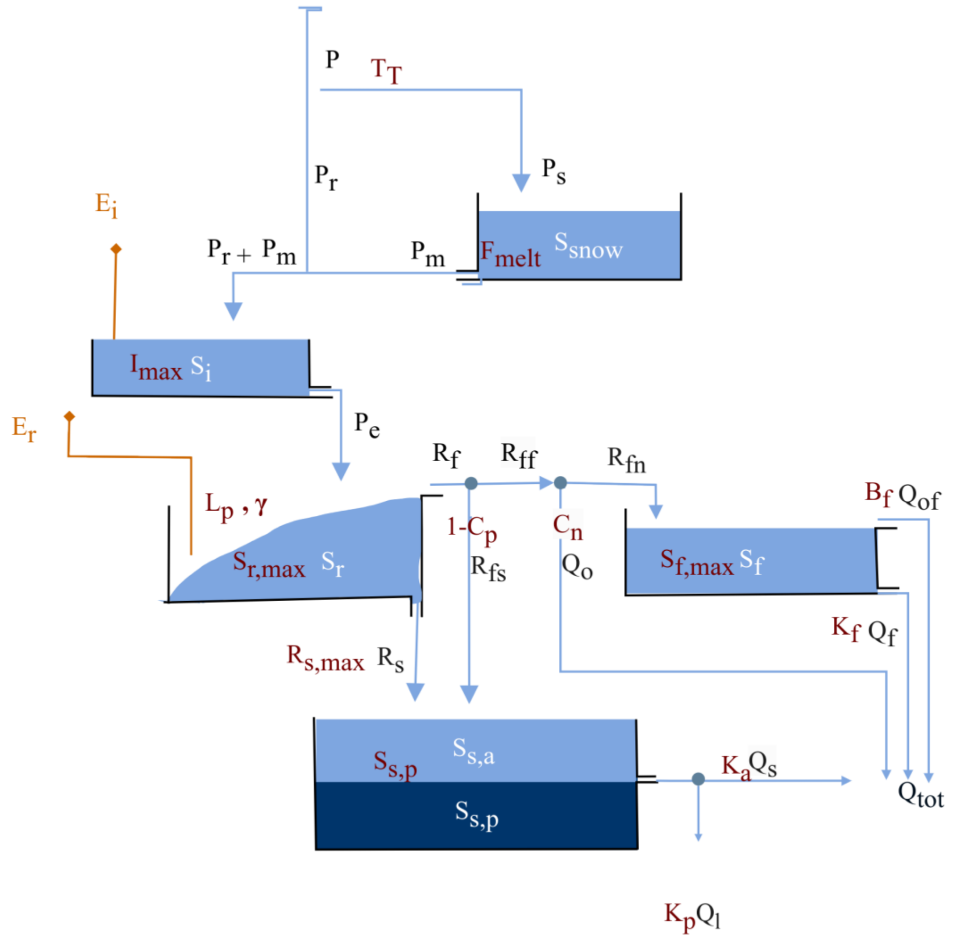

Figure S 1. The model structure used to represent the HOAL and the Wüstebach catchment. Light blue boxes indicate the hydrologically active individual storage volumes that contribute to total discharge Q_{tot}: Snow storage (Ssnow), canopy interception (S_i), fast response bucket (S_f), root zone (S_r), and "active" groundwater ($S_{s,a}$). The darker blue box($S_{s,a}$) indicates a hydrologically "passive" mixing groundwater volume. Blue lines indicate snow and water fluxes, while orange lines indicate water vapour fluxes. Model parameters are shown in red adjacent to the model component they are associated with. All symbols are defined in Table S 1

