



Deforestation reduces the vegetation-accessible water storage in the unsaturated soil and affects catchment travel time distributions and young water fractions

Markus Hrachowitz¹, Michael Stockinger^{2,3}, Miriam Coenders-Gerrits¹, Ruud van der Ent¹, Heye Bogena², Andreas Lücke², Christine Stumpp³

¹Department of Watermanagement, Faculty of Civil Engineering and Geosciences, Delft University of Technology, Stevinweg 1, 2628CN Delft, Netherlands

²Institute of Bio- and Geosciences, Agrosphere Institute (IBG-3), Forschungszentrum Jülich, Wilhelm-Johnen-Straße, 52425 Jülich, Germany

³Institute for Soil Physics and Rural Water Management, University of Natural Resources and Life Sciences Vienna, Muthgasse 18, 1190 Vienna, Austria

Correspondence to: Markus Hrachowitz (m.hrachowitz@tudelft.nl)

Abstract. Deforestation can considerably affect transpiration dynamics and magnitudes at the catchment-scale and thereby alter the partitioning between drainage and evaporative water fluxes released from terrestrial hydrological systems. However, it has so far remained problematic to directly link reductions in transpiration to changes in the physical properties of the system and to quantify these changes of system properties at the catchment-scale. As a consequence, it is difficult to quantify the effect of deforestation on parameters of catchment-scale hydrological models. This in turn leads to substantial uncertainties in predictions of the hydrological response after deforestation but also to a poor understanding of how deforestation affects principal descriptors of catchment-scale transport, such as travel time distributions and young water fractions. The objectives of this study are therefore to quantify the effects of deforestation in the Wüstebach experimental catchment on the partitioning of water fluxes and to directly associate these changes to changes in parameters of a hydrological model with integrated tracer routine based on the concept of storage age selection functions. Simultaneously modelling stream flow and stable water isotope dynamics using meaningfully adjusted model parameters both for the pre- and post-deforestation periods, respectively, the model is used to track fluxes through the system and to estimate the effects of deforestation on catchment travel time distributions and young water fractions F_{yw} .

It was found that deforestation led to a significant increase of stream flow, accompanied by corresponding reductions of evaporative fluxes. This is reflected by an increase of the runoff ratio from $C_R = 0.55$ to 0.68 in the post-deforestation period despite similar climatic conditions. This reduction of evaporative fluxes could be linked to a reduction of the catchment-scale water storage volume in the unsaturated soil ($S_{U,max}$) that is within the reach of active roots and thus accessible for vegetation transpiration from ~225 mm in the pre-deforestation period to ~90 mm in the post-deforestations period. The hydrological model, reflecting the changes in the parameter $S_{U,max}$ indicated that in the post-deforestation period stream water was characterized by slightly higher mean fractions of young water ($F_{yw} \sim 0.13$) than in the pre-deforestation period ($F_{yw} \sim 0.11$).



In spite of these limited effects on the overall F_{yw} , considerable changes were found for wet periods, during which post-
35 deforestation fractions of young water increased to values $F_{yw} \sim 0.40$ for individual storms. Deforestation also caused a
significantly increased sensitivity of young water fractions to discharge under wet conditions from $dF_{yw}/dQ = 0.25$ to 0.43.
Overall, this study demonstrates that deforestation has not only the potential to affect the partitioning between drainage and
evaporation as well as the vegetation-accessible storage volumes $S_{U,max}$, and thus the fundamental hydrological response
characteristics of catchments, but also catchment-scale tracer circulation dynamics. In particular for wet conditions,
40 deforestation caused higher proportions of younger water to reach the stream, implying faster routing of stable isotopes and
plausibly also solutes through the subsurface.

1 Introduction

Plant transpiration is, globally, the largest continental water flux (Jasechko, 2018). Notwithstanding considerable
uncertainties (Coenders-Gerrits, 2014), its magnitude depends on the interplay between canopy water demand and
45 subsurface water supply (Eagleson, 1982; Milly and Dunne, 1994; Donohue et al., 2007; Yang et al., 2016; Jaramillo et al.,
2018). The latter is regulated by water volumes that are within the reach of roots and can be taken up by plants. Many plant
species across humid climate zones develop only rather shallow root systems (Schenk, 2005) that do not directly tap the
groundwater (Fan et al., 2017). In such terrestrial hydrological systems that are dominated by shallow-rooting vegetation, the
water-filled pore volume between field capacity and permanent wilting point that is *within the reach of active roots* becomes
50 a core property of many terrestrial hydrological systems (Rodriguez-Iturbe et al., 2007). This maximum vegetation-
accessible water storage volume in the unsaturated root-zone of soils, hereafter referred to as vegetation-accessible water
storage capacity $S_{U,max}$ [mm], constitutes a major partitioning point of water fluxes as it regulates the temporally varying ratio
between drainage, such as groundwater recharge or shallow lateral flow, on the one hand and transpiration fluxes on the
other hand (Savenije and Hrachowitz, 2017), which can in turn generate considerable feedback effects on downwind
55 precipitation and drought generation (e.g. Seneviratne et al., 2013; Ellison et al., 2017; Teuling, 2018; Wang-Erlandsson et
al., 2018; Wehrli et al., 2019).

Traditionally, $S_{U,max}$ is determined as the product of root-depths or root-distributions and pore water content between field
capacity and permanent wilting point. Although correct in principle, this method has several weaknesses for applications at
the catchment-scale as much of the required data are typically not available at sufficient levels of detail. While soil maps and
60 the associated soil water retention curves have become globally available at resolutions $< 1\text{km}$ (Arrouays et al., 2017; Hengl
et al., 2017), they are characterized by considerable uncertainties. Similarly, direct and detailed observations of root-systems
are very scarce. They are, globally, limited to a few thousand individual plants only (e.g. Schenk and Jackson, 2002; Fan et
al., 2017) and many of the observations are based on biomass extrapolations after excavating only the first meter of soil or
less (Schenk and Jackson, 2003). Consequently, soil and root data largely remain inaccurate snapshots in space. As such,
65 they are likely to be inadequate reflections of the spatial heterogeneity of soils and roots. In addition, these available data are



also mostly snapshots in time and therefore disregard the adaptive behaviour of plant communities, whose compositions, and thus characteristics, at ecosystem level continuously evolve over multiple scales in space and time in response to changes in ambient conditions (e.g. Laio et al., 2006; Brunner et al., 2015; Tron et al., 2015).

There is increasing evidence that vegetation does not only actively adapt to its (changing) environment, but that it does so in an efficient way (e.g. Guswa, 2008; Schymanski et al., 2008). The vegetation, i.e. a collective of individual different plants within an area of interest, present at any given moment at any given location has survived. This in itself is a manifestation of the successful adaption of individual plants to their environment in the past. They have optimally allocated resources to balance sub- and above-surface growth to simultaneously meet water, nutrient and light requirements. This implies that these plants developed root-systems that, amongst other factors, ensure continuous access to *sufficient* water – but not more – to bridge dry periods. An individual plant that is not adapted to meet its water and nutrient requirements through its root-system as well as its light requirements through its foliage system in competition with other plants will disappear and be replaced by a better adapted plant. The root-system of vegetation at ecosystem level, and the associated vegetation-accessible water storage capacity $S_{U,max}$, is therefore at a dynamic equilibrium with and responding to the ever changing conditions of its environment. Similarly, any type of direct human interference with vegetation, such as deforestation, has an impact on transpiration water demand, the extent and structure of active root-systems and consequently on $S_{U,max}$ (Nijzink et al., 2016a). For a meaningful quantification of $S_{U,max}$ at larger scales, such as the catchment-scale, it is therefore necessary to adopt a Darwinian perspective (Harman and Troch, 2014) and to estimate effective values of $S_{U,max}$, reflecting the collective and adaptive behaviour of all individual plants within a catchment. Results from many previous studies suggest, broadly speaking, three methods to do so. The first is the use of inverse approaches that treat $S_{U,max}$ as model calibration parameter (Fenicia et al., 2008; Speich et al., 2018; Bouaziz et al., 2020; Knighton et al., 2020). Alternatively, the second type of methods is based on optimality principles that maximize variables such as net primary production or carbon gain (Kleidon, 2004; Guswa, 2008; Hwang et al., 2009; Yang, et al., 2016; Speich et al., 2018), nitrogen uptake (McMurtrie et al., 2012) or transpiration rates (Collins and Bras, 2007; Sivandran and Bras, 2012). Lastly, $S_{U,max}$ and its evolution over time can be directly estimated through magnitudes of annual water deficits as determined from observed water balance data (Gentine et al., 2012; Donohue et al., 2012; Gao et al., 2014; DeBoer-Euser et al., 2016).

For transpiration, shallow-rooting plants extract pore water of unsaturated soils that is held against gravity, i.e. between field capacity and permanent wilting point, and within the reach of roots. Significant vertical or lateral drainage only occurs at water contents above field capacity. Transpiration extracting soil water below that therefore effectively generates a root-zone water storage reservoir between field capacity and permanent wilting point that is characterized by a storage capacity $S_{U,max}$, i.e. a maximum vegetation-accessible storage volume, and that is at any given moment filled with a specific water volume $S_U(t)$, depending on the past sequence of water inflow and release.

Storage volumes as $S_{U,max}$, or others such as groundwater bodies, are key for hydrological functioning (Sprenger et al., 2019b) as they provide a buffer against hydrological extremes, such as floods and droughts. With increasing storage, the hydrological memory of a system increases as more water can be stored and held over longer periods of time (e.g.



100 Hrachowitz et al., 2015; Sprenger et al., 2019b). This implies that storage and changes thereof also control catchment residence time distributions (RTD) and travel time distributions (TTD; Soulsby et al., 2010). As fundamental descriptors of hydrological functioning RTDs and TTDs describe the age structure of water held in and released from catchments (Birkel et al., 2015; Rinaldo et al., 2015), which is critical for regulating solute transport and thus nutrient and contaminant dynamics (Hrachowitz et al., 2016).

105 However, neither the effects of land cover change (Blöschl et al., 2019) nor the individual roles of different storage compartments in terrestrial hydrological systems are well understood (McDonnell et al., 2010; Penna et al., 2018, 2020). This is mostly a consequence of the lack of suitable observational technology to directly observe their respective volumes at larger scales. It remains therefore also unclear how deforestation affects $S_{U,max}$ (e.g. due to a less developed and complex rooting system for subsequent younger vegetation) and how changes in $S_{U,max}$ may propagate to affect both, the partitioning
110 of water fluxes as well as the age structure of water stored in and released from catchments as described by residence and travel time distributions.

The objective of this study in the Wüstebach experimental catchment (Germany) is thus to analyse to which extent post-deforestation changes in the water balance can be attributed to changes in catchment-scale effective $S_{U,max}$ and to quantify the associated consequences on RTDs, TTDs and young water fractions F_{yw} . Specifically we test the hypotheses that (1)
115 deforestation affects water storage dynamics and the partitioning of water fluxes into transpiration and drainage, (2) the changes in water storage dynamics and partitioning of water fluxes are a direct consequence of a reduction of the vegetation-accessible water storage capacity in the unsaturated root-zone ($S_{U,max}$) after deforestation and that (3) the deforestation-induced reduction of $S_{U,max}$ affects the shape of travel time distributions and results in shifts towards higher fractions of young water in the stream.

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2 Study site

The experimental Wüstebach headwater catchment (0.39 km²; Fig. 1a) is part of the Lower Rhine/Eifel Observatory of the Terrestrial Environmental Observatories network (TERENO; Bogaen et al., 2018) located in the Eifel National Park in Germany (50°30'16"N, 06°20'00"E). The catchment is characterized by a humid, temperate climate with warm summers,
125 mild winters and a mean annual temperature of around 7°C (Zacharias et al., 2011). Mean annual precipitation is about 1200 mm yr⁻¹ and mean annual runoff about 700 mm yr⁻¹ (Fig. 2). Although most of the precipitation occurs in the winter months, the fraction that falls as snow is typically less than 10 % of the annual precipitation and snow cover is present for no more than 3-4 weeks per year.

The catchment is drained by a perennial 2nd-order stream and extends from 595 to 630 m asl. The landscape is characterized
130 by the gentle slopes of the surrounding hills and a flatter riparian area close to the stream, covering approximately 10 % of the catchment (Fig. 1a). The underlying bedrock is largely Devonian shales with sandstone inclusions (Richter, 2008)



covered by periglacial layers (Borchardt, 2012). While cambisols dominate the hillslopes, gleysols and histosols characterize much of the riparian area (Bogena et al., 2015). The average soil depth in the catchment reaches about 1.6 m with a maximum of 2 m (Graf et al., 2014). After the Second World War, the catchment was homogeneously and completely afforested (Fig. 1) with Sitka spruce (*Picea sitchensis*) and Norway spruce (*Picea abies*; Etmann, 2009). The maximum observed rooting depth of these spruce trees in the catchment is 50 cm and no roots were observed below this depth. In the course of the development of the area into a national park approximately 21 % of the catchment, including the entire riparian zone, were deforested in September 2013 and kept largely vegetation free since (Wiekenkamp et al., 2016; Fig. 1).

140 3 Data

3.1 Hydro-meteorological data

Daily hydro-meteorological data were available for the period 01/10/2009 – 30/09/2016 (Fig. 2a-c). Precipitation P [mm d⁻¹] and mean daily temperature T [°C] were available from the Monschau-Kalterherberg meteorological station operated by the German Weather Service (Deutscher Wetterdienst DWD station 3339), located 9 km northwest of the Wüstebach catchment. Stream discharge Q [mm d⁻¹] at the outlet of the Wüstebach was observed with a V-notch weir for low flow measurements and a Parshall flume for medium to high flows (Bogena et al., 2015). Daily potential evaporation E_P [mm d⁻¹] was estimated using the Penman-Monteith equation.

