Editor Comment:

Thank you very much for your revised manuscript. While reviewer 1 recommends publication you can see that reviewer 2 provided still a rather long list of recommendations, mainly to make your work transparent. I sincerely ask you to address these points thoroughly in a of revisions.

Authors' General Comment:

Dear Prof. Erwin Zehe,

We thank you for your time and for your assessment of our revised manuscript (EGUSPHERE-2025-2597). We are thankful that Reviewer 1 recommends the manuscript for publication. We also appreciate the thorough and detailed feedback provided by Reviewer 2, which has helped us to further improve the transparency and clarity of our work. We addressed all comments and recommendations raised by both reviewers.

Yours sincerely,

Hatice Türk and co-authors

Point-by-Point Authors' Responses to the Editor and Reviewers Comments.

In the following, we provide our detailed **point-by-point** responses to each of the comments. Reviewer comments are shown in **black**, while our responses are shown in **blue**. Line numbers refer to the revised version with track-changes.

Reviewer #1

The authors did an excellent job responding to this annoying reviewer. Indeed, the bucket model is inherently non-mechanistic, and any effort to expand its capacity to include unresolved aspects, such as preferential flows, is commendable. My requests aimed to encourage the authors to use this stage and the journal's discussion-oriented platform at HESS to promote dialogue on transitioning toward a more mechanistic approach. This feature distinguishes HESS from other journals and can serve as a collaborative space for sharing speculative hypotheses with the community and debating the experiments and analyses needed to test them. With that in mind, I reiterate my request to include the PDF in the supplementary materials. I understand that the eCDF is "cleaner" for figures 3 and 5, but the PDF offers additional information on infiltration and flow that could support ongoing discussions about moving toward mechanistic methods. Aside from that, the revisions create a standalone paper that broadens the field's boundaries and highlights the necessary steps forward. Thank you for that.

Response: We thank the reviewer for the positive feedback. To further support the discussion of infiltration and flow processes, we have included the PDF of the estimated transit time distributions for streamflow in the Supplementary Material (Figure S7).

Reviewer #2

The changes made by the authors have clearly improved the paper's quality, and I appreciate the effort, particularly in clarifying the theoretical background on the modelling.
However, several areas still need refinement for optimal clarity and precision in the overall narrative. This is particularly unfortunate, as many relate to points raised in my previous review, such as vague and imprecise language leading to inaccurate hydrological statements. I recommend consistently using the same terminology. For example, always referring to the SAS "framework" or "function" uniformly and correctly distinguishing between "groundwater" and "groundwater storage."

Response: We thank the reviewer for the re-evaluation of the manuscript and for acknowledging the improvements made in the revised version. We appreciate the feedback and the time invested in providing detailed comments. We have undertaken a thorough revision of the manuscript to improve the consistency and precision of terminology throughout.

My major concerns at this stage are as follows:
• Persistent vague and imprecise language throughout the manuscript. I recommend a thorough language review to enhance readability.

Response: We thank the reviewer for highlighting this concern. We conducted a thorough language revision of the manuscript to improve clarity, precision, and readability, with particular attention to eliminating vague or imprecise phrasing and ensuring consistent terminology throughout.

• Unclear calibration procedure: Was the bucket model calibrated first against streamflow, followed by calibration of the transport model against stable water isotopes? I recommend explicitly explaining these steps.

Response: The bucket model was calibrated simultaneously against both streamflow and stable water isotopes measurements in streamflow. The calibration was conducted using a function that aggregates the individual performance metrics for streamflow and the δ^2H into the Euclidean distance DE, with equal weights assigned to streamflow and the δ^2H signature. To further clarify this, we added a sentence in the manuscript to explicitly emphasize that calibration was performed concurrently for both streamflow and the δ^2H, rather than sequentially. The sentence reads as follows:

(Lines 272 - 274)

"We calibrated the model simultaneously against both streamflow and stable water isotopes to ensure that both hydrological and tracer information were integrated during parameter optimisation."

In addition, we clarified in the model definitions (lines 169–171) that the model simulates streamflow and tracer fluxes simultaneously as: "which allows for the simultaneous representation of water fluxes and tracer transport (Hrachowitz et al., 2013) and follows the concept of storage-age selection functions (Rinaldo et al.,2015)."

• Some results sections remain difficult to understand, making it challenging to grasp the main messages. I recommend streamlining these for better flow (see points below).