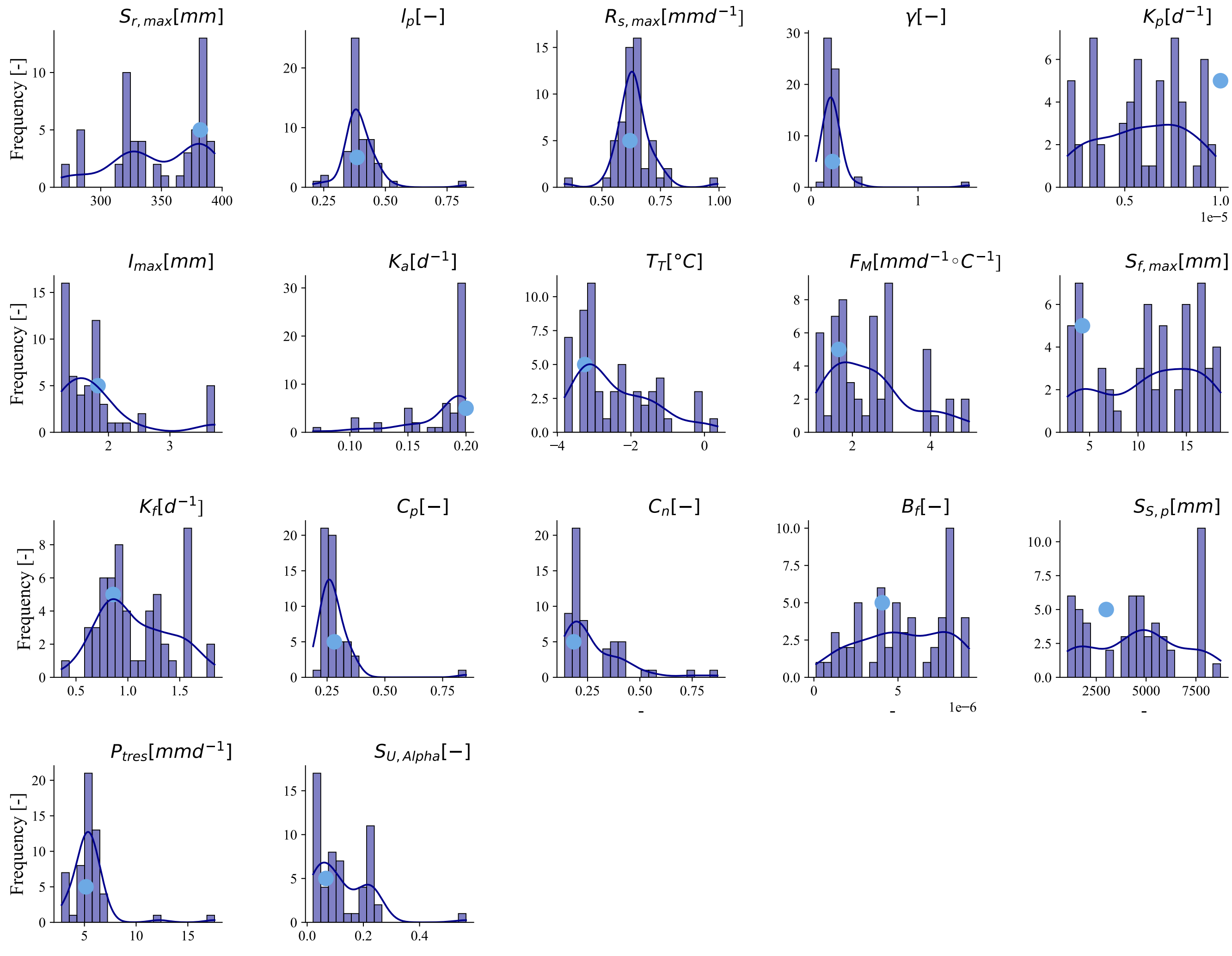

Figure S 2. Pareto optimal distributions of selected 55 parameters for the HOAL catchment. The model calibration results are shown as a frequency histogram. The dots indicate the parameter values associated with the best-balanced solution with the lowest DE.

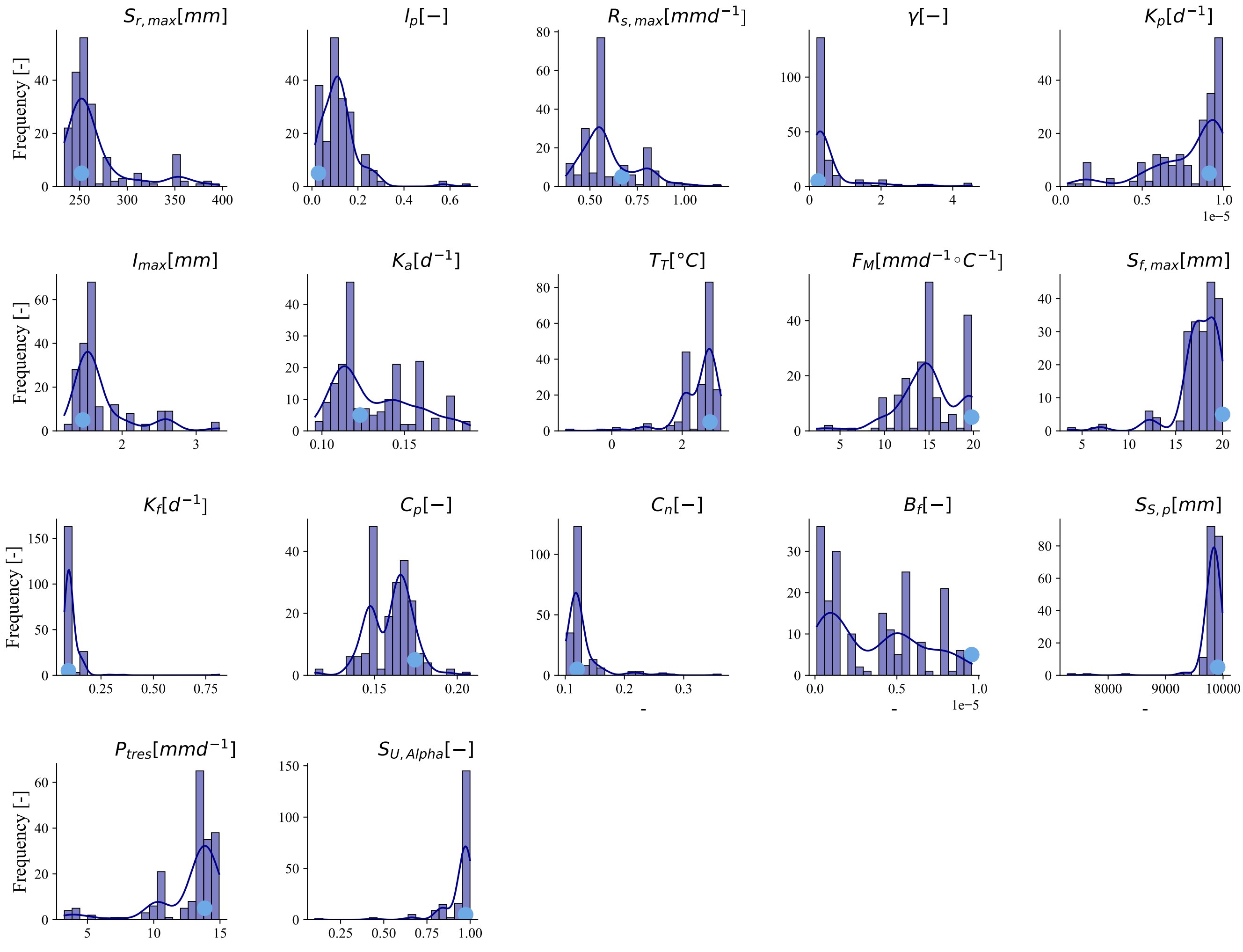

Figure S 3. Pareto optimal distributions of selected 190 parameters for the Wüstebach catchment. The model calibration results are shown as a frequency histogram. The dots indicate the parameter values associated with the best-balanced solution with the lowest DE.