3.2 Stable isotope data

Regular weekly $\delta^{18}\text{O}$ data from bulk precipitation samples collected in a cooled wet deposition gauge at the meteorological station Schleiden-Schöneseiffen (Meteomedia station) 3 km northeast of the catchment, were available for the period 01/10/2010 – 24/09/2012. After that, precipitation was sampled at half-daily intervals until 30/09/2016 using an automatic, cooled sampler (Eigenbrodt GmbH, Germany). The half-daily samples were precipitation volume-weighted to daily sampling intervals (Stockinger et al., 2016, 2017). Weekly stream water grab samples for stable water isotope analysis were taken at the outlet of the Wüstebach catchment in the 01/10/2010 – 30/09/2016 period (Fig. 2d). Isotope analysis was carried out using laser-based cavity ringdown spectrometers (L2120-i/L2130-i, Picarro Inc.). Internal standards calibrated against VSMOW, Greenland Ice Sheet Precipitation (GISP) and Standard Light Antarctic Precipitation (SLAP2) were used for calibration and to ensure long-term stability of analyses (Brand et al., 2014). The long-term precision of the analytical system was $\leq 0.1 \text{ ‰}$ for $\delta^{18}\text{O}$.

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4 Methods



To quantify effects of deforestation on $S_{U,max}$ and, as a consequence of that, on the age structure of water as described by TTDs, RTDs and young water fractions F_{yw} , the following stepwise experiment was designed: (1) quantify changes in the partitioning of annual water fluxes between the pre- and the post-deforestation periods based on observed water balance data; (2) estimate the effect of these changes on the magnitudes of pre- and post-deforestation $S_{U,max}$, respectively, using the same data; (3) calibrate a hydrological model to simultaneously reproduce stream flow and stream $\delta^{18}\text{O}$ dynamics for the pre-deforestation period; (4) use the calibrated parameter sets to run the model in the post-deforestation period and evaluate the model's post-deforestation performance without further calibration; (5) re-calibrate the model for the post-deforestation period and evaluate if changes in calibrated $S_{U,max}$ (and other parameters) are plausible and reflect changes in $S_{U,max}$ directly estimated from water balance data in step (2); and finally (6) use the calibrated pre- and post-deforestation parameter sets, respectively, to track modelled water fluxes through the system and quantify changes in TTDs, RTDs and F_{yw} between the pre- and the post-deforestation periods.

4.1 Water balance-based estimation of $S_{U,max}$

To survive, plants need continuous access to water to satisfy canopy water demand. The root-systems of vegetation are therefore adapted to provide access to water volumes that correspond to annual water deficits that result from the combination of (1) the phase lag between and (2) the difference in the respective magnitudes of seasonal precipitation and solar radiation signals (Donohue et al., 2012; Gentine et al., 2012; Gao et al., 2014). On a daily basis, these water deficits $S_{D,j}(t)$ can be estimated as the cumulative sum of daily effective precipitation P_E [mm d⁻¹] minus transpiration E_T [mm d⁻¹]. The maximum deficit $S_{D,j}$ for a specific year j is then equivalent to the water volume that was accessible to vegetation through its root system over that period (deBoer-Euser et al., 2016; Nijzink et al., 2016a):

$$S_{D,j}(t) = \begin{cases} \int_{t_0}^t (P_E(t) - E_T(t)) dt, & \text{if } S_{D,j}(t) \leq 0 \\ 0, & \text{if } S_{D,j}(t) > 0 \end{cases} \quad (\text{Eq. 1})$$

$$S_{D,j} = \max(|S_{D,j}(t)|) \quad (\text{Eq. 2})$$

where t is the time step [d], and t_0 is the last preceding time step for which the storage deficit $S_{D,j}(t) = 0$. As an approximation, Equation 1 implies that if $S_{D,j}(t) = 0$, the water content in the root-accessible pore space at day t is at field capacity and cannot hold additional water. If water supply then exceeds canopy water demand on that day, i.e. $P_E(t) - E_T(t) > 0$, this water surplus is drained from the root zone, e.g. to recharge groundwater or directly to the stream, and cannot be used for transpiration.

Daily effective precipitation P_E , i.e. precipitation that actually reaches the soil, was estimated on basis of the water balance of a canopy interception storage (Nijzink et al., 2016a):

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$$\frac{dS_I(t)}{dt} = P(t) - E_I(t) - P_E(t) \quad (\text{Eq. 3})$$

Where E_I [mm d^{-1}] is daily interception evaporation and S_I [mm] the canopy interception storage. For each time step, E_I can then be computed as:

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$$E_I(t) = \begin{cases} E_P(t), & \text{if } E_P(t)dt < S_I(t) \\ \frac{S_I(t)}{dt}, & \text{if } E_P(t)dt \geq S_I(t) \end{cases} \quad (\text{Eq. 4})$$

This then further allows to estimate P_E according to:

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$$P_E(t) = \begin{cases} 0, & \text{if } S_I(t) < I_{max} \\ \frac{S_I(t) - I_{max}}{dt}, & \text{if } S_I(t) \geq I_{max} \end{cases} \quad (\text{Eq. 5})$$

where I_{max} [mm] is the canopy interception capacity. In the absence of more detailed information P_E was estimated with a range of different interception capacities, i.e. $I_{max} = 0, 1, 2, 3,$ and 4 mm, in a sensitivity analysis approach.

Note that the catchment average P_E after deforestation was estimated as the areal weighted mean of P_E in the deforested area (21% of catchment area) computed with an assumed $I_{max} = 0$ mm and P_E from the remaining area computed based on the above range of I_{max} between 0 and 4 mm. In a next step, assuming negligible groundwater imports or exports (cf. Bouaziz et al., 2018), data errors and storage changes, long-term mean transpiration $\overline{E_T}$ was estimated according to the water balance:

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$$\overline{E_T} = \overline{P_E} - \overline{Q} \quad (\text{Eq. 6})$$

Where $\overline{P_E}$ [mm d^{-1}] is the long-term mean effective precipitation and \overline{Q} [mm d^{-1}] is the long-term mean observed stream discharge. Daily transpiration E_T [mm d^{-1}] for use in Eq. (1) is then estimated by scaling the long-term mean transpiration to the signal of daily potential evaporation to approximate the seasonal fluctuation of energy input (Bouaziz et al., 2020):

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$$E_T(t) = (E_P(t) - E_I(t)) \frac{\overline{E_T}}{\overline{E_P} - \overline{E_I}} \quad (\text{Eq. 7})$$



A range of previous studies provided evidence that mature forests develop root-systems that allow access to sufficiently large pore water storage volumes $S_{U,max}$ to bridge droughts with return periods $T_R \sim 20$ years (Gao et al., 2014; deBoer-Euser et al., 2016; Nijzink et al., 2016a; Wang-Erlandsson et al., 2016). The maximum annual water deficits $S_{D,j}$ (Eq. 2) for all j years in the pre-deforestation study period were therefore used to fit a Gumbel extreme value distribution (Gumbel, 1941). This subsequently allowed the estimation of a water deficit with a 20-year return period, which is for this study defined as vegetation-accessible water storage $S_{U,max}$ so that $S_{U,max} = S_{D,20yr}$.

Note that due to the limited length of the data series the $S_{U,max}$ estimates are rather uncertain and need to be understood as merely indicative approximations. This is in particular true for the post-deforestation period, where attempts to explicitly link $S_{U,max}$ to a specific return period are subject to additional uncertainty: as the catchment was not reforested and natural recovery of vegetation is negligible (see aerial images in Figure 1), it is not implausible to assume that the development of the root-system after the disturbance is far from equilibrium and likely to be actively evolving over time. Also note that although E_T is, for brevity, referred to as transpiration throughout this manuscript, it also contains soil evaporation. However, no explicit and quantitative distinction could be made between these two fluxes with the available data. A further critical assumption of the above method required that roots do not tap the groundwater and that water for transpiration is exclusively extracted from the unsaturated soil. In contrast to other landscapes (Fan et al., 2017), it is likely that this assumption largely holds in the Wüstebach as throughout the catchment the groundwater levels, also in the riparian zone, remains largely below a depth of 50 cm during the relatively dry growing season (Bogena et al., 2015) when storage deficits S_D typically accumulate (\sim May to October) and no roots have so far been observed for the dominant *picea* species below that depth in the Wüstebach catchment. This is also broadly consistent with the results of Evaristo and McDonnell (2017), who show rather limited groundwater use by *picea* species.

4.2 Model architecture

A semi-distributed, process-based catchment model, iteratively customized and tested within the previously developed DYNAMITE modular modelling framework (Hrachowitz et al., 2014; Fovet et al., 2015), was adapted with additional, hydrologically passive storage volumes to allow for simultaneous representation of water fluxes and tracer transport (Hrachowitz et al., 2013) based on the general concept of storage-age selection functions (SAS; Rinaldo et al., 2015).

4.2.1 Hydrological model

The model domain of the Wüstebach catchment was spatially discretized into two functionally distinct response units, i.e. hillslopes and riparian areas. These are represented in the model as two parallel suites of storage components, linked by a common groundwater body as shown in Figure 3 (e.g. Euser et al., 2015; Nijzink et al., 2016b). According to elevation data and distribution of soil types (Fig.1), 90% of the catchment area was classified as hillslope and the remaining 10% as riparian area. Below a threshold temperature T_T [°C] precipitation P [mm d⁻¹] accumulates as snow P_S [mm d⁻¹] in S_{Snow} [mm]. Above that temperature precipitation is falling as rain P_R [mm d⁻¹] and snow melt P_M [mm d⁻¹] is released from S_{Snow}



according to a melt factor F_M [$\text{mm d}^{-1} \text{ } ^\circ\text{C}^{-1}$] using a simple degree-day method (e.g. Arsenault et al., 2015; Ala-aho et al.,
 255 2017; Gao et al., 2017). The total liquid water input $P_R + P_M$ [mm d^{-1}] entering the hillslope is routed through the canopy
 interception storage $S_{I,H}$ [mm]. Water that is not evaporated as $E_{I,H}$ [mm d^{-1}] enters the unsaturated root-zone $S_{U,H}$ [mm],
 whose storage capacity is defined by the calibration parameter $S_{U,max,H}$ [mm]. Water can be released from $S_{U,H}$ as combined
 root-zone transpiration and soil evaporation flux $E_{T,H}$ [mm d^{-1}] or eventually recharge the groundwater $S_{S,a}$ [mm] over a fast,
 preferential recharge pathway as $R_{F,H}$ [mm d^{-1}] and a slower percolation flux $R_{S,H}$ [mm d^{-1}]. Similarly, water entering the
 260 riparian zone, i.e. $P_R + P_M$ [mm d^{-1}], is routed through $S_{I,R}$ [mm]. Excess water $P_{E,R}$ [mm d^{-1}] that is not evaporated infiltrates
 into the unsaturated root-zone $S_{U,R}$ [mm], defined by calibration parameter $S_{U,max,R}$ [mm]. In addition, a fraction of the
 upwelling groundwater $R_{S,R}$ [mm d^{-1}] replenishes $S_{U,R}$ in the riparian zone, while the remainder Q_S [mm d^{-1}] drains directly
 into the stream. While water stored in $S_{U,R}$ is available for transpiration (and soil evaporation) $E_{T,R}$ [mm d^{-1}], water that
 cannot be held is released as $R_{F,R}$ [mm d^{-1}] to a fast responding reservoir $S_{F,R}$ [mm] from where it is reaches the stream as Q_R
 265 [mm d^{-1}]. The relevant model equations can be found in Table 1.

4.2.2 Tracer transport model

The $\delta^{18}\text{O}$ composition of water fluxes and storages was tracked through the model using the storage age selection approach
 (SAS; Rinaldo et al., 2015), which allows a catchment-scale description of conservative transport based on time-variant
 travel time distributions. The method builds on the fact that a water volume S [mm] stored in any storage component can, at
 270 any moment t [d], consist of parcels of water of different age T [d]. The composition of ages in the stored volume at t
 depends on the history of water inflows and outflows. Consequently, it evolves over time as new inputs enter into and
 outflows are released from the storage component, whereby each inflow I [mm d^{-1}] and outflow volume O [mm d^{-1}] can have
 a different age composition. A convenient way to implement the SAS approach is the use of age-ranked storage $S_T(T,t)$
 [mm], which represents, “at any time t the cumulative volumes of water in a storage component as ranked by their age T ”
 275 (Benettin et al., 2017). Similarly, decomposing each inflow and outflow of a storage component into their respective
 cumulative, age-ranked volumes $I_T(T,t)$ and $O_T(T,t)$ [mm d^{-1}], respectively, then allows to update the age-ranked storage
 $S_T(T,t)$ at each time step according to the general water age balance (Botter et al., 2011; van der Velde et al., 2012; Benettin
 et al., 2015a, 2017; Harman, 2015):

$$280 \quad \frac{\partial S_{T,j}(T,t)}{\partial t} + \frac{\partial S_{T,j}(T,t)}{\partial T} = \sum_{n=1}^N I_{T,n,j}(T,t) - \sum_{m=1}^M O_{T,m,j}(T,t) \quad (\text{Eq.36})$$

where the term $\partial S_T / \partial T$ represents the aging of water in storage. Reflecting the slightly more abstract approach by Rodriguez
 and Klaus (2019) and similar to previous studies based on the functionally equivalent mixing coefficient approach (e.g.
 Fenicia et al., 2010; McMillan et al., 2012; Birkel and Soulsby, 2016; Hrachowitz et al., 2015), the water age balance is here



285 individually formulated for each storage reservoir j (e.g. $S_{l,H}$, $S_{u,H}$, etc.), which each can have varying numbers N and M of
 inflows I (e.g. P_R , P_M , $R_{S,H}$, etc.) and outflows O (e.g. P_M , $R_{S,H}$, Q_S , etc.), respectively (see Figure 3). It is assumed that the
 entire volume of a precipitation signal $P(t)$ entering the system at t has an age T of zero so that the associated $I_{T,P,j}(T,t) =$
 $P_T(T,t) = P(t)$ for all T . As all other inflows to any following storage component in the system are outflows of storage
 components prior in the sequence (see Figure 3), the corresponding $I_{T,n,j}(T,t)$ entering a storage component are identical to
 290 the $O_{T,m,j}(T,t)$ released from the storage component above.

