e.g., Wilusz et al., 2017). A time series of ^3H measurements or other young groundwater tracers such as CFC's or SF₆—preferably spanning a period of 5–10 years—would be beneficial for validation purposes (e.g., Kolbe et al., 2016; Modica et al., 1998), and if sampled with high enough frequency, could also reveal and confirm the dynamics of the TTDs (as found by e.g., Morgenstern et al., 2010). Unfortunately these data were not available in our study area. Inversely, a more efficient sampling design and tracer interpretation could be achieved predicting the age tracer signals beforehand using our TTD model approach, which was outside the scope of this paper.

Because flow paths have different chemical characteristics, the TTs of groundwater can be used to predict or interpret the chemistry of the surface water that is fed by the groundwater. More in depth knowledge of the behavior of time-variable SAS functions for different catchments helps research in which TTs are calculated from an assumed SAS function (see section 4.5). The use of spatially distributed models opens more opportunities to unravel the landscape characteristics controlling time and spatially varying TTDs and SAS functions, but we must not forget the limitations of such models regarding preferential flow paths and subsurface heterogeneity.

5. Conclusions

To explore the variation in the groundwater contribution to streams, an approach was presented to calculate dynamic monthly RTDs, TTDs, and SAS functions which builds upon recent advances in TT research. With this method, the groundwater contribution to three Dutch streams was characterized. The TTD of the groundwater input to streams is highly variable as a result of the activation of different groundwater flow paths. Old flow paths are more stable than younger flow paths in time, but still vary due to the intensification of fluxes in wet periods.

The dynamical SAS functions showed that our study catchments have a preference for the discharge of young water throughout the year, just like previous studies have shown for relatively flat catchments with thick aquifers (Benettin, Soulsby, et al., 2017; Van der Velde et al., 2012). In addition, for two of the study catchments the storage selection shifts towards a preference for older water in summer, as young flow paths cease. On the other hand, this “inverse storage effect” was not found for the Roelinksbeek catchment which is probably caused by spatial mixing within the catchment, which complicates interpretation of catchment SAS

functions. It was shown that the dominant process that governs the shape of the SAS function relates to sub-surface structure, while the overall preference for young water seems strongly controlled by drainage density and the variability in storage selection is controlled by the extent to which shallow-flow paths are activated when a catchment wets up.

Time-varying SAS functions are required for realistic computation of dynamic TTDs with lumped catchment-scale reservoir approaches and this research showed realistic ranges for the variability in time and space in the SAS functions of lowland catchments. For some catchments SAS functions can be varied with storage or discharge, whereas in another catchment SAS functions did not clearly show this relation. In addition, catchments composed of multiple aquifer layers may require the mixing of multiple SAS functions. Further research is needed to gain more insight into the relation between the TTD and the chemistry of both ground-water and surface water and to find the landscape characteristics controlling catchment functioning.

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