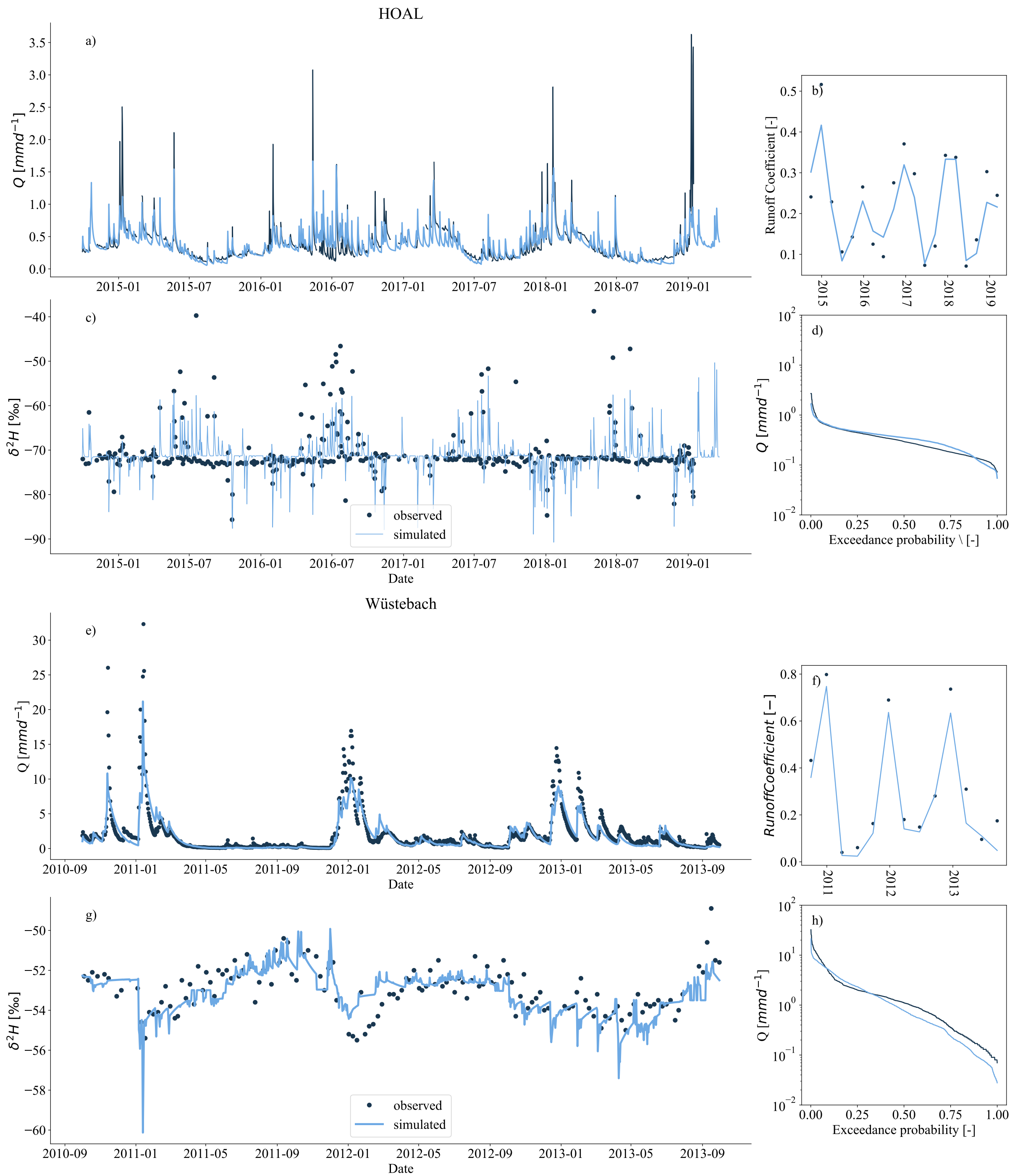

Figure S 4. Modelling results for streamflow, the signature of streamflow, and δ^2H ratios in the stream for HOAL (a-d) and Wüstebach (e-h). Observed values are presented as black lines and dots, while simulations based on the best optimal solution are shown as blue lines for all panels. (a, e) observed and modelled daily streamflow Q (mm-1). (b, f) observed and modelled a three-month averaged runoff coefficient (RC). (c, g) observed and modelled δ^2H signals in streamflow. (d, h) Observed and modelled flow duration curves for HOAL and Wüstebach, respectively.

Table S 1. Definitions and uniform prior distributions of the parameters of the solute-transport model (Fig. S 1).

