

Dear Editor, dear Dr. Dai

let me first of all thank you on behalf of all co-authors for providing extra time to complete the revisions. Let me furthermore thank the three reviewers for their valuable points, which were most helpful for improving our study.

Attached you find the revised version of our manuscript "On the relevance of molecular diffusion for travel time distributions inferred from different water isotopes" as track change version and as clean version. In the clean version we highlighted all substantial changes in yellow, to make it easier for the reviewers to double check how we addressed their recommendations.

In line with the recommendations of Markus Hrachwitz and Mikes we:

- Removed the term self-diffusion from the manuscript, as it created more confusion than clarity.
- Extended the review of the relevant literature to better reflect the methodological development for assessing travel time distributions in lines 50-51, 66 -67 and 84-90 of the revised manuscript.
- We acknowledge the truly interesting and relevant work of Wang et al. (2023) both in the introduction (line 111 -115) and the discussion section (line 488 -492).
- Figures 2 and 3 were changed to assure their readability, and we restructured the presentation of the three different scenarios in section 2.2.
- And we completely revised the results section 3, with new figures 4-6 showing now the cumulated distribution function of travel times (instead of the pdfs) to better highlight the differences between the isotopes. We furthermore added three tables showing mean and standard deviation of travel times for all isotopes as well as the 10%, 25%, 50%, 75% and 90% quantiles of the TTD (line 336-448). The color code in these tables is consistent with the one in Figure 4-6. Naturally we provided a more detailed description of the figures and tables.

The recommendations of the anonymous reviewer are addressed as follows:

- We kept the section on Taylor Aris dispersion, a) because the reviewers Markus Hrachowitz and Mike Stewart, both experts in isotope hydrology and travel time modelling, found it instructive and helpful, and b) because our simulations reveal a clear growth of the standard deviation of the travel time distributions with declining diffusion coefficients of the isotopes (see Tables 2 and 3). However, the revised manuscript stresses, as recommended, clearly that dispersion in porous media is distinctly different from Taylor dispersion (line 217 -228)
- In this context we also corrected Eq. 3 and the dependence of the diffusion coefficient on the fluid viscosity (line 162).
- As requested, we conducted test simulations to assure independence of the particle tracking simulations on the particle number and explain how fluid velocities were interpolated to the particle positions (line 262 -266).
- We provide the requested details on the Catflow model (line 285 -290).
- We re-conducted all our simulation at variable time steps and found convergence for time steps of 1h. These new simulations yielded indeed, in line with reviewers perception, stronger differences in the travel time distributions of the different isotopes, particularly for the high variance case in scenario 5. We are very grateful to the reviewer for this hint.

For your convenience we attached also or line to line response to the reviewers from the open discussion.

Thank you again, best regards,

Erwin Zehe

## Reply to the review by Markus Hrachowitz

Reviewer MH: The manuscript “On the relevance of molecular diffusion for travel time distributions inferred from different water isotopes” by Zehe et al. addresses a so far, at least for studies at larger spatial scales, rarely considered aspect of tracer circulation and its potential effects on water ages and its distributions. Conceived as a modelling study, the experiment is very well designed and quite comprehensive in its extent, analyzing a broad spectrum of potential scenarios. The manuscript is similarly well written, logically structured and, due to clear explanations, easy to follow. Overall, I will be more than glad to eventually see this study published. However, I think the manuscript could be further strengthened by considering the following rather minor points:

Reply EZ: On behalf of all co-authors, I thank Markus Hrachowitz for his insightful assessment of our work and his constructive recommendations.

Reviewer MH: (1) Many different model runs with changing structures of the spatial domains (scenarios A-C), tracers ( $^2\text{H}$ ,  $^3\text{H}$ ,  $^{18}\text{O}$ ), hydraulic conductivities, Peclet numbers, etc. were executed. This is very well described and an excellent approach as it analyses many different aspects and influences on the experiment. However, the results of these model runs are reported in somewhat of a rush and with little detail. I think that systematically showing the results of all model runs – at least as figures in the Supplementary Material – and to provide some more detail in the descriptions of the results will benefit the analysis as it will allow the reader to appreciate these different aspects much more.

Reply EZ: We agree that the results section is maybe a little bit too condensed. In the revised manuscript we will happily go to a greater detail and provide additional Figures, when necessary, in the Annex, to make the study comprehensive and fully accessible.