Response: we have streamlined the results sections to improve flow and clarity, directly addressing the specific points listed below to ensure that the main messages are clear

• I appreciate the addition of a discussion section titled "Implications." However, the section does not contain any actual implications for me. To improve this aspect, I recommend discussing why and how the results matter for water quality, how the approach could apply to other catchments beyond the discussed uncertainties (e.g., whether the a priori assumptions here need adjustment per catchment), and in which catchments (especially regarding active/passive storage dynamics) similar or distinct outcomes might be expected.

Response: We thank the reviewer for this comment. We revised the "Implications and limitations" section to explicitly discuss the relevance of the modelling approach for water-quality interpretation, including the role of preferential flow in mobilizing solutes during event-driven conditions, the transferability of the framework to other catchments, and how differences in active–passive groundwater storage dynamics may lead to similar or contrasting outcomes across hydrological settings. Lines 570 -574 now read as:

"At the catchment scale, isotope-based modelling proved useful in capturing preferential flow in the unsaturated zone, but was limited in doing so in groundwater due to the damping of water stable isotope signals by large passive groundwater storage volumes. This damping does not necessarily indicate the absence of preferential

flow; rather, it implies that when isotope variability in streamflow is strongly attenuated, groundwater age mixing will be difficult to constrain using isotopes alone. This limitation is relevant for water-quality applications, as catchments with large passive groundwater storage may still exhibit rapid contaminant responses during hydrological events through preferential flow pathways, despite long mean transit times. Consequently, applying this modelling approach to other catchments requires careful consideration of active–passive groundwater storage dynamics."

I have also raised several minor points which I believe are relevant to improving the manuscript. Some of these concern language clarity and style, where the final decisions are left to the authors.

Response: We thank the reviewer for these additional comments. We have considered all minor points raised and revised the manuscript accordingly to improve language clarity and style

• **Line 1**: "Preferential flow paths in hydrological systems (e.g., macropores or subsurface pipe networks)". Macropores/pipe networks are not systems. Please clarify.

Response: We revised sentence as "Preferential flow paths (e.g., macropores or subsurface pipe networks) in hydrological systems"

• Line 4: Specify "unsaturated zone and groundwater storage" or "groundwater aquifer"? Current wording is incorrect.

Response: We revised the sentence as "unsaturated zone and groundwater aquifers"

• Line 6: Use "hypothesize" (present tense).

Response: We updated the tense in the sentence

• Line 7: Clarify "this effect". Replace "represented through" with "studied with".

Response: We replaced "represented through" with "studied with" and removed "effect" from the sentence for clarity.

• Line 8: Please use "describe" instead of "specify".

Response: We used to "describe" instead of "specify".

• Line 9: "function for the unsaturated zone…": Vague. Consider "functions describing how

flow paths from unsaturated/groundwater…".

Response: We revised the sentence as "functions describing how water of different ages is released from the unsaturated zone and groundwater aquifer'

• Line 14: Delete "where r values…." (no novelty); "corresponding" unclear.

Response: We revised the sentence and removed the sentences with r values and"corresponding"

• Line 18: Consider deleting "with r…." (not essential for abstract).

Response: We deleted "with r…." from the abstract.

• Line 18: Please introduce "passive groundwater storage" concept earlier.

Response: We were unable to move this description earlier in the manuscript without disrupting the flow of abstract; however, we have added a brief clarification here defining passive groundwater storage as water that mixes with the tracer signal of the active groundwater volume".

• Line 20 ff.: Main message unclear; please revise for focus.

Response: We revised the sentence as "Consistent with this interpretation, the degree of attenuation in the simulated streamflow isotope signal increased with increasing passive groundwater storage volumes and became pronounced when passive storage was an order of magnitude larger than active groundwater storage.

• Line 24: Delete "actually".

Response: We deleted "actually" from the sentence

• Line 28: "Processes" misused. Likely means preferential flow paths.

Response: We replaced the "Processes with "preferential flow"

• Line 37 ff.: Unclear key message; consider deleting.

Response: We deleted "which have the potential to alter stream chemical composition: from the sentence to avoid repetition.

• Line 43: "Simpler": Compared to what? Maybe delete?

Response: We deleted "Simpler"

• Line 44: Please clarify "top-down". "Groundwater flow" means streamflow contribution?

Response: We replaced "top-down" with "conceptual" for clarity and revised the sentence as "it remains uncertain whether conceptual catchment-scale isotope-based transport models can meaningfully represent preferential groundwater flow contributions to streamflow."

• Line 46: Reposition "(transit time, TT)". Delete "statistical". Revise sentence and link to next.