Each age-ranked outflow $O_{T,m,j}(T,t)$ of a specific storage component j depends on the outflow volume $O_{m,j}(t)$ along this
 outflow pathway and the cumulative age distribution $P_{o,m,j}(T,t)$ of that outflow:

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$$O_{T,m,j}(T,t) = O_{m,j}(t)P_{o,m,j}(T,t)$$

(Eq.37)

The outflow volume $O_{m,j}(t)$ is estimated via the hydrological model (see Section 4.2.1; Figure 3) and thus assumed to be
 known. In contrast, the cumulative age-distribution $P_{o,m,j}(T,t)$ can in general not be directly parametrized, as it depends on the
 temporally varying age distribution of water in the storage component j represented by $S_{T,j}(T,t)$ and thus on the history of past
 300 inflows and outflows (Botter et al., 2011; Harman, 2015). Instead, it is possible to define a SAS function $\omega_{o,m,j}$ (or $\Omega_{o,m,j}$ in its
 cumulative form) for each outflow m from each storage component j that describes how outflow is sampled (or selected)
 from the temporally varying water volumes of different age present in the age-ranked storage $S_{T,j}(T,t)$ at any time t :

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$$P_{o,m,j}(T,t) = \Omega_{o,m,j}(S_{T,j}(T,t), t)$$

(Eq.38)

From the cumulative age-distribution $P_{o,m,j}(T,t)$ the associated probability density function, which represents the outflow age
 distribution $p_{o,m,j}(T,t)$, frequently also referred to as backward travel time distribution of that outflow (TTD; e.g. Benettin et
 al., 2015a; Wilusz et al., 2017), can be obtained according to:

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$$p_{o,m,j}(T,t) = \omega_{o,m,j}(S_{T,j}(T,t), t) \frac{\partial S_{T,j}}{\partial T}$$

(Eq.39)

Note that conservation of mass requires that any SAS function $\omega_{o,m,j}$ integrates to the total storage volume $S_j(t)$ present in j at
 any time t . To avoid the resulting need for rescaling $\omega_{o,m,j}$ at each time step, it is helpful to normalize the age-ranked storage
 to $S_{T,norm,j}(T,t) = S_{T,j}(T,t)/S_j(t)$ so that it remains bounded to the interval $[0,1]$ and defines a residence time distribution (RTD).
 For this study beta distributions, which are conveniently bound between the limits $[0,1]$ and defined by two shape parameters
 315 α and β , were used as SAS functions $\omega_{o,m,j}$ to sample water of different age for outflows from storage components. The
 parameters β were fixed at a value of 1 for all SAS functions $\omega_{o,m,j}$ used here. However, there is substantial evidence for
 preferential flow through macropores in the shallow subsurface (e.g. Weiler and Naef, 2003; Zehe et al., 2006, 2007; Weiler



and McDonnell, 2007; Beven, 2010; Beven and Germann, 2013; Klaus et al., 2013; Angermann et al., 2017; Loritz et al., 2017). Such preferential flow can, with increasing wetness, increasingly bypass water volumes stored in small pores with
 320 little exchange (Sprenger et al., 2016, 2018, 2019a; Cain et al., 2019; Evaristo et al., 2019; Knighton et al., 2019). This then leads to an increasing preferential release of younger water as the system becomes wetter (Brooks et al., 2010). To mimic this, the shape parameters α of the preferential fluxes $R_{F,H}$ and $R_{F,R}$ released from the two unsaturated root-zone storage components $S_j = S_{U,H}$ and $S_{U,R}$ (Figure 3), were allowed to vary as a function of the water volumes stored in $S_{U,H}$ and $S_{U,R}$, respectively (Hrachowitz et al., 2013; van der Velde et al., 2015):

325

$$\alpha_{m,j}(t) = 1 - \left(\frac{S_j(t)}{S_{U,max,j}} (1 - \alpha_0) \right) \quad (\text{Eq.40})$$

Where α_0 is a calibration parameter representing a lower bound so that $\alpha_{m,j}(t)$ can vary between α_0 and 1. A value of $\alpha_{m,j} = 1$ indicates complete mixing in dry conditions. Any value below that entails incomplete mixing and thus increases the
 330 preference towards releasing younger water in wet conditions (Benettin et al., 2017). Although there is evidence for the presence of preferential flow in other components of the system, such as in the groundwater (e.g. Berkowitz and Zehe, 2020), initial model testing suggested that the inclusion of the additional calibration parameters is not warranted by the available data. For simplicity and following the principle of model parsimony we assumed complete mixing for all other outflows from all other storage components (Figure 3; cf. Fenicia et al., 2010; Rodriguez et al., 2018). Parameter α was
 335 therefore fixed to value of 1 for these SAS functions.

The $\delta^{18}\text{O}$ precipitation input signals had to be damped to the level of fluctuation observed in the stream. To achieve this a calibration parameter defining a hydrologically passive storage volume $S_{S,p}$ [mm] was added to the active groundwater storage $S_{S,a}$. (Zuber, 1986; Hrachowitz et al., 2015, 2016), so that $S_{S,tot} = S_{S,a} + S_{S,p}$ (Figure 3). While $dS_{S,p}/dt = 0$, the age-ranked groundwater storage was computed as $S_{T,S,tot}$ and the outflows from the groundwater component consequently
 340 sampled from the entire storage volume $S_{S,tot}$.

Each individual volume with different age in $I_{T,n,j}(T,t)$ and, as a consequence, also in $S_{T,j}(T,t)$ is also characterized by a different tracer concentration $C_{I,n,j}(I_{T,n,j}(T,t),t)$ and $C_{S,j}(S_{T,j}(T,t),t)$, respectively. For a conservative tracer such as $\delta^{18}\text{O}$ that is not significantly affected by decay, evapoconcentration, retention or any other biogeochemical transformation (e.g. Bertuzzo et al., 2013; Benettin et al., 2015b; Hrachowitz et al., 2015) the concentration $C_{O,m,j}(t)$ in any outflow at any time t can then
 345 be obtained from:

$$C_{O,m,j}(t) = \int_0^{S_j} C_{S,j}(S_{T,j}(T,t),t) \varpi_{O,m,j}(S_{T,j}(T,t),t) dS_T \quad (\text{Eq.41})$$



In contrast to other regions (e.g. Soulsby et al., 2017), isotopic fractionation in the Wüstebach was previously observed to
350 mostly affect canopy interception evaporation (Stockinger et al., 2015). Fractionation was therefore here accounted for in the
two interception storage components ($S_{I,H}$, $S_{I,R}$). Following the approach described by Birkel et al. (2014), the respective $\delta^{18}\text{O}$
compositions $C_{S,S_{I,H}}$ and $C_{S,S_{I,R}}$ were accordingly updated for every time step.

Due to data availability, age tracking was here limited to 4 years in the pre- and 3 years in the post-deforestation period. For
age beyond that it can only be said that water is older than these 4 and 3 years, respectively. The RTDs and TTDs reported
355 hereafter are thus truncated at these ages.

4.3 Model calibration and post-calibration evaluation

The model was run with a daily time step and has a total of 14 free calibration parameters, which were calibrated for the
model to simultaneously reproduce flow and $\delta^{18}\text{O}$ dynamics in the stream. The uniform prior parameter distributions (Table
2) were sampled using a Monte Carlo approach with 10^6 realizations. To limit equifinality (Beven, 2006) and to ensure
360 robust posterior parameter distributions for a meaningful process representation, an extensive multi-objective calibration
strategy was applied. Briefly, this was done using a total of 14 performance metrics that describe the model's skill to
reproduce different signatures associated to streamflow (E_Q) and $\delta^{18}\text{O}$ dynamics ($E_{\delta^{18}\text{O}}$) as shown in Table 3.

Combining these metrics into two equally weighted classes describing stream and $\delta^{18}\text{O}$ dynamics, respectively, solutions
with balanced overall model performances were then obtained using the mean Euclidean Distance D_E [-] from the “perfect”
365 model (i.e. $D_E = 1$; Hrachowitz et al., 2014; Hulsman et al., 2020):

$$D_E = 1 - \sqrt{\frac{1}{2} \left(\frac{\sum_{n=1}^N (1 - E_{Q,n})^2}{N} + \frac{\sum_{m=1}^M (1 - E_{\delta^{18}\text{O},m})^2}{M} \right)}$$

(Eq.42)

Where N is the number of different performance metrics describing streamflow and M the number of different performance
370 metrics for $\delta^{18}\text{O}$. To construct the posterior parameter distributions and the corresponding model uncertainty intervals, the
retained parameter sets were then weighted according to a likelihood measure $L = D_E^p$ (cf. Freer et al., 1996), where the
exponent p was set to a value of 2 to emphasize models with good overall calibration performance.

In a first step, the model was calibrated for the pre-deforestation period 01/10/2009 – 31/08/2013. Note that due to a lack of
regular and weekly $\delta^{18}\text{O}$ precipitation data before 01/10/2010, the performance metric $E_{\delta^{18}\text{O}}$ describing the $\delta^{18}\text{O}$ dynamics
375 was computed from that date onwards only. The feasible parameter sets were then used to test the model without further
calibration in the post-deforestation period. In a second step, the model was re-calibrated for the 01/09/2013 – 30/09/2016
post-deforestation period and the changes in the resulting model performance and posterior distributions compared to those
from the pre-deforestation calibration. The estimation of the effects of deforestation on RTDs and TTDs is based on model
parameter sets obtained from calibration in the pre-deforestation and post-deforestation periods, respectively.



380

5 Results and Discussion

5.1 Deforestation effects on the hydrological system

Initial analysis of water balance data suggests that the hydro-meteorological conditions as expressed by the aridity index $I_A = \overline{E_P}/\overline{P}$, do not show significant differences between the pre-deforestation ($I_A = 0.50 \pm 0.02$) and the post-deforestation periods ($I_A = 0.51 \pm 0.03$), respectively (Figure 4a). However and in spite of these comparable climatic conditions, the data show a shift in the partitioning of water fluxes between runoff Q and actual evaporation E_A (note that $E_A = E_I + E_T$). While the fraction of precipitation that was released into the atmosphere as vapour was reduced ($\overline{E_A}/\overline{P}$; Figure 4a), the mean runoff ratio ($C_R = 1 - \overline{E_A}/\overline{P}$) increased correspondingly from $C_R = 0.55 \pm 0.04$ to $C_R = 0.68 \pm 0.03$ after deforestation of 21 % of the catchment with $p = 0.049$ based on a Wilcoxon rank sum test. These results correspond well with the findings of an earlier study in the Wüstebach, based on a shorter study period (2011 – 2015; Wiekenkamp et al., 2016), which estimated an increase of C_R from ~ 0.58 to ~ 0.66 during that period using eddy-covariance measurements. In absolute terms this entails that, notwithstanding rather stable mean annual precipitation $P = 1269 \pm 24 \text{ mm yr}^{-1}$ and potential evaporation $E_P = 632 \pm 9 \text{ mm yr}^{-1}$ over the entire study period, the annual actual evaporation E_A decreased from $576 \pm 11 \text{ mm yr}^{-1}$ to $401 \pm 6 \text{ mm yr}^{-1}$ whereas annual runoff Q increased by $\sim 25 \%$ from $694 \pm 47 \text{ mm yr}^{-1}$ to $870 \pm 63 \text{ mm yr}^{-1}$.

The overall pattern found here also broadly reflect the effects of land cover/use change in many different environments (Creed et al., 2014; Jaramillo and Destouni, 2014; Renner et al., 2014, van der Velde et al., 2014; Moran-Tejada et al., 2015; Nijzink et al., 2016; Zhang et al., 2017; Jaramillo et al., 2018). The vast majority of these studies suggest that forest removal leads to an increase in the runoff ratio C_R at the cost of reduced evaporation E_A , although the magnitudes of these changes do substantially vary between individual catchments and studies, which is consistent with our physical understanding of the importance of forest for transpiration in hydrological systems. Under the assumption that reduction of E_A is largely a direct consequence of forest removal in the Wüstebach, a plausible hypothesis to directly attribute this shift in water partitioning from E_A to Q to a physical process can be formulated as follows: the roots of harvested trees stopped extracting water for transpiration from the subsurface. In addition, the decrease of turbulent exchange of vapour with depth effectively limits soil evaporation to the first few centimetres of the soil (e.g. Brutsaert, 2014). Thus, the felling of trees led to a situation where under comparable atmospheric water demand E_P , water volumes held at depths below that and previously within the reach of active roots became largely unavailable for transpiration and evaporation after deforestation. This implies that the water volumes accessible to satisfy atmospheric water demand, i.e. $S_{U,max}$ and I_{max} , are drastically reduced.

This becomes evident when comparing the catchment-scale maximum annual storage deficits $S_{D,j}$ (Eq. 2) of the pre- and post-deforestation periods, respectively, which are indicative of differences in soil depths affected by E_A in the two periods.