Reviewer MH: (2) Related to (1), I also think that the Figures can benefit from being developed a bit more systematically and carefully, so that they are easily readable and consistent in their respective structures across the individual experiments. For example, it is unclear why for scenario A the results of  $^3\text{H}$  and  $^{18}\text{O}$  are not shown in Figure 4. I believe it is important to show the entire sequence of results – even if they do not exhibit major differences between the runs – to allow the reader a complete assessment. Similarly, why are estimates of  $\text{RT}_{\text{hyd}}$  only given in Figure 4 for scenario A but not in Figures 5-7 for the other scenarios? Overall, the figures will benefit from a more consistent structure and appearance. In other words, while figure 4 shows  $\text{mean}(T)$  and  $\text{RT}_{\text{hyd}}$  as annotations next to the graphs, and hydraulic conductivities in the legend, the subsequent figures show  $\text{mean}(T)$  in the legend (omitting  $\text{RT}_{\text{hyd}}$ ) and definitions of the model runs, i.e.  $L$ ,  $Pe$ ,  $\text{var}$  next to the graphs in the figures. These inconsistencies bring unnecessary “noise” into the manuscript.

Reply EZ: We omitted the TTD of  $^3\text{H}$  and  $^{18}\text{O}$  for scenario A, because the differences compared to  $^2\text{H}$  were not large. Yet, we fully agree with the reviewer here, and thus will happily revise the figures assuring a more consistent appearance as requested. We particularly think that the comparison of the mean travel time  $\text{mean}(T)$  inferred from the tracer breakthrough and the hydraulic retention times  $\text{RT}_{\text{hyd}}$  is informative and needs to be presented for all cases. The same applies to hydraulic conductivities (their mean and variance).

Reviewer MH: In addition, it will be good to increase symbol sizes at least in the legends, as right now the individual model runs are difficult to discern in the individual figures. Also make sure to include (a), (b), (c)... labels for sub-figures. Right now, it is not always immediately clear which subfigure is which. The figures could also benefit from the use of more systematic colour schemes throughout the manuscript.

Reply EZ: We agree that the symbols were actually too small, and we will assure a more consistent color coding in the revised manuscript.

Reviewer MH: (3) This is a study that builds exclusively on model experiments. That is fine. However, I think it would benefit the manuscript if a stronger link to experimental studies with real world data is established to allow the reader to place the results into a wider context. While the authors refer to previous studies by Stewart et al. and Rodriguez et al. to frame their work, it is surprising that they do not refer to a recent study by Wang et al. (2023). In that study we found considerable evidence that the considerable differences in  $^3\text{H}$  and  $^{18}\text{O}$  estimates of water ages reported by Stewart et al. (2010), are to a large extent an artefact of the choice of model type used by Stewart et al. (2010) and the cites studies therein. Overall, the study by Wang et al. (2023) found that  $^{18}\text{O}$  and  $^3\text{H}$  result in estimates of water ages that are broadly consistent, with estimates from  $^{18}\text{O}$  even showing older (!) ages and thus the opposite of what was reported by Rodriguez et al. and here in the results of this study. Thus, discussion of the results found here in the context of the results reported by Wang et al. (2023) will give the reader a much more complete picture of the current state-of-the-art.

Reply EZ: Our work is indeed a theoretical study, though case A is motivated by lab experiments by Elhanati et al (in press; <https://doi.org/10.5194/egusphere-2025-3365>), though the dimensions are a little bit different. Yet the results can be easily scaled to the dimension of the experiment, and we will do so in the revised manuscript.

We must admit that the main reason for this study was the discussion between Mike Stewart and Julian Klaus, when Mike Stewart (<https://doi.org/10.5194/hess-25-6333-2021>) commented on the paper of Rodriguez et al., which was co-authored by the main author these days. Our argument was basically that HDO and HTO are two times the same molecule, so there is no physical reason why travel time distributions of HDO and HTO may be different. After a while we realized, they are not the same! There is a difference in mass and in the (self-)diffusion coefficient, and these differences may in case of imperfect mixing, even cause difference in average travel times. Thus, our study is in a certain way a late acknowledgment that Mike Stewart had a point.