Response: We replaced the "(transit time, TT)", deleted statistical" from the sentence and revised the sentence. New sentence now read as lines 43-46:"Water molecules entering at different locations within a catchment travel along distinct flow paths and take different times (transit time, TT) to exit the catchment via streamflow or evaporation. The distribution of transit times is referred to as the transit time distribution (TTD), which reflects key information about how quickly water moves through a control volume, such as a catchment"

• Line 49: Water flows above, not through, surface. Clarify "their".

Response: We updated the sentence as “from the surface, through the subsurface” we replaced “their” by “TTs to clarify that it refers to “TTs” in the sentence.

• Line 51: Replace "catchment wide input output signals of tracers" with "measured tracer signal". Start new paragraph before "Many studies…".

Response: We replaced the "catchment wide input output signals of tracers" with "measured tracer signal in streamflow". And we started a new paragraph before "Many studies…"

• Line 55: "Its long-term storage": Vague.

Response: We replaced the word with “its prolonged retention”

• Line 57 ff.: Why? Meaning unclear. Please add reference.

Response: We added a reference to lines 55-57 support the statement. In addition we added “to simulate both, transport and flow simultaneously” to provide reasoning

• Line 59: SAS function definition incorrect due to vagueness.

Response: We revised the definition of SAS function for further clarification. New sentence lines 58-60 read as “The SAS function represents water age dynamics of storage and release in hydrological systems by defining the relationship between the distribution of water ages stored within the system at a given time (residence time distribution, RTD) and the distribution of water ages leaving the system as outflows (TTD)”

• Line 63: "Vary over time" restates time-variable SAS.

Response: We removed “vary over time” to avoid repetition

• Lines 71–82: Unclear key message and "chosen mixing assumptions". Please specify.

Response: We specified “chosen mixing assumptions” as “mixing assumptions (e.g., complete mixing vs. partial mixing) in SAS function”. The new lines 77- 79 sentence now reads: “Several studies highlight that TTD estimates are sensitive to mixing assumptions (e.g., complete mixing vs. partial mixing) which are reflected in the shape of the SAS function and lead to uncertainty in transport timescale estimates.”

• Line 83: Delete "indeed".

Response: We deleted “indeed”

• Line 92: Please delete or explain “the conceptualization of Zuber (1986)”. Currently unclear.

Response: We removed “the conceptualization of Zuber (1986)” from the sentence.

• Line 93: Use "can be" instead of "is typically".

Response: We used “can be” instead of “typically”

• Line 98: Please clarify SAS assumptions earlier in introduction.

Response: We clarified SAS assumptions earlier in introduction in line 78.

• Line 99: Please consider using "Applying" instead of "However, adopting…".

Response: We used to “Applying” instead of "However, adopting…"

• Line 100: Beven citation is the wrong reference here. Please rephrase. The word

"Consequently,…." Is not connecting a logical consequence here. Please clarify.

Response: We removed the "Consequently,…." and added relevant citations to the sentence.

• Lines 105–112: Vague, especially last sentence.

Response: We revised the paragraph to explicitly state the tested hypotheses, the modelling choices evaluated, and the diagnostic role of isotope data, thereby clarifying the study objectives and reducing vagueness.

• Line 113: Introduce "shape" earlier. "process(es)" misused.

Response: We replaced “shape” with parameterizations for consistency and replaced “processes” with “preferential groundwater flow”

• Line 116: Please use "younger" (not "young"). Please use "described by" (not "through"). Applies manuscript wide.

Response: We used to younger and replace through by described by in several part of the manuscript

• Line 117: "Catchment-scale" is incorrect here (because this sentence refers to streamflow-linked SAS?). Please check third research question for clarity.

Response: We removed the “Catchment-scale” from the sentence. We revised the third research question as “How and to what extent do different groundwater mixing assumptions, in combination with varying passive storage volumes, affect the fit measured streamflow tracer signals, and the estimation of transit time distributions??”

• Line 119: Specify model. Please use "fit measured" (not "reproduce").

Response: We removed “model” from the sentence and used “fit measured tracer signal in streamflow” instead of “reproduce”

• Line 128: Remove one "study".

Response: We removed one of "study"

• Line 133: Use "geology of the catchment" (not "area's").

Response: We used "geology of the catchment"

• Section 2.1.: Report soil distribution % consistently for both catchments (or none).

Response: Soil distribution was consistently reported without %

• Lines 143–146: Poor writing; please reformulate.

Response: We reformulated the section as follows lines 143-147:

"We used daily hydro-meteorological data from October 2013 to 2019 for the HOAL catchment (Fig. 1a, 1b) and from October 2009 to October 2013 for the Wüstebach catchment (Fig. 1c, 1d). For the Wüstebach catchment, partial deforestation in October 2013 led to changes in streamflow generation processes, affecting catchment travel time distributions and increasing young- water fractions in streamflow (Hrachowitz et al., 2021). Therefore, the time series after deforestation was not used for the analyses as it would introduce additional model constraints."