In spite of similar climatic conditions, the mean annual maximum storage deficit in the pre-deforestation period is



significantly higher ($p = 0.047$) than in the post-deforestation period. In the pre-deforestation period values between 105 ± 23 mm for $I_{max} = 0$ mm and 95 ± 21 mm for $I_{max} = 4$ mm, respectively, were found (Figure 4b). Whereas in the post-deforestation period the mean storage deficit only reached between 49 ± 10 mm and 33 ± 7 mm for the same values of I_{max} (Figure 4b). Note that in both periods, $S_{D,j}$ is relatively insensitive to the magnitude of I_{max} (cf. Gerrits et al., 2009). From the
415 above maximum annual storage deficits $S_{D,j}$, the corresponding catchment-scale vegetation-accessible water storage capacity, assuming vegetation adaptation to dry conditions with 20-year return periods (see Section 4.1), was estimated at values of $S_{U,max} = 225 \pm 62$ mm for the pre-deforestation ($R^2 = 0.91$, $p = 0.04$; Figure 4c) and $S_{U,max} = 90 \pm 149$ mm for the post-deforestation period ($R^2 = 0.83$, $p = 0.27$; not shown). Note that in particular the estimates for the post-deforestation period need to be understood as merely indicative because of only 3 years of data, and a system that is likely to be far from
420 equilibrium, because the deforested part cannot have adapted yet (e.g. Nijzink et al., 2016; Teuling and Hoek van Dijke, 2020). These considerable uncertainties are also reflected in the surprisingly low post-deforestation $S_{U,max}$. Notwithstanding these limitations, the above results illustrate that here the reduction of transpiration due to deforestation is likely to go hand in hand with a considerably reduction of $S_{U,max}$ and thus the catchment-scale sub-surface pore volume (between field capacity and permanent wilting point) that is actively accessed by vegetation to satisfy the evaporative demand.

425 5.2 Deforestation effects on the catchment model

5.2.1 Model calibration for pre-deforestation period

The model parameter sets retained as feasible after calibration in the 2009-2013 pre-deforestation period reproduce the general features of the hydrograph in that period rather well (Figure 2c), similar to a previous modelling study (Cornelissen et al., 2014). This is true for both, the timing and magnitudes of high flows, with an associated Nash-Sutcliffe Efficiency
430 $E_{NS,Q} = 0.79$ for the best performing model in terms of D_E (Figure 5a) but also for low flows ($E_{NS,\log(Q)} = 0.79$), with the exception of some overestimation in summer 2011. In addition, the model could also simultaneously mimic most other observed flow signatures reasonably well (Figure 5a), in particular the flow duration curve ($E_{NS,FDC} = 0.89$; Figure 5b), the peak distribution ($E_{NS,PD} = 0.91$; Figure 5d), the auto correlation function ($E_{NS,AC} = 0.90$; Figure 5f) and the runoff ratio ($E_{R,CR} = 0.97$). Similarly, the model captures the substantial attenuation of the precipitation $\delta^{18}O$ variability, while at the same time
435 largely preserving the limited but visible low-frequency temporal fluctuations in the stream $\delta^{18}O$ composition (Figure 2d). In comparison to the flow performance metrics the Nash-Sutcliffe Efficiency of the $\delta^{18}O$ composition for the best model is somewhat lower ($E_{NS,\delta^{18}O} = 0.38$; Figure 5a), which mostly results from the low variability of such a damped signal, where even very small absolute errors and a few scattered outliers can lead to very low Nash-Sutcliffe Efficiencies (cf. Hrachowitz et al., 2009).
440 The posterior distributions (Table 2, Figure 6) show that most model parameters are reasonably well identified. Individually calibrated for their respective landscape class, i.e. hillslope and riparian zone, $S_{U,max,H} = 246$ mm (5/95th IQR: 233 – 309 mm) and $S_{U,max,R} = 234$ mm (194 – 287 mm) showed similar optimal values and distributions (Figures 6a,b), reflecting the



catchment-wide relatively homogenous forest cover in the pre-deforestation period (Figure 1). Remarkably, these values also come close to the water balance-derived catchment-scale estimates of $S_{U,max} = 225 \pm 62$ mm, as described in 5.1 (Figure 4c).

445 5.2.2 Application of pre-deforestation model to post-deforestation period

In a next step, the parameter sets obtained from the above calibration in the pre-deforestation period were used to run the model without further re-calibration in the post-deforestation period. This entails the implicit and clearly wrong assumption that the physical characteristics of the system remained unaffected by deforestation. The consequence of that can be seen in Figure 2c (red line). While the low flows remain well reproduced, the post-deforestation application of the model substantially and systematically underestimates high flows, partly by 50% or more, such as in November 2013 or August 450 2014. The inability of the model to reproduce post-deforestation high-flow dynamics of the system is also evident in the partly much lower model performance metrics associated with high flows (Figure 5a). Besides the time series of flow ($E_{NS,Q} = 0.63$), notably the model's skill to capture the peak distribution ($E_{NS,PD} = 0.81$; Figure 5e), the autocorrelation function ($E_{NS,AC} = 0.71$; Figure 5g) and the runoff ratio ($E_{R,CR} = 0.73$) were negatively affected. In contrast to the pre-deforestation 455 period, the modelled runoff ratio $C_R = 0.52$ ($0.48 - 0.67$) in the post-deforestation period considerably underestimates the observed $C_R = 0.68 \pm 0.03$ (Figure 4a). The above implies that the model also overestimates post-deforestation evaporative fluxes E_A . Therefore, it can, without re-calibration, not deal with the observed changes in the partitioning between drainage and evaporative fluxes (Figure 4a). A likely explanation for the pattern produced by the model is that, in contrast to the real world, no reduction in E_A due to the reduced forest cover is achieved because the model still relies on the catchment-scale 460 vegetation-accessible storage volume $S_{U,max}$ that characterizes the extent of the catchment-scale active root-system before deforestation. This $S_{U,max}$ falsely provides sufficient water supply to sustain E_A at high levels comparatively close to E_P throughout the year (see red line in Figure 2b), although, in the parts of the catchment where trees were removed, water stored at depths below a few centimetres is not available for significant evaporation anymore. Such an overestimation of $S_{U,max}$ implies also that in the model a more pronounced water storage deficit can and does develop throughout dry periods. 465 The model therefore assumes that soils dry out to deeper depths. Consequently, to establish connectivity and to eventually generate flow during and after rainstorms, more water needs to be stored in the model than in the real world system to overcome this deficit. This water is then in the model held against gravity and thus only available for evaporation but not for drainage. Although it is reasonable to assume that groundwater recharge is affected in a similar way, the model can better reproduce low flows. The reason for this is that the draining groundwater body, which sustains summer low flows, is, due to 470 limited recharge during these drier periods, largely disconnected from and thus largely unaffected by subsurface – vegetation interaction in shallower parts of the subsurface. In the parts of the catchment where trees were removed a similar reasoning also holds for the interception capacity I_{max} and the associated likely overestimation of interception evaporation E_I , yet, due to the smaller magnitude of I_{max} , to a lesser extent than for $S_{U,max}$. The above described problems for the high flow periods are accompanied by the model's inability to describe the post-deforestation $\delta^{18}O$ dynamics in stream water ($E_{NS,\delta^{18}O} < 0$). While 475 this is partly an effect of the above explained low signal-to-noise ratio of such a damped signal and thus of the chosen



performance metric, the model also struggles to adequately reproduce the low-frequency fluctuations, such as between February and July 2014, when the model indicated rather stable $\delta^{18}\text{O}$ values while the observed values show a slight yet clear decrease over the same period (not shown). This is also underlined by the more than doubling of the mean absolute errors of $\delta^{18}\text{O}$ from $\text{MAE} = 0.09 \text{ ‰}$ in the pre-deforestation period to $\text{MAE} = 0.21 \text{ ‰}$ in the post-deforestation period. Together with
480 the significant lower overall model performance metric D_E (Figure 5a), these results illustrate that the pre-deforestation model parameter sets provide an unsuitable characterization of the system characteristics in the post-deforestation period.

5.2.3 Recalibrate model for post-deforestation period

To estimate the effect of forest removal on the characteristics of the hydrological system and thus on the model parameters, the model was in a next step recalibrated for the post-deforestation period. This led to a significant improvement of the
485 overall model performance from $D_E = 0.22$ to 0.58 (Figure 5a). It can be observed that the recalibrated model can much better reproduce the increased high flows in that period (Figure 2c), as reflected by improvements in the performance metrics associated with high flows (Figure 5a), but most notably $E_{NS,Q} = 0.69$, $E_{NS,PD} = 0.93$ (Figure 5e) or $E_{NS,AC} = 0.87$ (Figure 5g). In addition and perhaps most importantly, the runoff ratio also increased and was with a modelled value of $C_R = 0.58$ ($0.56 - 0.61$) closer to the observed C_R ($E_{R,CR} = 0.84$). This further implies that, in contrast to the initial model, the recalibrated
490 model also features expected reductions of evaporative fluxes E_A by about 10%, which can be seen in Figure 2b. Mirroring the improvements in the reproduction of flows, recalibration also allowed the model to better capture the stream water $\delta^{18}\text{O}$ dynamics ($E_{NS,\delta^{18}\text{O}} = 0.24$; $\text{MAE} = 0.10 \text{ ‰}$; Figure 5a). While there is little change in the model's ability to mimic the general level of damping of the $\delta^{18}\text{O}$ signal and its low-frequency fluctuations, the more pronounced, albeit in absolute terms still small, high-frequency fluctuations, as short-term response to individual storms are better described (Figure 2d).

495 It is of course unsurprising that recalibration leads to an improved model performance in the post-deforestation period. Without further analysis, such a mere model fitting exercise allows in the presence of model equifinality only little insight into the underlying processes (Beven, 2006; Kirchner, 2006). To gain more confidence that the improvements in the recalibrated model are at least partly due to the right reasons (Kirchner, 2006), the changes in the posterior parameter distributions resulting from the two calibration runs were thus analysed. It was hypothesized above that reductions in
500 evaporative fluxes are directly linked to reduced water volumes accessible and available for evaporation and transpiration at the catchment-scale. In the theoretical ideal case, the representations of the associated storage capacities in the model, i.e. the parameters $S_{U,max}$ and I_{max} , should thus be the only ones to significantly change after deforestation. However, note that this is unlikely for two reasons. First, while it is plausible to assume that these storage capacities are significantly affected by forest removal, it is not unlikely that other system characteristics and their mutual interactions, thus far unknown and not
505 considered, are similarly influenced, potentially causing considerable ontological uncertainty. Second, model parameter interactions that arise as artefacts to compensate overly simplistic process representations and/or data uncertainty are also likely to affect parameters seemingly unrelated to deforestation.



Inspection of the posterior parameter distributions reveals that the catchment-scale $S_{U,max}$ experienced considerable reductions after recalibration. While in the hillslope parts of the catchment, which were less affected by deforestation (~ 10% of the hillslope area; Figure 1) an average decrease by ~ 75 mm to $S_{U,max,H} = 137$ mm (118 – 249 mm) can be seen (Figure 510 6a), the completely deforested riparian area exhibits an average decrease by ~ 130 mm to $S_{U,max,R} = 67$ mm (53 – 126 mm; Figure 6b). As an indicative value, the area-weighted catchment-average $S_{U,max} = 120$ mm of the best performing parameter set falls remarkably well into the plausible range of $S_{U,max} = 90 \pm 149$ mm as described in Section 5.1. Similarly, significant reductions in I_{max} can be observed, which are with average reduction of ~ 2 mm not as pronounced on the less deforested 515 hillslopes (Figure 6d) than in the riparian areas where $I_{max,R}$ decreased on average by ~ 3 mm (Figure 6e). Comparing to the posterior distributions of other parameters, the results illustrate that the storage parameters $S_{U,max}$ and I_{max} of the riparian zone, and to a lesser extent of the hillslope, were subject to the most pronounced changes. In contrast, for most other parameters, the 5/95th interquantile range of the pre- and post-deforestation posterior distributions largely remained overlapping (Figure 6). However, it can also be observed that the individual parameter values associated with the best model 520 solutions in the pre- and post-deforestation periods, respectively, do vary to a stronger degree for most parameters. Notwithstanding the distinct overall effects of forest removal on the individual posterior distributions, this clearly highlights the influence of parameter compensation effects and related uncertainties. After preliminary unsuccessful testing, no further attempts were thus made to re-calibrate only the above discussed four storage parameters, i.e. $S_{U,max,H}$, $S_{U,max,R}$, $I_{max,H}$ and $I_{max,R}$.

525 Overall the results suggest that the model formulation together with the multi-objective calibration strategy ensured the identification of solutions that provide a robust description of the system and allow a simultaneous representation of flow and isotope dynamics in the stream. There are indications that at least some processes and parameters can be directly linked to real world quantities. In particular, the results provide supporting evidence that the parameters $S_{U,max,H}$ and $S_{U,max,R}$ are not merely abstract quantities, but that it is not implausible to assume that they, taken together, provide a catchment-scale 530 representation of vegetation-accessible and -accessed water volumes as defined by Equation 2.

5.3 Deforestation effects on travel time distributions, SAS-functions and young water fractions

While the volume weighted mean $\delta^{18}\text{O}$ contents of observed precipitation with -7.9 ‰ and stream water with -8.2 ‰ are comparable, a substantial difference in their respective fluctuations, with standard deviations of 3.6 ‰ and 0.2 ‰, respectively, is evident (Figure 2d). This difference suggests a remarkably elevated degree of damping rarely found 535 elsewhere (e.g. Speed et al., 2010), indicative of the importance of old water contributions to the stream in the study catchment. No significant difference in damping ratios was observed between the pre- and post-deforestation period, which further corroborates the prevalence of old water.