Parameter	Unit	Definition	Lower, Upper Bound
Hydrological			
T_r	(°C)	Threshold temperature for snow melt	[-4.0, 5.0]
γ	(–)	Shape factor	[0.0, 5.0]
B_f	(–)	Saturation excess overland flow coefficient	[0.0, 0.00001]
C_n	(–)	Division parameter for fraction of overland flow	[0.0, 1.0]
C_p	(–)	Division parameter for fast groundwater recharge	[0.0, 1.0]
F_{melt}	(mm d^{-1} $°C^{-1}$)	Melt factor	[1.0, 5.0]
I_{max}	(mm)	Interception capacity	[1.2, 5.0]
K_a	(d^{-1})	Storage coefficient of the slow-responding reservoir	[0.01, 1.2]
K_f	(d^{-1})	Storage coefficient of the fast-responding reservoir	[0.01, 2.0]
K_p	(d^{-1})	Storage coefficient of deep infiltration losses	[0.0, 0.00001]
L_p	(–)	Transpiration water stress factor	[0.0, 1.0]
P_{thresh}	(mm d^{-1})	Threshold precipitation for overland flow	[2.0, 20.0]
R_{max}	(mm d^{-1})	Maximum percolation rate	[0.0, 1.2]
S_{fmax}	(mm)	Fast response storage capacity	[0.0, 20.0]
S_{rmax}	(mm)	Root-zone storage capacity	[100, 500]
Tracer			
S_{sp}	(mm)	Passive storage capacity	[1000, 10000]
$S_{U,\text{Alpha}}$	(–)	SAS alpha shape parameter for root zone	[0.0, 1.0]

Table S 2. Signatures for streamflow, δ^2Hsignal and the associated performance metrics used for model calibration scenarios and efficiency evaluation. The performance metrics included the Nash-Sutcliffe efficiencies of streamflow (NSE_Q), of logarithmic streamflow (NSE_{logQ}), the flow duration curve (NSE_{FDC}),the runoff coefficient averaged over three months (NSE_{RC}) as well as the NSE of the δ^2H signal in streamflow $NSE_{\delta^2\text{H}}$

Signatures	Abbreviation	Performance Metric	Reference
Time series of streamflow		NSE_Q	(Nash and Sutcliffe, 1970)
	Q	$\text{NSE}_{\text{log}Q}$	
Flow duration curve	FDC	NSE_{FDC}	(Jothityangkoon et al., 2001)
Seasonal runoff ratio	RC	NSE_{RC}	(Yadav et al., 2007)
Time series δ^2H in streamflow	δ^2H	$\text{NSE}_{\delta^2\text{H}}$	(Birkel et al., 2011b)

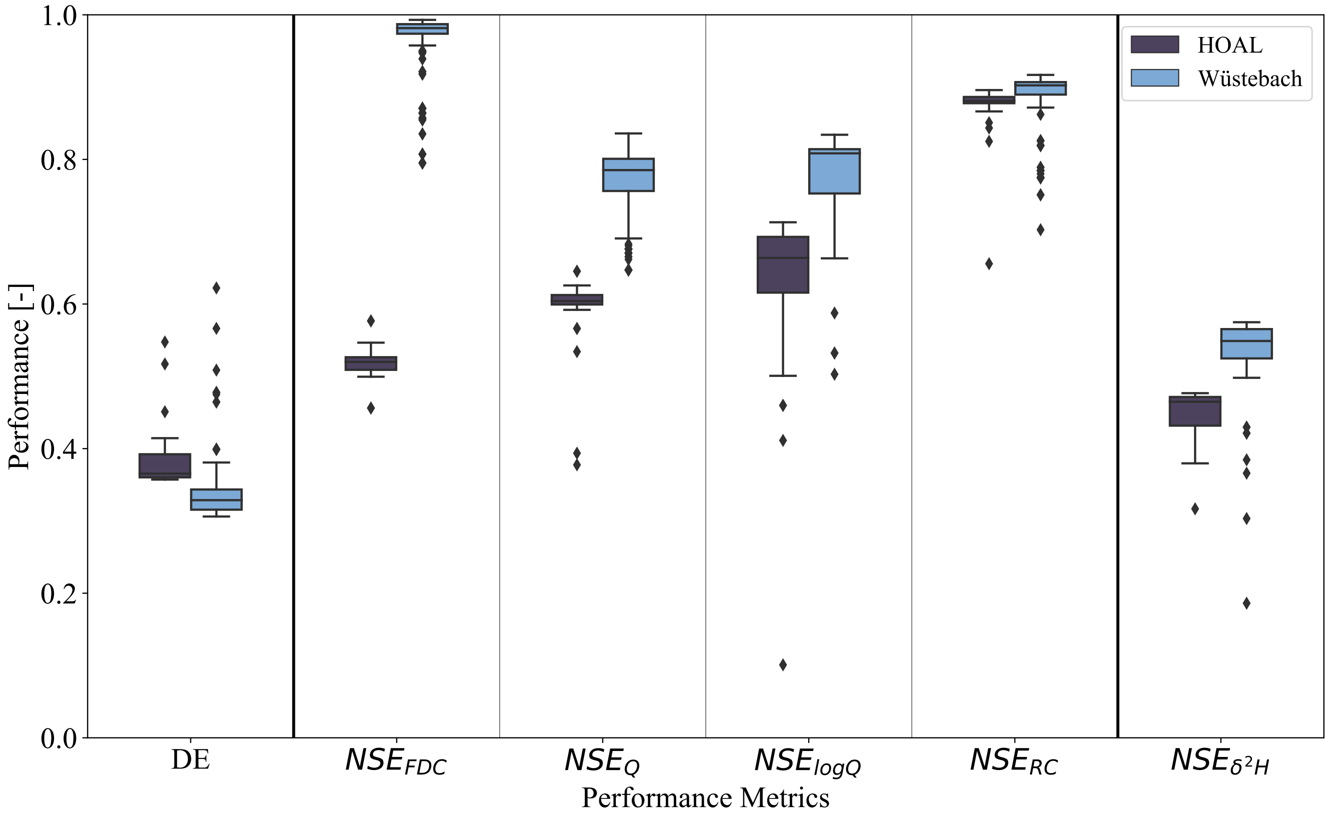

Figure S 5. Boxplots of performance metrics of the 190 best-performing parameter sets. The performance metrics included the Nash-Sutcliffe efficiencies of the flow duration curve (NSE_{FDC}), streamflow (NSE_Q), of logarithmic streamflow (NSE_{logQ}), the runoff coefficient averaged over three months (NSE_{RC}) as well as the NSE of the δ^2H signal in streamflow $NSE_{\delta^2\mathrm{H}}$