• Line 165: Please revise title. A tracer model is also a hydrological model.

Response: We revised the title as "tracer transport model" only

• Line 166 ff.: Unclear. Please reformulate.

Response: We reformulated the sentence lines 167-170: "We used a process-based tracer transport model (Türk et al., 2024) based on the previously developed dynamic mixing tank (DYNAMITE) modelling framework (Hrachowitz et al., 2014), which allows for the simultaneous representation of water fluxes and tracer transport (Hrachowitz et al., 2013) and includes the concept of storage-age selection functions (Rinaldo et al.,2015). "

• Line 168: Define "fast response storage".

Response: We defined that the fast responding storage corresponds to shallow soil water.

• Line 177: Please clarify calibration sequence (hydrological model then tracer model?). Please address over calibration risk.

Response: We clarified in the model definitions (lines 168 - 170): "…which allows for the simultaneous representation of water fluxes and tracer transport (Hrachowitz et al., 2013) and follows the concept of storage-age selection functions (Rinaldo et al.,2015)." In addition, we added in the section 2.3.1 "Model calibration and evaluation" section (lines 271-272): "We calibrated the model simultaneously against both streamflow and stable water isotopes to ensure that both hydrological and tracer information were integrated during parameter optimization"

• Line 178: Use "trace" (not "route").

Response: we used “trace” instead of “route”

• Line 192: Which storage do you refer to?

Response: It is any storage in model. we added “(e.g., S_r, S_f)” for clarity

• Figure 2: Please define all letters.

Response: We thank the reviewer for this suggestion. The symbols for hydrological fluxesnow are defined in the caption, while all symbols for calibration parameters shown in the figure are defined in Tables~S1 and S2, which are explicitly referenced in the figure caption.

• Line 207: Please replace "binds".

Response: We replaced “binds” with “constrains”

• Line 222: Please justify fixing beta at 1. While reducing calibrated parameters is understandable to avoid overfitting, the specific choice of beta (and value 1) appears arbitrary without physical or conceptual reasoning (e.g., prior studies or theoretical basis). Explicitly state here that alpha was varied during calibration. Initially, it seemed both were fixed at 1 (implying uniform distribution), which would undermine the SAS function's purpose and question the entire approach, as uniform selection requires no SAS modelling.

Response: We added justification of parameter selection to lines 225-228 as follows: “In this study, α was varied during calibration to represent different degrees of preferential selection of younger or older water, while β was fixed at 1 to reduce parameter dimensionality and avoid over parameterization. Fixing β = 1 follows previous applications of SAS functions (van der Velde et al., 2015; Hrachowitz et al., 2021) and provides a parsimonious way to explore age-selection behaviour by varying a single shape parameter, while still allowing deviations from uniform mixing.”

• Line 227: Please define "outfluxes"(not used before).

Response: We replaced “outfluxes” with “hydrological fluxes”

• Line 228: Please use "interactions" (not "complexity").

Response: We used "interactions" (not "complexity")

• Line 229: Clarify "outflow" vs. prior "outflux".

Response: we replaced with “hydrological fluxes”

• Line 240: Why isn’t overland flow considered when Sr/Sr,max > 1? This seems physically

plausible, so if there is a specific reasoning or threshold concept behind this choice, please clarify it.

Response: The overland flow is considered when Sr/Sr,max > 1, we added additional information to lines 248-249 the sentence to clarify that: "In addition to saturation excess overland flow (when Sr/Sr,max > 1), infiltration excess overland flow also occurs."

• Line 264: Use "spin-up" (not "warm-up").

Response: We have changed to “Spin-up”

• Line 293: Clarify "fixing".

Response: We used “by using” instead of “fixing”

• Line 298: Please reformulate “It should be noted….”.

Response: We reformulated the sentence by removing “It should be noted….”.

• Fig. 3 caption: Define "stepwise analysis" (Is this the sensitivity analysis?).

Response: We used “sensitivity analysis”.

• Lines 310–314: Reformulate for clarity. It is conceptually confusing to first state that passive storage does not contribute to streamflow, then claim it influences streamflow age composition. Please explain the mechanism.