Tracking the $\delta^{18}\text{O}$ signals through the model then allowed to estimate residence (RTD) and travel time distributions (TTD). Note that any results reported hereafter are necessarily conditional on the assumptions made in and the uncertainties arising 540 from the modelling process.



In general and consistent with the observed high degree of damping, it was found that pre-deforestation the system was characterized by rather old water. On average only ~ 16% of the water stored in the system is younger than three years, as illustrated by the range of the truncated marginal RTDs that combine water stored in all model storage components (Figure 7b). The young water fractions (F_{yw}), here defined as the fractions of water younger than three months (Kirchner, 2016), are
545 similarly low but exhibit somewhat more pronounced variations from < 0.01 to 0.04 between dry and wet periods. In contrast, the range of truncated TTDs of stream water not only shows more variability in response to changing wetness conditions but also somewhat younger water, with on average about 24 % of the discharge younger than 3 years (Figure 7b). Stream water can contain F_{yw} of up to ~ 0.30 for individual storm events in the wet period, while frequently dropping to < 0.01 during elongated summer dry periods (Figures 7c, 8a), similar to what has been reported elsewhere (e.g. Gallart et al.,
550 2020b). It can also be observed that the age composition of stream water (Figure 7c) and the associated F_{yw} (Figure 8a) do considerably vary throughout wet periods. Dry periods are characterized by considerably less variability and more stable stream water TTDs. This is a consequence of increased bypass flow that has little interaction with resident water as the system gets wetter and which may reach the stream over preferential flow paths and increased contributions from the riparian zone with its shorter flow paths. In other words, in a wet system where little additional water can be stored, the precipitation
555 volumes of individual storm events control the shape of TTDs (Heidbüchel et al, 2020). In the summer dry season, however, precipitation is to a higher degree buffered in the root-zone and used for transpiration (Stockinger et al., 2014). Conversely, stream flow is then mostly sustained by groundwater which is characterized by large volumes of older water. This effectively attenuates fluctuations by the proportionally much lower volumes of younger precipitation water that cannot be stored and is thus quickly released to the stream. This is further corroborated by the significantly higher sensitivity of F_{yw} to changes in
560 stream flow in wet periods as compared to dry periods (Figure 8c). In spite of the low mean F_{yw} ~ 0.11 (Figure 8a), the above also entails that very fast switches towards higher young water fractions can be observed when the system is wetting up after dry periods as well as for storm events throughout the wet season. In general, the above observations are also encapsulated in the catchment-overall storage age selection functions ω , that represent the ratio of stream water TTD over the combined RTD of all model storage elements (Benettin et al., 2015a). While for dry periods under-sampling of young water ages with
565 relatively little variability is evident, it can also be seen that in particular during wet-up and wet periods a considerable, yet highly variable preference for very young water can be seen (Figure 9a).

The overall picture did not change in the post-deforestation period. Water stored in the subsurface remains to be characterized by RTDs with very low young water fractions F_{yw} between < 0.01 and 0.04 with limited sensitivity to changes in wetness conditions (Figure 7i). Similar to the pre-deforestation period, the TTDs exhibit much more variability compared
570 to RTDs, depending on the wetness conditions. However, in contrast to the pre-deforestation period and irrespective of the wetness conditions, partly considerable shifts towards younger water can be observed for the TTDs (Figure 7d-g). While individual summer storms led to increases of almost exclusively very young water <10 – 20 days in the stream (Figure 7d), considerable shifts towards younger water can be observed throughout the entire spectrum of tracked ages during wet-up and wet conditions (Figure 7e-f). During the wet period ~ 28 % of the stream water are on average younger than the tracked three



575 years (Figure 7i). The mean F_{yw} only slightly increased to 0.13 (Figure 8b), compared to 0.11 in the pre-deforestation period
(Figure 8a). For individual winter storm events, however, F_{yw} increased to up to ~ 0.40 (Figures 7j, 8b) compared to F_{yw} of
up to ~ 0.30 in the pre-deforestation period (Figures 7c, 8a). Besides the generally higher F_{yw} during wet periods, the F_{yw}
became more sensitive to flow during wet-up and wet conditions, with $dF_{yw}/dQ \sim 0.27$ and 0.43 , respectively (Figure 8d),
similar to what has been previously reported by von Freyberg et al. (2018) and Gallart et al. (2020a). At the end of dry
580 periods and the beginning of the wet period, elsewhere also referred to as “autumn flush” (e.g. Dawson et al., 2011), the
switches towards younger water at given flow levels occur considerably faster in the post-deforestation period than in the
pre-deforestation period. Therefore, where, at the same discharge, previously relatively little young water reached the stream,
a much higher fraction of young water can now be observed in the stream. This is a direct effect of the reduced evaporative
removal of relatively young near-surface water, which in turn is intimately linked to the reduced water supply for
585 evaporative fluxes, i.e. smaller storage volumes $S_{U,max}$ and I_{max} . This relatively young, surface-near water, not taken up by
vegetation anymore is thus to a higher degree flushed from the system mostly via fast responding flow paths to the stream.
Once connectivity and the associated higher degree of bypass flow are established in the wet period, the peak sensitivity of
 dF_{yw}/dQ to flow increased to ~ 0.43 , as under these conditions when little additional water can be stored in the shallow
subsurface, F_{yw} is largely controlled by magnitude of the individual precipitation signals and to a lesser extent by the
590 footprint of the pre-storm history of evaporative fluxes in the shallow subsurface storage. In contrast, no significant changes
could be observed for the sensitivity of F_{yw} to discharge during dry periods, as during that period, the composition of water
ages is controlled by large volumes of old water. The above described post-deforestation changes are also manifest in the
corresponding storage age selection function ω (Figure 9b) for that period. While the degree of under-sampling of young
water during dry periods significantly decreased, a substantially higher preference for young water during wet-up and wet
595 periods can be observed than during the pre-deforestation period, with a clear overall shift towards younger water for all
wetness conditions.

5.4 Uncertainties, unresolved questions and limitations

As emphasized above, all results are conditional on the assumptions taken throughout the modelling process. These
600 assumptions, present in model structure, parameterization and parameters, can lead to uncertainties. Yet, notwithstanding
these potential uncertainties, extensive preliminary model testing together with the use of multiple model calibration and
evaluation criteria suggest there is relatively strong evidence to support the main results in this study: the post-deforestation
reduction of evaporative fluxes can, at least partially, be linked to a relatively clear reduction in the catchment-scale storage
capacities $S_{U,max}$ and I_{max} , which in turn triggered a shift towards younger water ages in the stream, particularly during wet-up
605 and wet conditions.

One specific interesting observation was that the results suggest that a passive mixing volume $S_{S,p}$ of at least ~ 10.000 mm is
necessary for the model to attenuate the amplitudes of the precipitation $\delta^{18}\text{O}$ signals to those in the stream water. Although,



$S_{S,p}$ is rather well constrained (Figure 6h), there has in the past been no hydrogeological evidence for the presence of such a surprisingly large groundwater volume nor for its hydrological relevance in the study catchment. Indeed, the authors are not aware of any catchment-scale study that reported similarly high values for $S_{S,p}$ or functionally equivalent parameters (e.g. Birkel et al., 2011a,b; Hrachowitz et al., 2013,2015; Benettin et al., 2013,2015a; Harman, 2015; van der Velde et al., 2015). Yet, to achieve the degree of damping observed in the stream water, such a volume is necessary, if the current understanding of conservative tracer dynamics holds. Reflecting our insufficient knowledge to which depths exchange with surface water occurs (e.g. Condon et al., 2020), a potential explanation for this observation is that the frequently layered and fractured structure of the Devonian shale bedrock may provide relatively high-permeability pathways for the circulation of and exchange with water at depth. Another, yet, given the current understanding of the Wüstebach (e.g. Graf et al., 2014), less likely hypothesis is the presence of significant lateral groundwater exchange (e.g. Bouaziz et al., 2018). In other words the possibility that the subsurface catchment does not match with the surface catchment (Figure 1) and that older groundwater is imported from “outside” the surface catchment, while an equivalent volume of younger groundwater is exported, maintaining the mass balance. These are hypotheses to be tested in future studies, as the currently available data do not allow a conclusive answer to this question.

6 Conclusions

The small Wüstebach catchment experienced significant deforestation in 2013. Analyzing the effects of this deforestation on the hydrology and stable isotope circulation dynamics in the study catchment our main findings are:

- (1) Water balance data suggest that deforestation led to a significant increase of stream flow, accompanied by corresponding reductions of evaporative fluxes. This is reflected by an increase of the runoff ratio from $C_R = 0.55$ to 0.68 in the post-deforestation period despite similar climatic conditions, supporting previous results based on eddy covariance measurements (Wiekenkamp et al., 2016).
- (2) Based on water balance data, this reduction of evaporative fluxes, as a consequence of reduced vegetation water uptake, could at least partly be linked to a reduction of the catchment-scale water storage volume in the unsaturated soil ($S_{U,max}$) that is within the reach of active roots and thus accessible for vegetation transpiration from ~ 225 mm in the pre-deforestation period to ~ 90 mm in the post-deforestations period.
- (3) Estimating $S_{U,max}$ as calibration parameter of a process-based hydrological model led to similar conclusions. The catchment-average calibrated model parameters representing $S_{U,max}$ for both, the pre- and deforestation periods, respectively, correspond with ~ 240 mm and ~ 120 mm remarkably well with $S_{U,max}$ directly estimated from water balance data. Other model parameters, assumed to have a less direct link to vegetation, exhibited much lower levels of systematic change following deforestation.
- (4) Using the model to track the age composition of stream water suggested that, in general, water reaching the stream in the pre-deforestation period was rather old with a mean young water fraction $F_{yw} \sim 0.11$. In spite of the overall low F_{yw} , clear



640 shifts in the shape of travel time distributions towards younger water can be seen under wet conditions with young water fractions increasing up to $F_{yw} \sim 0.30$.

(5) Deforestation and the associated reduction of $S_{U,max}$ led to shifts in travel time distributions towards younger water. Under wet conditions, this resulted in increases of young water fractions to up to $F_{yw} \sim 0.40$ for individual storms. In contrast, dry period travel time distributions exhibited only minor changes. Overall the mean fraction of young water in the stream
645 increased to $F_{yw} \sim 0.13$.

(6) Deforestation resulted in a considerable increase of the sensitivity of young water fractions to discharge under wet conditions from $dF_{yw}/dQ = 0.25$ to 0.43. This implies faster switches towards younger water and thus faster routing of solutes during and shortly after storm events and thus faster routing of solutes with increasing wetness.

Overall, this study demonstrates that deforestation has not only the potential to affect the partitioning between drainage and
650 evaporation, and thus the fundamental hydrological response characteristics of catchments, but also catchment-scale tracer circulation dynamics. In particular for wet and wet-up conditions, sometimes also referred to as “autumn flush”, deforestation caused higher proportions of younger water to reach the stream, implying faster routing of water and plausibly also solutes through the subsurface.

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Author contributions. MH and MS designed the experiment. MH did the analysis and wrote the first draft. All authors discussed the design, results and the first draft and contributed to writing the final manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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Table 1: Water balance, state and flux equations used in the hydrological model. Symbols shown in bold are model parameters. Subscripts H and R indicate hillslope and riparian zone, respectively. Model variables: P is total precipitation [mm d⁻¹], P_S is solid precipitation (snow) [mm d⁻¹], P_M is snow melt [mm d⁻¹], P_R is rain [mm d⁻¹], P_E is effective precipitation [mm d⁻¹], E_P is potential evaporation [mm d⁻¹], E_I is interception evaporation [mm d⁻¹], R_F is preferential recharge [mm d⁻¹], R_S is slow recharge [mm d⁻¹], E_T is transpiration [mm d⁻¹], Q_S is flow from slow responding reservoir [mm d⁻¹], Q_R is flow from the fast responding riparian reservoir [mm d⁻¹], Q is the total flow [mm d⁻¹] and E_A is the total actual evaporation [mm d⁻¹]. Model parameters: T_T is the threshold temperature [°C], F_M is a melt factor [mm d⁻¹ °C⁻¹], I_{max} is the interception capacity [mm], $S_{U,max}$ is the root-zone storage capacity [mm], γ is a shape factor [-], $R_{S,max}$ is the maximum percolation rate [mm d⁻¹], L_p is a transpiration water stress factor [-], f_{Q_S} is a factor determining the fraction of groundwater flow that is upwelling into the riparian zone [-], k_S is the storage coefficient of the slow responding reservoir [d⁻¹], k_R is the storage coefficient for the fast responding riparian reservoir [d⁻¹] and f is the areal fraction of the riparian zone [-].