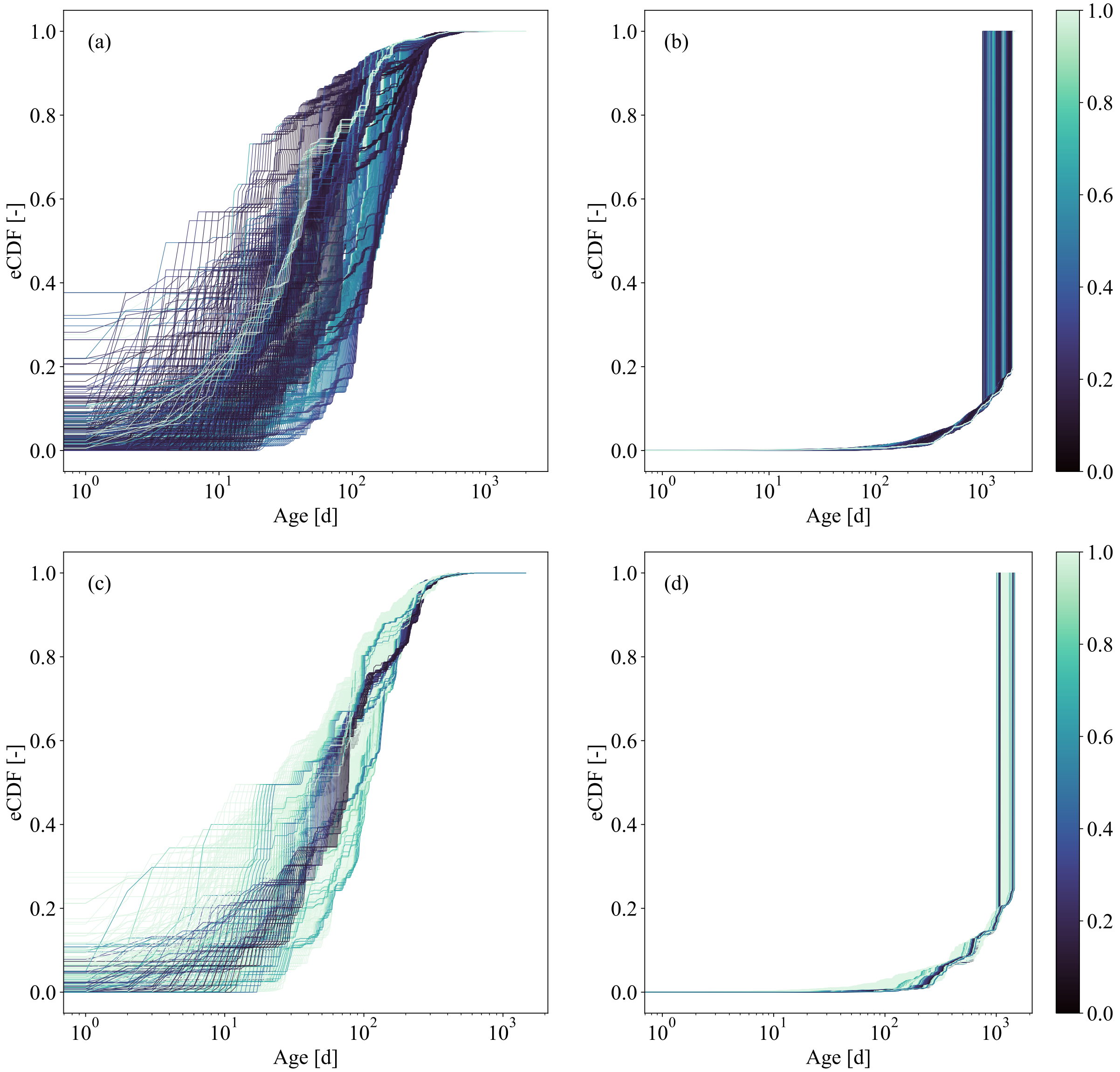

Figure S 6. Residence time distribution of the root zone (a, c) and groundwater (b, d) for the HOAL (top) and Wüstebach (bottom) catchments, respectively. The color of the lines corresponds to normalized streamflow, where light green indicates high flow and dark blue indicates low flow.