Response: We reformulated the sentence for clarity lines 222 -226“The passive groundwater storage ($S_{s,p}$) does not contribute to the quantity of baseflow; it exchanges older water with the active groundwater storage ($S_{s,a}$), thereby influencing the age composition and isotopic signature of the baseflow (Qs , Fig. 2) that contributes to streamflow. For the SAS formulation, total groundwater storage was defined as the sum of active and passive components ($S_{s,tot} = S_{s,a} + S_{s,p}$), such that the age-ranked total storage ($S_{s,tot}$) represents the combined influence of active and passive storage on the age composition of baseflow and thereby on the age composition streamflow”

• Line 315: Please use "based on values" (not "to cover the range").

Response: We used “based on the values”

• Line 343: Specify "main features".

Response: We revised it as “main features (e.g. the rise and recession limbs)”

• Line 360: Use "sensitivity analysis" consistently (not "experiments").

Response: We used “sensitivity analysis”

• Line 375: Please reformulate "SAS shape parameter lower bound".

Response: We reformulated as "the lower limit of the SAS shape parameter".

• Line 397 ff.: Specify where shown.

Response: We cited the Figures. 7a, 7b in the sentence

• Table 1: Use "parameterizations" (not "variation").

Response: We used parameterizations

• Section 4.1.: Consider deleting this section. While it reads well, the conclusion (line 462) is not supported by the discussion, as the model was designed with catchment-specific assumptions tailored to observations thus validating against those same data does not demonstrate generalizability or independence

Response: We thank the reviewer for this comment. We considered this suggestion but found it necessary to retain Section 4.1, as it provides context for interpreting the model behaviour and results within the other catchment-specific modelling framework adopted in this study.

• Line 474 ff.: Please delete repetition.

Response: We deleted the repetition in the sentence

• Line 482: Please soften: "This suggests…." (because this is not measured).

Response: We rephrased the sentence. “This suggests younger water to reach the stream with minimal mixing with stored water”

• Line 486: Please add "to streamflow".

Response: We added “to streamflow"

• Line 493: Use "contributions to streamflow". Please define "correlation strength".

Response: We used contributions to streamflow and replaced “correlation strength" by correlation coefficient.

• Line 505: The current consequence reads too technically, resembling a result rather than a

broader implication. Please expand to discuss wider impacts, such as applications to water

quality modelling or transferability to other systems.

Response: We added discussion lines 517-521 as follows to discuss wider impacts, such as applications to water quality modelling or transferability to other systems. “Consequently, uncertainty in estimating passive groundwater storage volumes in modelling leads to uncertainty in transit time estimation and contaminant transport time scales, with critical implications for assessing water-quality risks. For water-quality modelling, this implies that rapid contaminant transport through preferential flow paths can occur in response to hydrological events; however, stable water isotope data alone are insufficient to capture these dynamics in catchments with substantial passive groundwater storage.”

• Section 4.3: This section lacks a clear key message and reads like a results summary rather than discussion (e.g., no interpretation, implications, or context). Consider deleting or rewriting to include analysis of findings' meaning and broader relevance.

Response: We revised the section for clarity and included analysis of findings' meaning and broader relevance, see lines (528-540).

• Line 526: Please restate the meaning of "fell below 1%".

Response: We revised the sentence as “was less than 1 %, indicating that passive groundwater storage volumes orders of magnitude larger than active groundwater storage are required to damp isotope variability in streamflow,”

• Line 537: Please use "TTD".

Response: We used “TTD”

• Line 539: "Under uniform…": Please delete this sentence, as it repeats results. End the paragraph with a strong final statement summarizing key insights instead.

Response: We deleted the sentence "Under uniform”. We revised the end of paragraph lines 558- 564 as follow: “ Because the groundwater storage SAS function was formulated based on the sum of active and passive groundwater storage ($S_{S,tot} = S_{S,a} + S_{S,p}$), increasing passive storage volumes systematically increased the likelihood of older water contributions to streamflow, thereby extending the tails of the transit time distributions (TTDs; $100 < T < 1000$ days). These results underscore that passive groundwater storage is a dominant control on catchment memory (the retention of the past hydrological inputs in streamflow due to long residence and transit times) substantially masking young-water contributions and promoting delayed solute and pollutant transport at the catchment scale.”

• Section 4.5: Please rewrite this section for better flow and clarity. Currently, it reads poorly

(jumpy, lacks cohesion) and fails to convince why the hydrological community needs a model for preferential flow path contributions. Please specify where, when, and why such modeling is essential (e.g., water quality prediction, solute transport, extreme events). Add a clear take home message.

Response: We revised the “Implications and limitations” section lines 565-609 to explicitly discuss the relevance of the modelling approach for water-quality interpretation, including the role of preferential flow in mobilizing solutes during event-driven conditions, the transferability of the framework to other catchments, and how differences in active–passive groundwater storage dynamics may lead to similar or contrasting outcomes across hydrological settings.