Landscape unit	Storage component	Water balance	Eq.	Constitutive equations	Eq.
	Snow storage	$dS_{snow}/dt = P_S - P_M$	(8)	$P_S = \begin{cases} P, & T < T_T \\ 0, & T \geq T_T \end{cases}$	(15)
				$P_M = \begin{cases} 0, & T < T_T \\ \min\left(F_M(T - T_T), \frac{S_{snow}}{dt}\right), & T \geq T_T \end{cases}$	(16)
Hillslope	Interception storage	$dS_{I,H}/dt = P_R + P_M - P_{E,H} - E_{I,H}$	(9)	$P_R = \begin{cases} 0, & T < T_T \\ P, & T \geq T_T \end{cases}$	(17)
				$P_{E,H} = \max\left(0, \frac{S_{I,H} - I_{max,H}}{dt}\right)$	(18)
				$E_{I,H} = \min\left(E_P, \frac{S_{I,H} - P_{E,H}}{dt}\right)$	(19)
	Unsaturated root-zone storage	$dS_{U,H}/dt = P_{E,H} - R_{F,H} - R_{S,H} - E_{T,H}$	(10)	$S'_{U,H} = (1 + \gamma)S_{U,max,H} \left(1 - \left(1 - \frac{S_{U,H}}{S_{U,max,H}}\right)^{\frac{1}{1+\gamma}}\right)$	(20)
				$R_{F,H} = P_{E,H} - \left(S_{U,max,H} + S_{U,H} + S_{U,max,H} \left(1 - \frac{P_{E,H}dt + S_{U,H}'}{(1 + \gamma)S_{U,max,H}}\right)^{(1+\gamma)}\right) dt^{-1}$	(21)
				$R_{S,H} = \min\left(R_{S,max} \frac{S_{U,H}}{S_{U,max,H}}, \frac{S_{U,H}}{dt}\right)$	(22)
				$E_{T,H} = \min\left(E_P - E_{I,H}, \min\left(\frac{S_{U,H}}{S_{U,max,H} L_p}, 1\right), \frac{S_{U,H}}{dt}\right)$	(23)
	Slow responding storage	$dS_{S,a}/dt = (1 - f)(R_{F,H} + R_{S,H}) - R_{S,R} - Q_S$	(11)	$R_{S,R} = f_{Q_S} S_{S,a} (1 - e^{-k_S t}) dt^{-1}$	(24)
				$Q_S = (1 - f_{Q_S}) S_{S,a} (1 - e^{-k_S t}) dt^{-1}$	(25)
Riparian zone	Interception storage	$dS_{I,R}/dt = P_R + P_M - P_{E,R} - E_{I,R}$	(12)	$P_{E,R} = \max\left(0, \frac{S_{I,R} - I_{max,R}}{dt}\right)$	(26)
				$E_{I,R} = \min\left(E_P, \frac{S_{I,R} - P_{E,R}}{dt}\right)$	(27)
	Unsaturated root-zone storage	$dS_{U,R}/dt = P_{E,R} + R_{S,R}/f - R_{F,R} - E_{T,R}$	(13)	$S'_{U,R} = (1 + \gamma)S_{U,max,R} \left(1 - \left(1 - \frac{S_{U,R}}{S_{U,max,R}}\right)^{\frac{1}{1+\gamma}}\right)$	(28)
				$R_{F,R} = P_{E,R} + \frac{R_{S,R}}{f} - \left(S_{U,max,R} + S_{U,R} + S_{U,max,R} \left(1 - \frac{P_{E,R}dt + S_{U,R}'}{(1 + \gamma)S_{U,max,R}}\right)^{(1+\gamma)}\right) dt^{-1}$	(29)
				$E_{T,R} = \min\left(E_P - E_{I,R}, \min\left(\frac{S_{U,R}}{S_{U,max,R} L_p}, 1\right), \frac{S_{U,R}}{dt}\right)$	(30)
	Fast responding storage	$dS_{F,R}/dt = R_{F,R} - Q_R$	(14)	$Q_R = S_{F,R} (1 - e^{-k_R t}) dt^{-1}$	(31)
				$Q = Q_S + fQ_R$	(32)
				$E_I = (1 - f)E_{I,H} + fE_{I,R}$	(33)
				$E_T = (1 - f)E_{T,H} + fE_{T,R}$	(34)
				$E_A = E_I + E_T$	(35)

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995 **Table 2: Parameter prior distributions and 5/95th percentiles of the posterior distributions. Note that *) parameter f , characterizing the areal proportion of the riparian zone was fixed according to soil and elevation data and **) the interception capacity I_{max} was assumed to be identical on the hillslopes and the riparian zone in the pre-deforestation period.**

Model	Parameter	Prior distribution	Posterior distribution	
			Pre-deforestation	Post-deforestation
Hydrological model	f [-]*	0.1	0.1	0.1
	F_M [mm d ⁻¹ °C ⁻¹]	1.0 – 5.0	2.1 – 4.2	1.8 – 4.6
	f_{QS} [-]	0.00 – 0.20	0.02 – 0.11	0.01 – 0.10
	$I_{max,H}$ [mm]	0.0 – 6.0	0.8 – 4.5	0.1 – 1.7
	$I_{max,R}$ [mm]**	0.0 – 6.0	0.8 – 4.5	0.0 – 0.9
	k_R [d ⁻¹]	0.01 – 2.00	0.20 – 1.60	0.40 – 1.40
	k_S [d ⁻¹]	0.01 – 0.15	0.04 – 0.07	0.03 – 0.09
	L_p [-]	0.0 – 1.0	0.2 – 0.7	0.1 – 0.2
	$R_{S,max}$ [mm d ⁻¹]	0.0 – 2.0	0.2 – 1.9	0.4 – 1.6
	$S_{U,max,H}$ [mm]	0 – 400	233 – 309	118 – 249
	$S_{U,max,R}$ [mm]	0 – 400	194 – 287	53 – 126
	T_T [°C]	-1.5 – 1.5	-0.6 – 1.2	-0.7 – 1.0
	γ [-]	0.0 – 5.0	0.3 – 4.2	0.7 – 4.3
Tracer model	α_0 [-]	0.00 – 1.00	0.77 – 0.99	0.58 – 0.96
	$S_{S,p}$ [mm]	1000 – 30 000	8132 – 16 457	8387 – 16314

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Table 3: Signatures of flow and $\delta^{18}\text{O}$ and the associated performance metrics used for model calibration and evaluation. The performance metrics used include the Nash-Sutcliffe efficiency (E_{NS}), the volume error (E_V) and the relative error (E_R).

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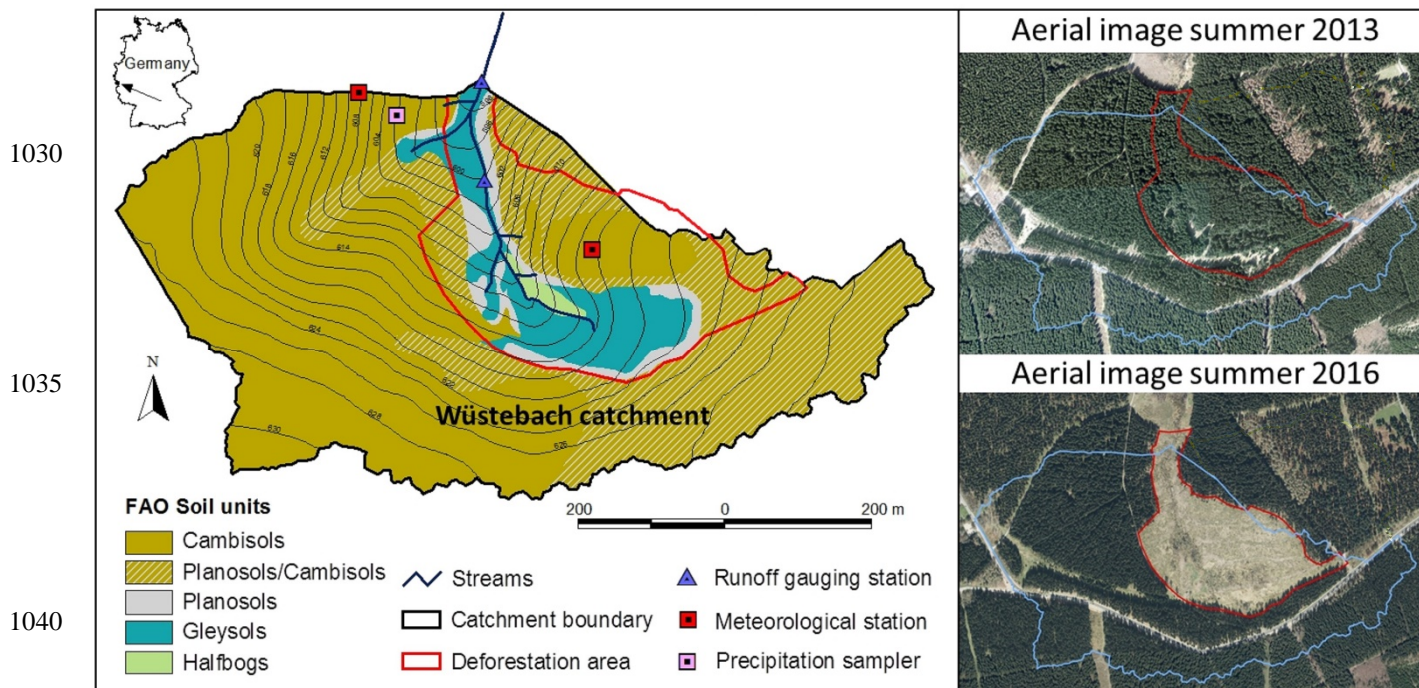
Variable/Signature	Symbol	Performance Metric	Reference
Time series of flow	Q	$E_{NS,Q}$	Nash and Sutcliffe (1970)
	$\log(Q)$	$E_{NS,\log(Q)}$	
	Q	$E_{V,Q}$	Criss and Winston (2008)
Flow duration curve	FDC	$E_{NS,FDC}$	Jothityangkoon et al. (2001)
Flow duration curve high flow period	FDC,h	$E_{NS,FDCh}$	Yilmaz et al. (2008)
Peak distribution	PD	$E_{NS,PD}$	Euser et al. (2013)
Rising limb density	RLD	$E_{R,RLD}$	Shamir et al. (2005)
Declining limb density	DLD	$E_{R,DLD}$	Sawicz et al. (2011)
Autocorrelation function of flow	AC	$E_{NS,AC}$	Montanari and Toth (2007)
Lag-1 autocorrelation	AC1	$E_{R,AC1}$	Hrachowitz et al. (2014)
Lag-1 autocorrelation low flow period	AC1,l	$E_{R,AC1,l}$	Fovet et al. (2015)
Runoff ratio	CR	$E_{R,CR}$	Yadav et al. (2007)
Time series of $\delta^{18}\text{O}$ in stream water	$\delta^{18}\text{O}$	$E_{NS,\delta^{18}\text{O}}$	Birkel et al. (2011a)

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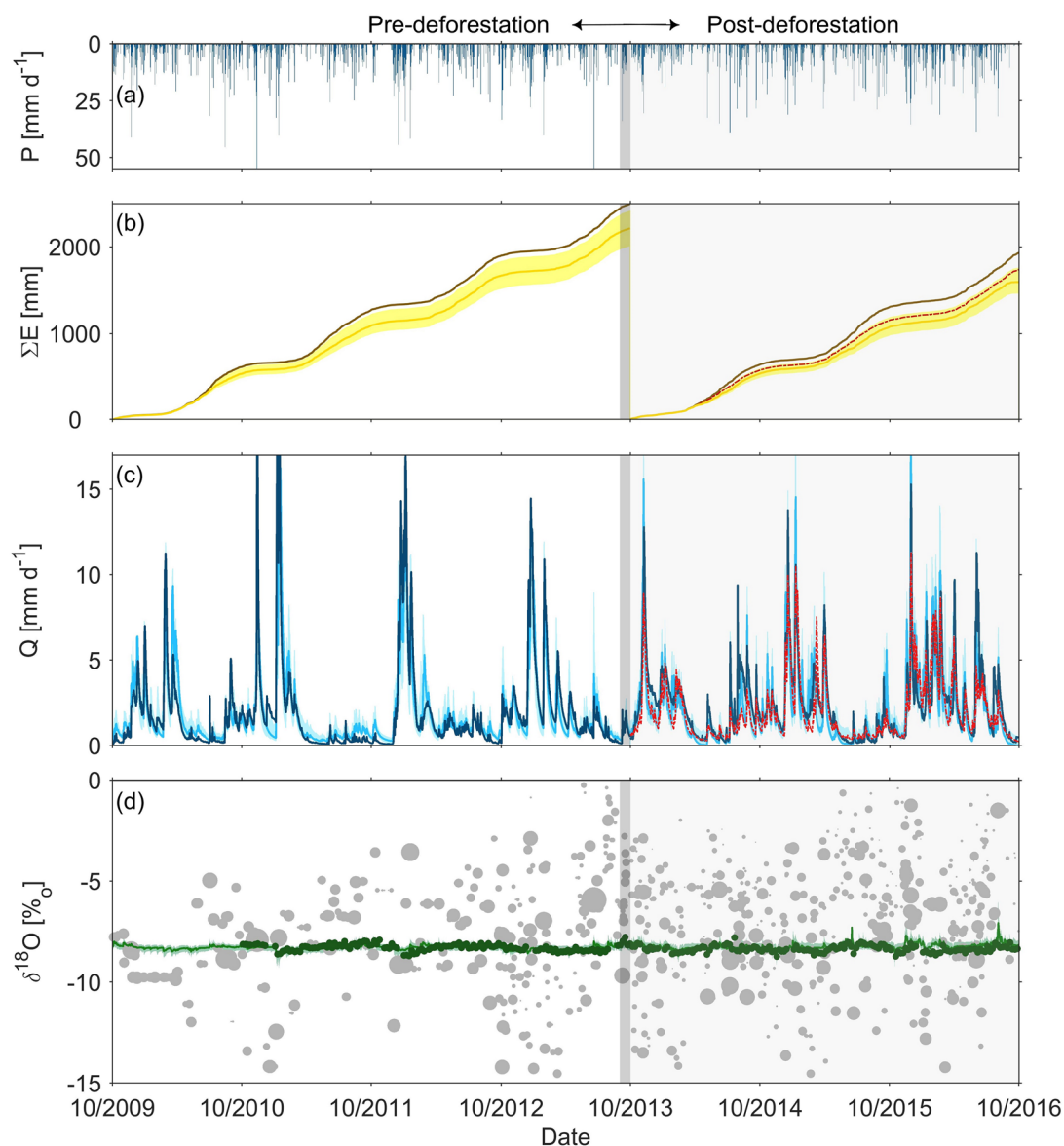


1025 **Figure 1: Map of the Wüstebach study catchment showing the spatial distribution of soil types. The riparian zone is defined by the**
parts of the catchment covered by Gleysols, Planosols and Halfbogs. The red line indicates the outline of the deforested part of the
catchment, as can also be seen on the aerial images (© Google Earth, Maxar Technologies 2020) from 2013 and 2016.