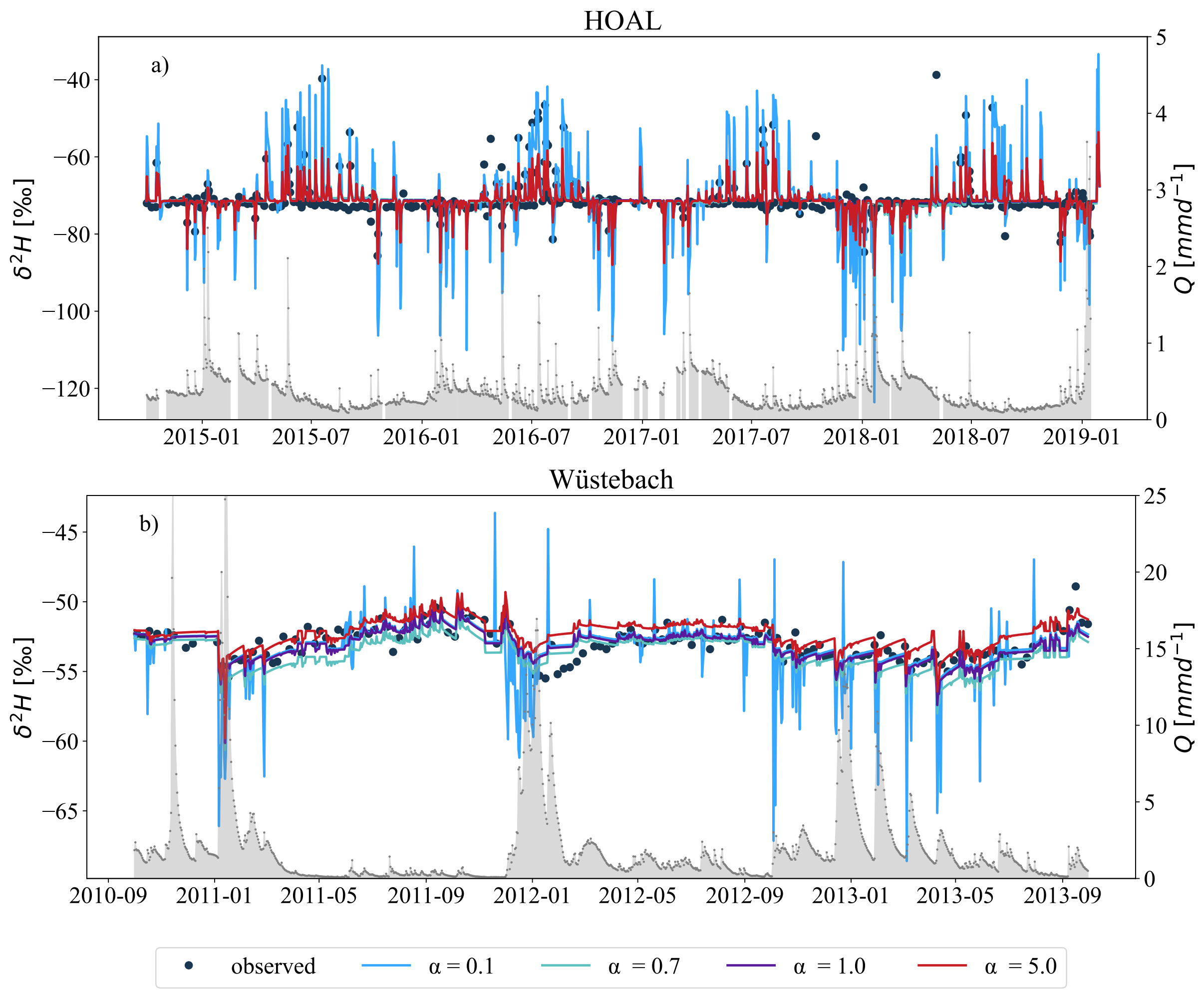

Figure S 7. Simulation of δ^2H in streamflow based on varying SAS shape parameter α [-] in groundwater for (a) HOAL and (b) Wüstebach. The simulations ranged from very young water preference (α = 0.1) to old water preference (α = 5) for the groundwater, while for the root zone compartment, a calibrated value was used (α = 0.14, for HOAL and 0.98 for Wüstebach). Line colours correspond to the simulation based on α values: blue for 0.1, turquoise for 0.7, purple for 1.0, and red for 5.0. The grey-shaded area shows the measured streamflow (Q, mm d^{-1}) for both catchments.