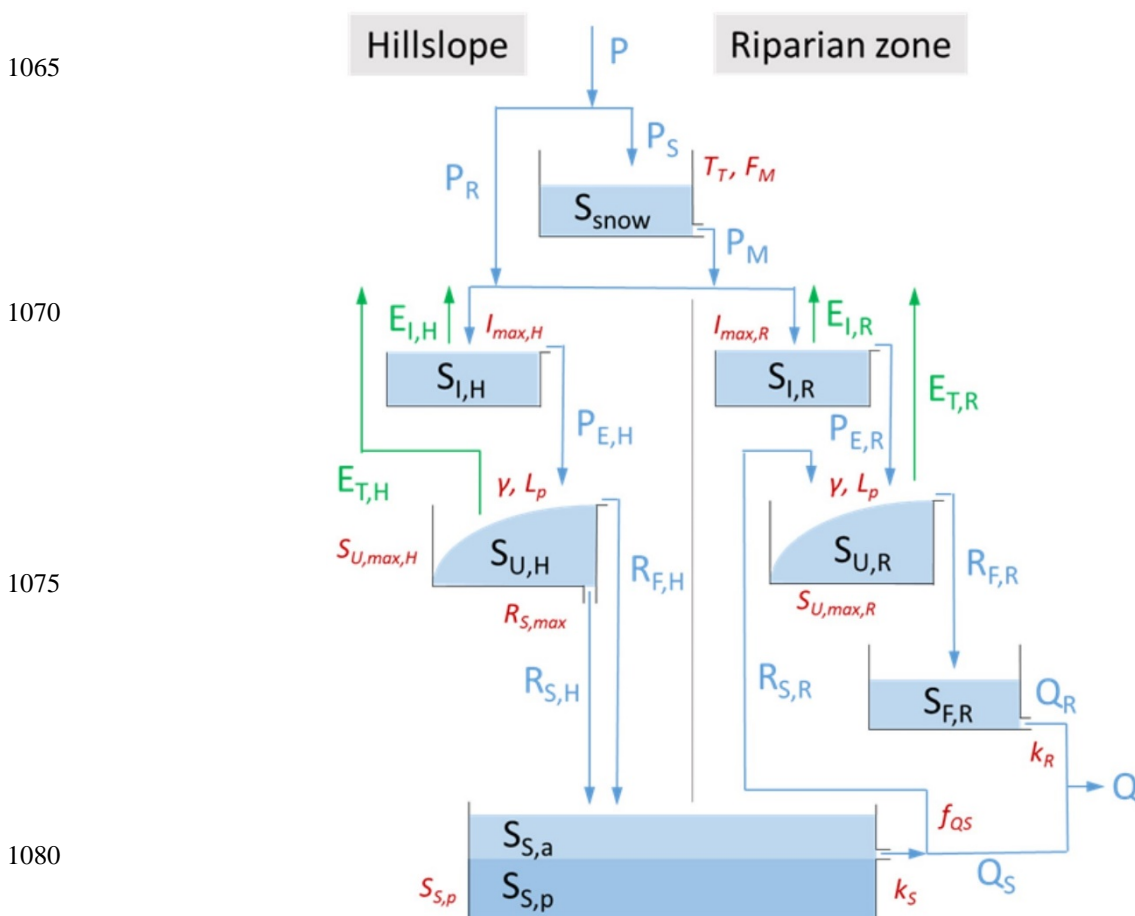
1045 **Figure 2: (a) Time series of observed daily precipitation P ; (b) daily cumulative evaporative fluxes for the pre- and post-**
deforestation period, where the dark brown line indicates potential evaporation E_P and the orange lines and the yellow shaded
areas show the actual evaporation E_A modelled using the best fit parameter sets and the associated 5/95th percentiles of all feasible
solutions of the pre- and post-deforestation periods, respectively. The dashed red line indicates the modelled E_A in the post-
deforestation period using the best fit pre-deforestation parameter set; (c) observed (dark blue line) and modelled stream flow Q ;
1050 **light blue line indicates best fit model and the shaded area the 5/95th percentile of all feasible solutions for the pre- and post-**
deforestation periods, respectively. The dashed red line indicates the modelled stream in the post-deforestation period using the
best fit pre-deforestation parameter set; (d) Observed $\delta^{18}\text{O}$ signals in precipitation (grey dots; size of dots indicates the
precipitation volume) and stream flow (green dots) as well as the modelled $\delta^{18}\text{O}$ signal in the stream, shown as the 5/95th percentile
of all feasible solutions from pre- and post-deforestation calibration (green shaded area). The grey shaded area indicates the
deforestation period.



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1060 **Figure 3: Model structure used in this study. The light blue boxes indicate the hydrologically active individual storage volumes in the hillslope and riparian zones, respectively. The darker blue box $S_{S,p}$ indicates a hydrologically passive, i.e. $dS_{S,p}/dt = 0$, mixing volume. The blue lines indicate liquid water fluxes, the green lines indicate vapour fluxes. Model parameters are shown in red, adjacent to the model component they are associated with. All symbols are defined in Table 1.**

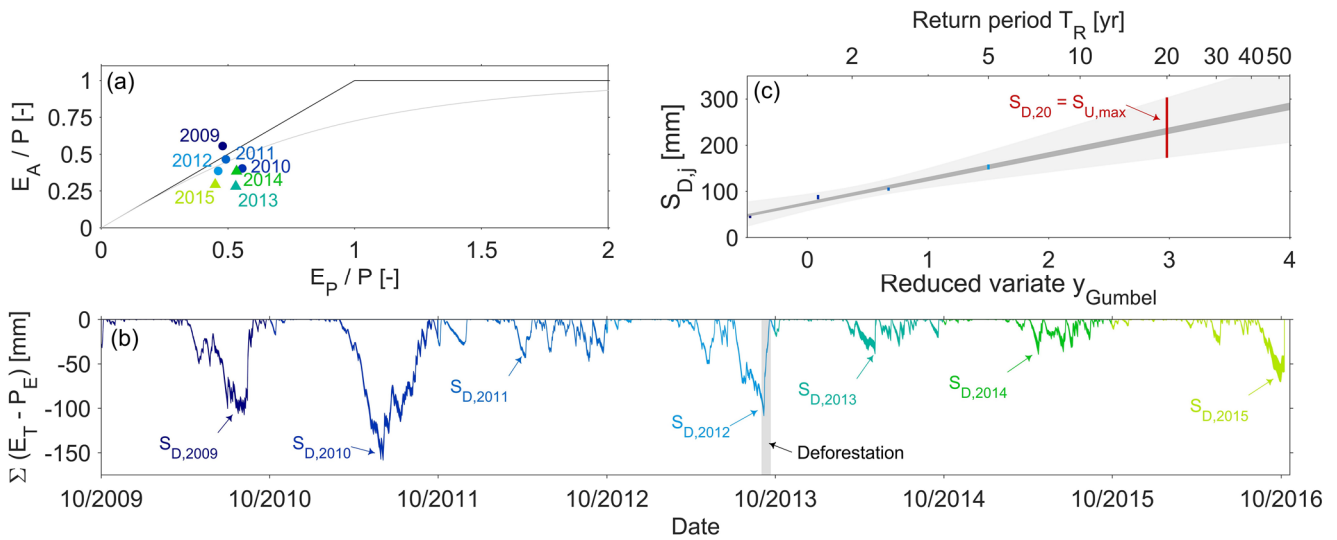




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Figure 4: (a) Positions of the individual years of the study period in the Budyko framework. The x-axis shows the aridity index $I_A = E_p/P$, the y-axis indicates the evaporative ration E_A/P and the runoff ratio $C_R = 1 - E_A/P$. Pre-deforestation years are shown with blueish shades, post-deforestation years with greenish shades. The bold black lines indicate the energy and water limits, respectively. The dashed grey line is the theoretical-analytical Turc-Mezentsev relationship (Turc, 1954; Mezentsev, 1955). (b) The range of time series of storage deficits as computed according to equation 2, using values of I_{max} from 0 to 4 mm. The maximum annual storage deficits $S_{D,j}$ are indicated by the arrows. The grey shaded area indicates the deforestation period. (c) Estimation of $S_{U,max}$ as the storage deficit associated with a 20-year return period $S_{D,20yr}$ using the Gumbel extreme value distribution for the pre-deforestation period. The blueish dots indicate the range of maximum annual storage deficits $S_{D,j}$ for the four years pre-deforestation period. The dark grey shaded area indicates the envelop of least-square fits for the individual values of I_{max} . The light grey shaded area indicates the envelope of the 5/95th confidence intervals. The red line shows the plausible range for $S_{U,max}$.