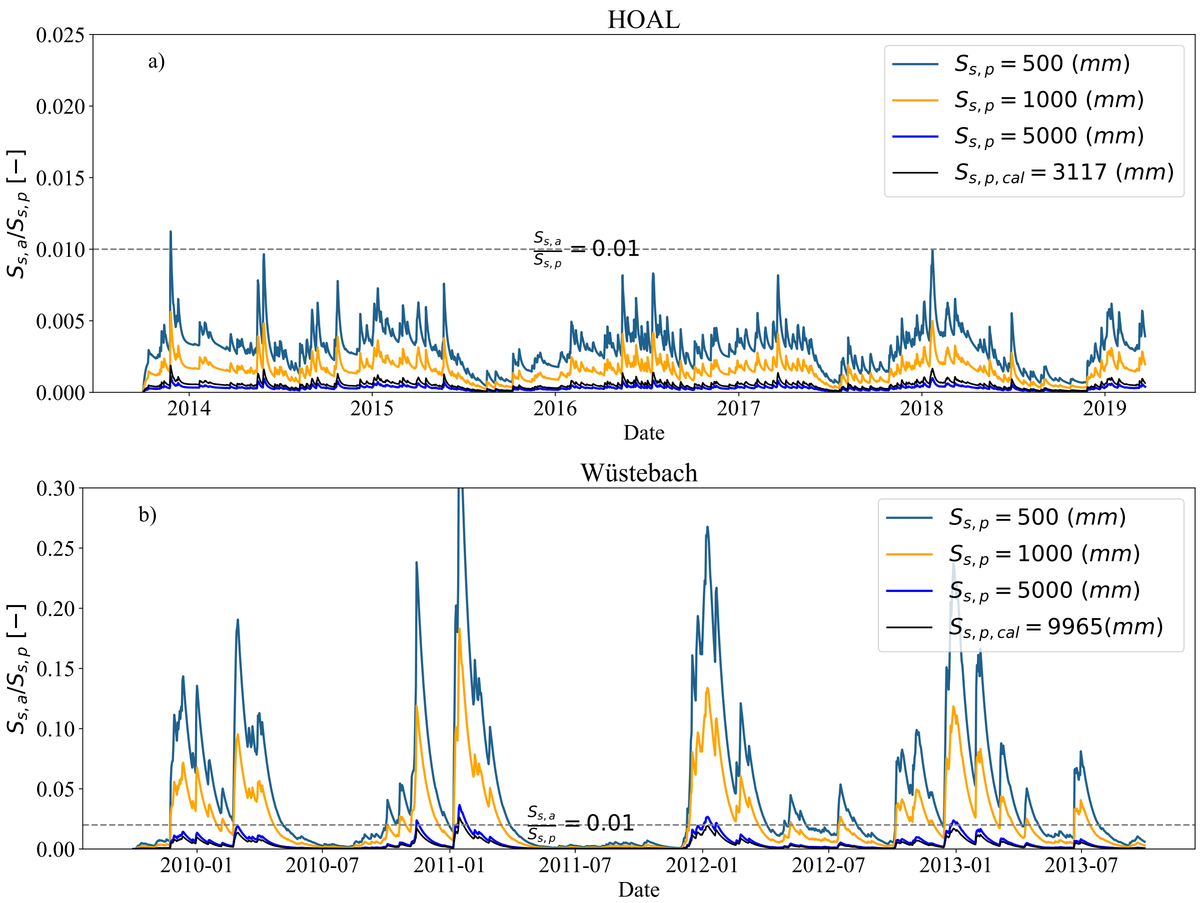

Figure S 8. Time series of the ratio of active to passive groundwater storage volumes $(S_{s,a}/S_{s,p})$ for (a) the HOAL catchment and (b) the Wüstebach catchment under varying passive storage volumes ($S_{s,p}$). Lines represent different scenarios of passive storage volumes: 500 mm (blue), 1000 mm (orange), 5000 mm (dark blue), and the calibrated passive storage volume ($S_{s,p,\text{cal}}$) of 3117 mm for HOAL and 9965 mm for Wüstebach (black). The dashed line indicates $S_{s,a}/S_{s,p} = 0.01$, representing a threshold for significant active storage contribution. The results highlight the impact of the passive storage volume on the temporal dynamics of groundwater storage partitioning in both catchments.

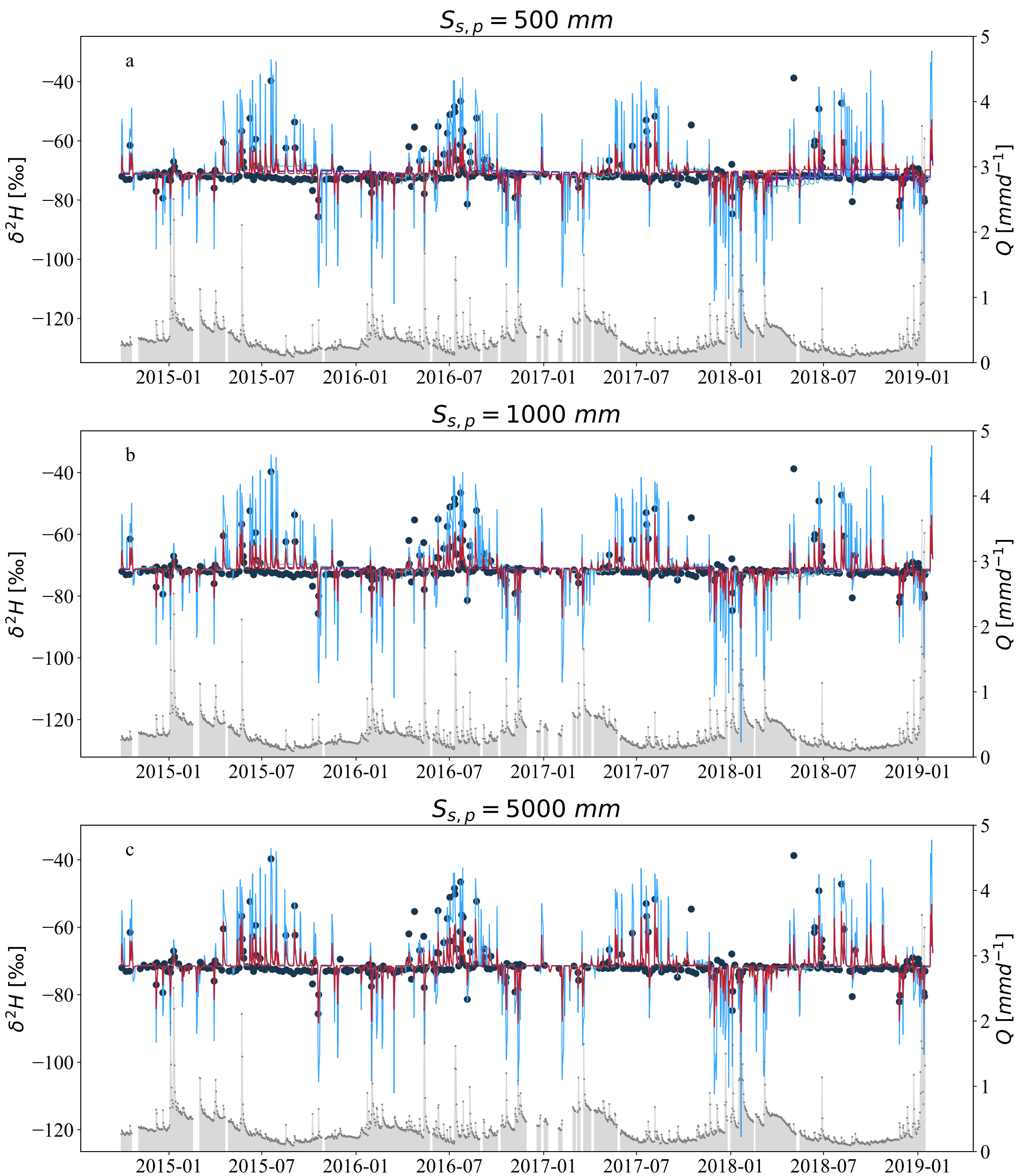

Figure S 9. The simulation results for δ^2H signals from streamflow (Q mm d^{-1}) based on varying passive groundwater storage volumes ($S_{S,p}$ = 50 mm, $S_{S,p}$ = 500 mm, 1000 mm, and 5000 mm) and different mixing assumptions determined by SAS function shape factors (α = 0.1, 0.7, 1.0, and 5.0) for the HOAL catchment (a-c). (a-c), black dots represent grab samples of δ^2H from streamflow. Line colours correspond to the simulation based on α values: blue for 0.1, turquoise for 0.7, purple for 1.0, and red for 5.0.

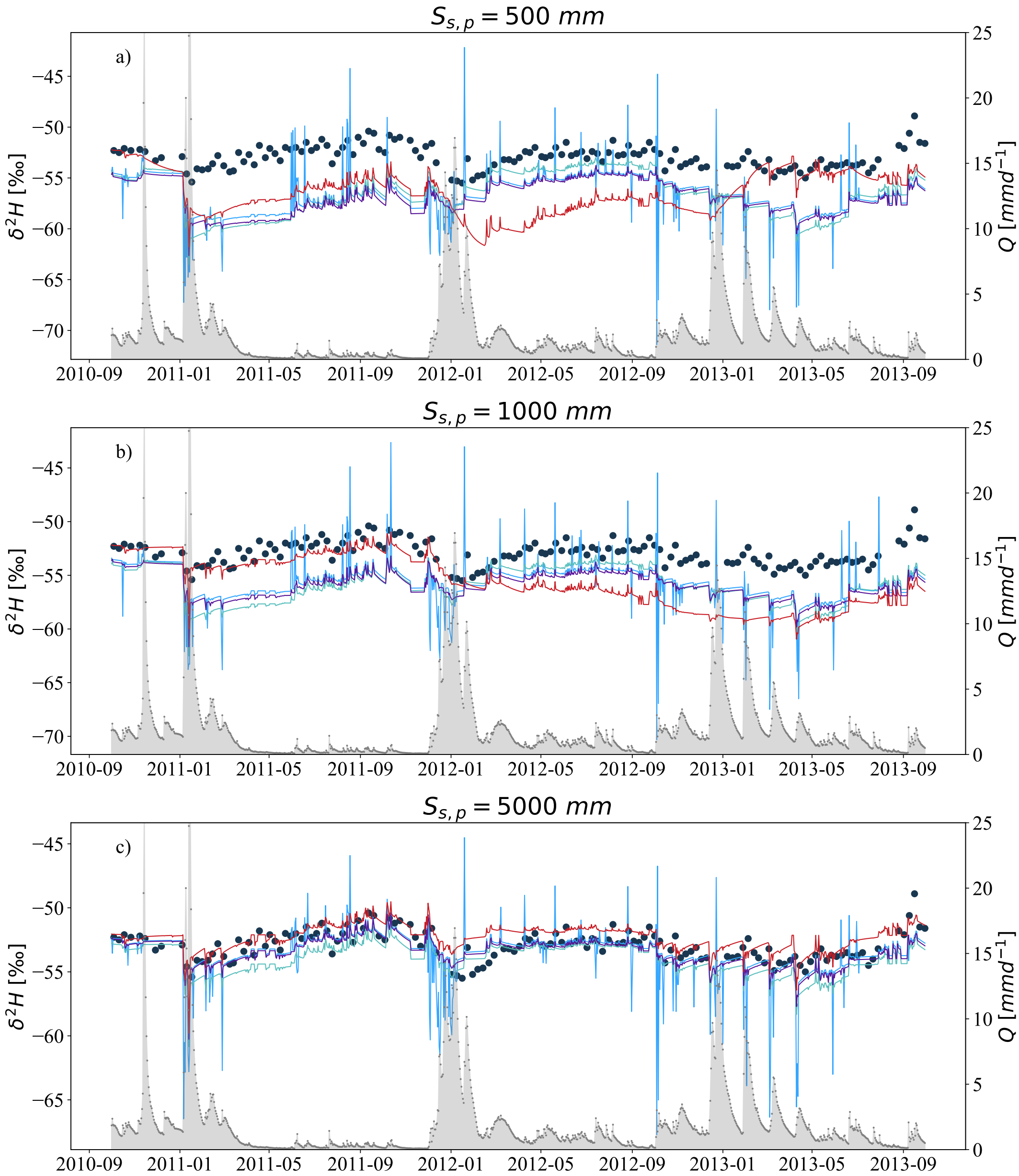

Figure S 10. The simulation results for δ^2H signals from streamflow (Q mm d^{-1}) based on varying passive groundwater storage volumes ($S_{S,p}$ = 500 mm, 1000 mm, and 5000 mm) and different mixing assumptions determined by SAS function shape factors (α = 0.1, 0.7, 1.0, and 5.0) for the Wüstebach catchment (e-f). (e-f) black dots represent weekly grab samples of δ^2H from streamflow. Line colours correspond to the simulation based on α values: blue for 0.1, turquoise for 0.7, purple for 1.0, and red for 5.0.