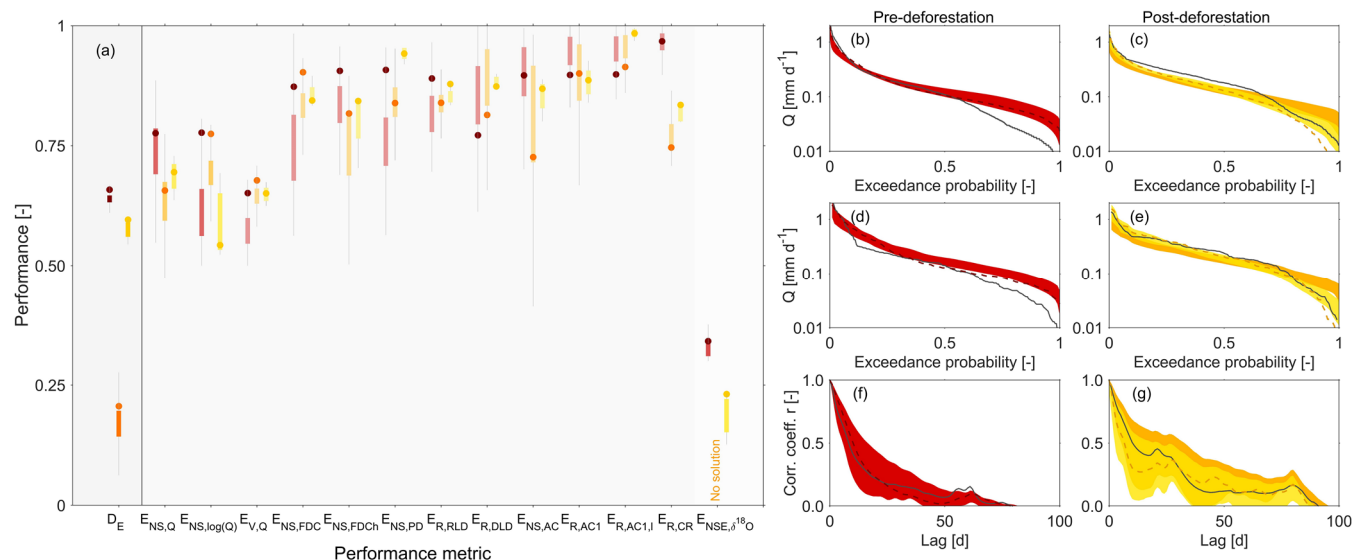


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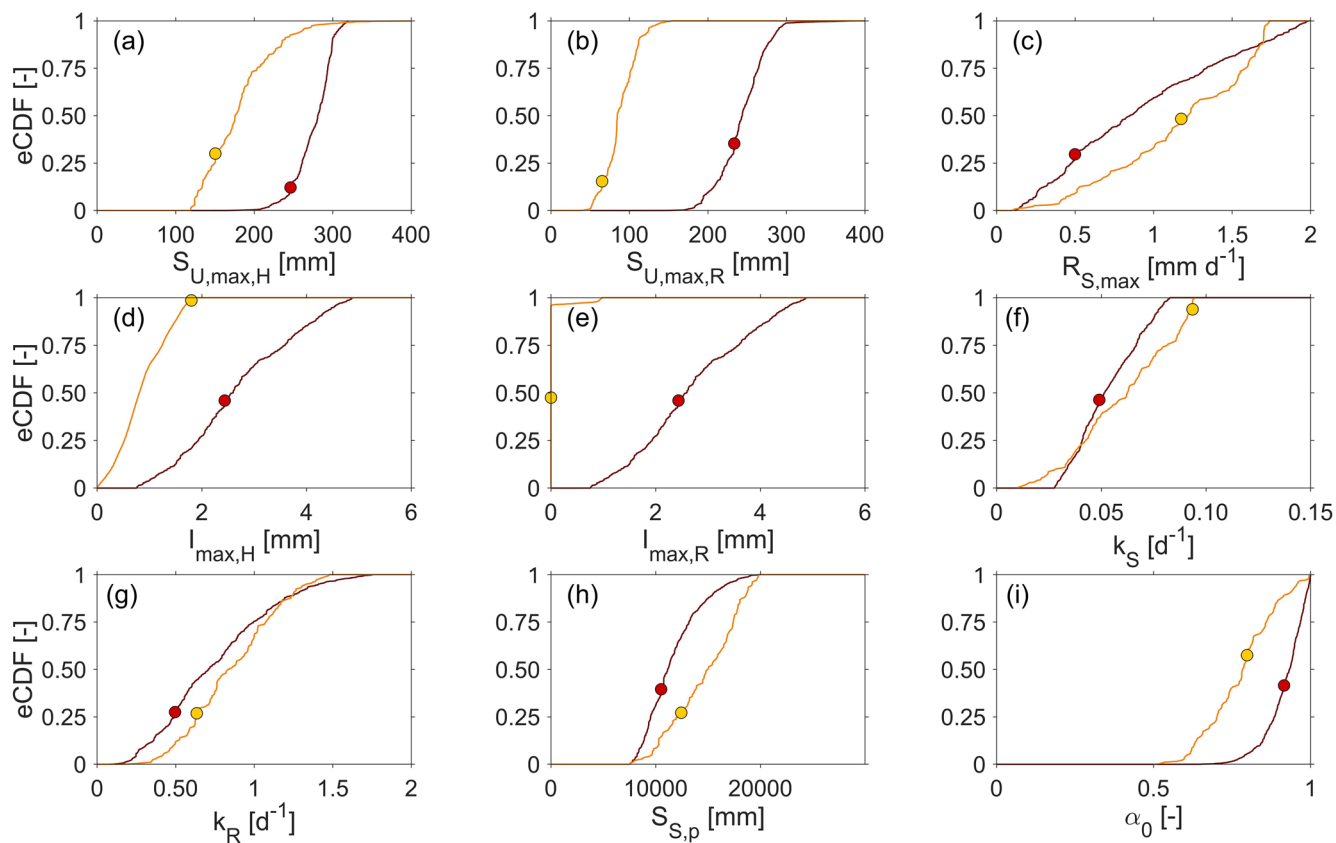
1105 **Figure 5: (a) Model performance metrics for all variables and signatures. D_E is the Euclidean distance to the perfect model. It**
combines all other performance metrics (Table 3) into one number (Eq.42). All performance metrics are formulated in a way that
a value of 1 indicates a perfect fit. The boxplots summarize the performances of all parameter sets retained as feasible. The circle
symbols indicate the performance of the best performing model in terms of D_E . The dark red shades indicate pre-deforestation
model performance based on calibration in the pre-deforestation period. Orange shades indicate post-deforestation performance
using the pre-deforestation parameter sets without further re-calibration. Yellow shades show the post-deforestation performance
after model re-calibration in the post-deforestation period. (b)-(c) show flow duration curves, (d)-(e) show the peak distributions and
(f)-(g) the autocorrelation functions for the pre- (red) and the post deforestation periods (orange and yellow), respectively. The
black lines indicate the observed values, the dashed lines indicate the best fits and the shaded areas the 5/95th uncertainty interval
of all solutions retained as feasible. The dark red shades indicate pre-deforestation model results based on calibration in the pre-
deforestation period. Orange shades indicate post-deforestation model results using the pre-deforestation parameter sets without
further re-calibration. Yellow shades show the post-deforestation model results after model re-calibration in the post-deforestation
period.



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Figure 6: Posterior distributions of selected parameters. Red shades indicate calibration in the pre-deforestation period, Yellow shades indicate post-deforestation calibration. The dots indicate the parameter values associated with respective best fit models.



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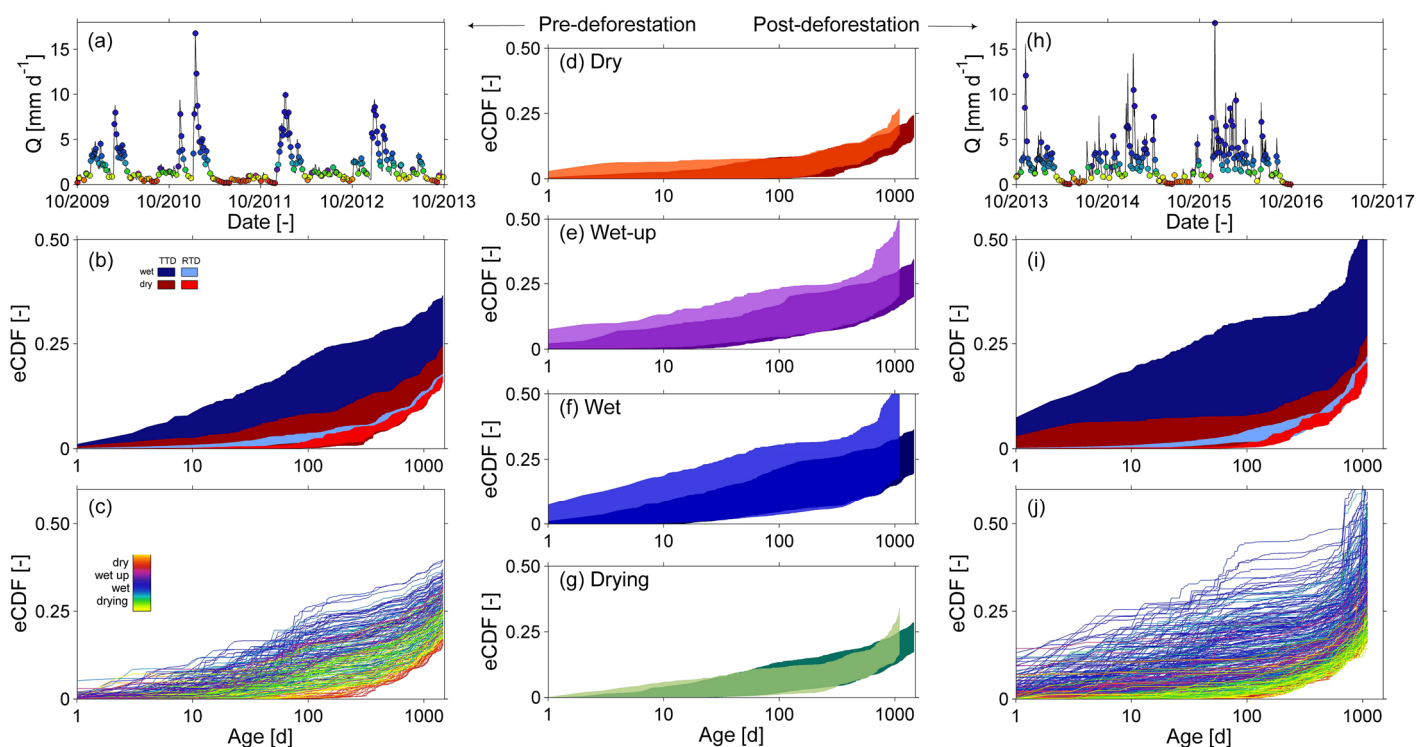


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Figure 7: Panels in the left column show pre-deforestation (a) discharge, the coloured dots indicate to which period (dry, wet-up, wet, drying) the individual selected time steps belong; (b) the 5/95th percentiles of the empirical cumulative RTDs (light shades) and TTDs (dark shades) for wet (blue) and dry (red) periods, respectively; (c) the ensemble of the individual TTDs at the time steps indicated in (a). Panels in the middle column (d-g) compare the 5/95th percentiles of empirical cumulative TTDs between pre-deforestation (dark shades) and post-deforestation (light shades) periods for dry, wet-up, wet and drying conditions, respectively. Panels in the right column show post-deforestation (h) discharge, the coloured dots indicate to which period (dry, wet-up, wet, drying) the individual selected time steps belong; (i) the 5/95th percentiles of the empirical cumulative RTDs (light shades) and TTDs (dark shades) for wet (blue) and dry (red) periods, respectively; (j) the ensemble of the individual TTDs at the time steps indicated in (h). All distributions shown are truncated at 3 (post-deforestation) for 4 years (pre-deforestation), which coincides with the tracked period. For the remaining fractions, i.e. the difference to 1, it can only be said that they are older than 3 years but nothing more than that.

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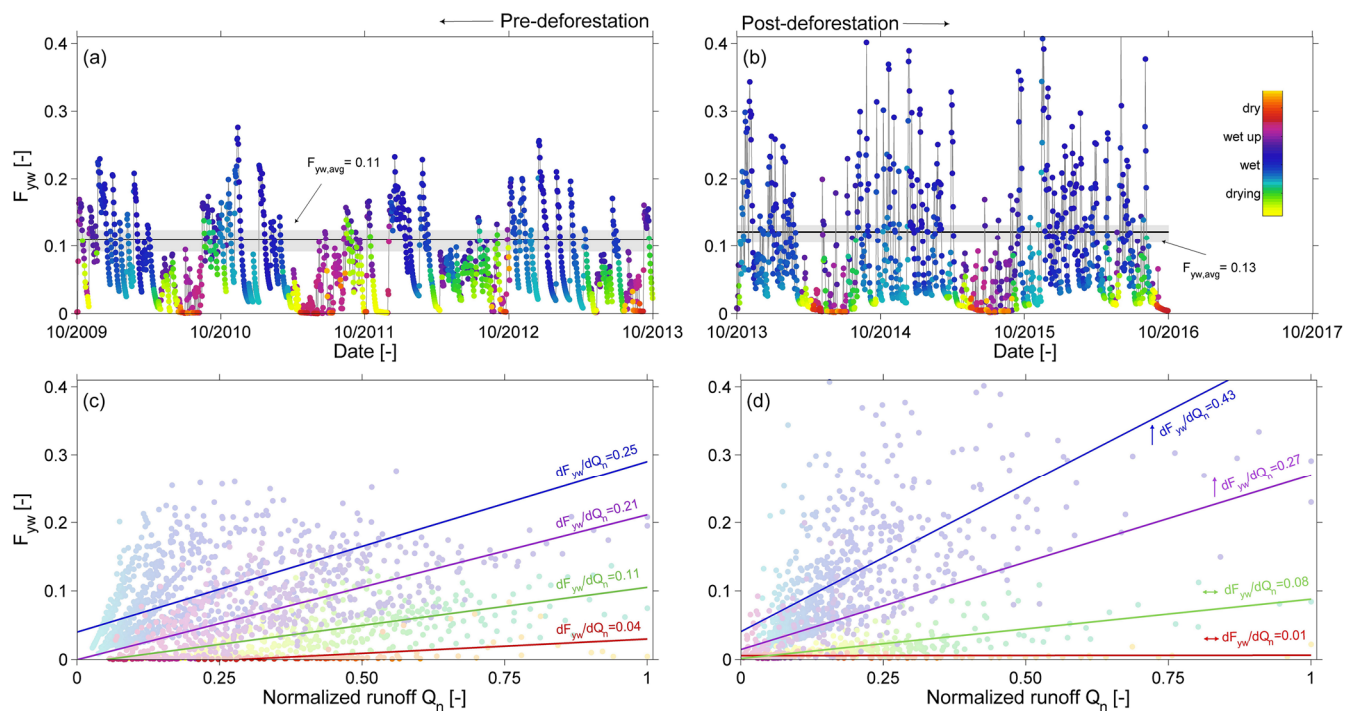
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Figure 8: (a)-(b) Pre- and post-deforestation time series of young water fractions F_{yw} in discharge. The colour code indicates the transition between dry, wetting-up, wet and drying conditions. The bold black line shows the mean F_{yw} of the best model fit, the grey shaded area shows the 5/95th percentile of F_{yw} for all feasible model solutions; (c)-(d) pre- and post-deforestation sensitivity of F_{yw} to discharge, using the same colour code as above to indicate dry, wetting-up, wet and drying conditions. The arrows in (d) indicate if there are statistically significant (\uparrow ; $p < 0.05$) changes or not (\leftrightarrow) in the sensitivities between the post-deforestation period and the pre-deforestation period.

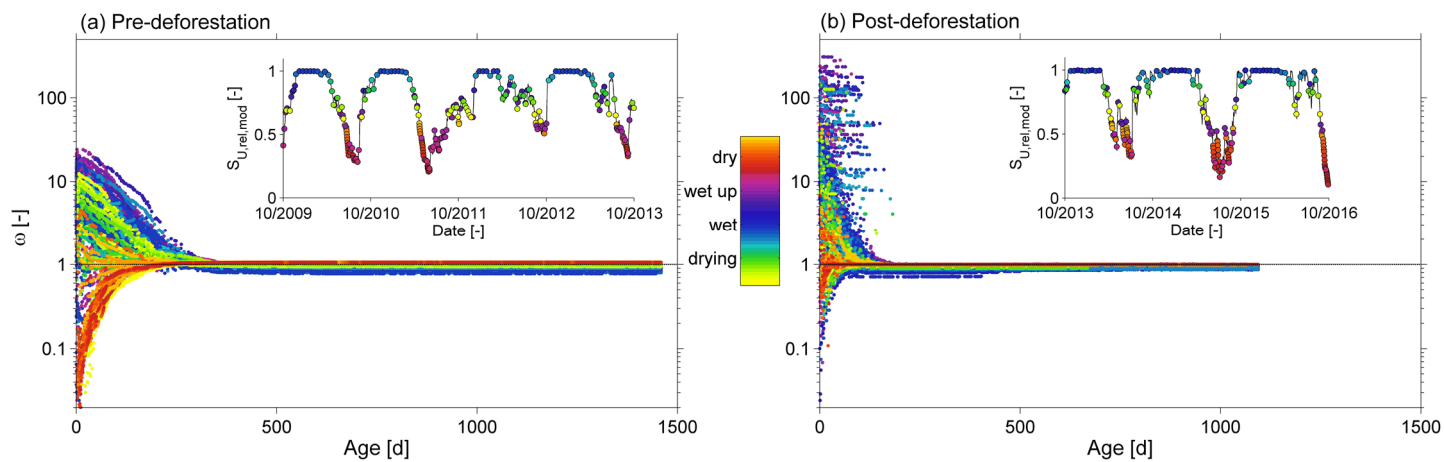


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1170 **Figure 9: Individual catchment overall SAS ω -functions for individual time steps under different wetness conditions in the (a) pre-deforestation period and (b) post-deforestation period. The insets show the relative water content in $S_{U,rel,mod} = S_U/S_{U,max}$ at the individual time steps.**



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