That said, we absolutely agree that we missed the opportunity to reflect our findings, against the truly very interesting work of Wang et al (2023). What we understood from this work so far, is that the water ages inferred from  $^{18}\text{O}$  and  $^3\text{H}$  with SAS were largely consistent, while related differences were substantial, when using a convolution model. So obviously, we must be very precise about the methods which have been used to infer travel times, otherwise one might compare apples with oranges. As far as we understood the study, it relies on a smart, HRU based hydrological model structure. While this makes much sense, such a structure is per default not sensitive to differences arising from different diffusion coefficients, which requires a spatially distributed velocity field. We will further elaborate on this in the revised manuscript.

Detailed suggestions:

Reviewer MH: p.1,l.15: not clear what is meant by “assessing...chemical species...”. Perhaps rephrase

Reply EZ : maybe better of “water and tracers”

Reviewer MH: p.2,l.48-50: ok, but the concept was also already known and used before that, e.g. Eriksson (1958), Bolin and Rodhe (1973). Perhaps good to include these references, as well

Reply EZ : You are never too old to learn, I was not aware of these studies. We will happily acknowledge those in the revised manuscript.

Reviewer MH: p.2,l.61-64: not sure if this is a valid generalization. The dependence of water ages on water supply has been known for quite a while (e.g. Nir, 1973) and even explicitly accounted for in time-variable formulations of transfer function approaches (e.g. Niemi, 1977). Please rephrase and include the references.

Reply EZ : Sorry for being unclear. We refer here exclusively to the use of transfer functions in the partially saturated zone. We will clarify this in the revised manuscript.

Reviewer MH: p.3,l.82ff: I think a more precise formulation of the history of the various concepts here would benefit the manuscript. Early studies approached the question in fact with both approaches. While indeed many of them relied on time-invariant or time-variant transfer functions (see e.g. review by McGuire and McDonnell, 2006), many others used methods that are equivalent to the SAS function approach, such as the many studies of hydro-chemical dynamics based on the Birkenes and HBV models (Christophersen and Wright, 1981; Christophersen et al., 1982; Seip et al., 1985; de Groisbois et al., 1988; Hooper et al., 1988; Barnes and Bonell, 1996) and in particular nicely illustrated by Fig. 1 in Bergström et al. (1985). The same is true for Hrachowitz et al. (2013) that is now cited as a transfer function based study. This is factually incorrect. In that study we estimated water ages based on tracking tracers through the systems using “mixing coefficients”, which are functionally the same things as the SAS-approach with piecewise linear age sampling distributions (see Hrachowitz et al., 2016 and Benettin et al., 2022 for more detail).

Reply EZ : Fair enough. We will happily elaborate in more detail on the history of both concepts and acknowledge the proposed, relevant work.

Reviewer MH:P3.l.89: probably better to replace Hrachowitz et al. (2010) by Hrachowitz et al. (2021)

Reply EZ : Thanks, we will change the references accordingly.

Reviewer MH: P3.l.89: p.3,l.103-105: true, but this is likely an artefact due to the choice of model by Stewart et al. (2010, 2021) and the studies cited therein. A detailed comprehensive demonstration thereof can be found in a recent study by Wang et al. (2023), who found broadly equivalent magnitudes of water ages inferred from  $^3\text{H}$  and  $^{18}\text{O}$ . Would be good to rephrase and add the perspectives by Wang et al. (2023)

Reply EZ . We will happily address this key issue in the revised manuscript. However, in this context it is also important that Rodriguez et al. found differences in mean water ages in the Weiherbach, although they are both based on SAS.

Reviewer MH: p.3,l.107: Nice!! This is really excellent.

Reply EZ: Thanks.

Reviewer MH: p.3,l.130ff: please rephrase and add Wang et al. (2023)

Reply EZ: With pleasure.

Reviewer MH: p.4,l.137: this is a very useful and clearly described section. One additional thing that I personally would find useful would be to include an explicit description of the difference between self-diffusion and molecular diffusion. After reading this section I am still not sure whether they are the same thing or not and if not, what the difference is.

Reply EZ: We are glad that this is helpful and agree that the distinction is misleading. Molecular diffusion relates to Brownian motion/diffusive motion of a solute, e.g., Br<sup>-</sup> in water, we speak of two different chemicals. Self-diffusion relates to Brownian motion/diffusive motion of water isotopologues (HDO, TDO, 18OHH) in water H<sub>2</sub>O. So, it is the same chemical but not the same molecule.

Reviewer MH: p.3,l: p.10,figure 3: I do not understand the legend in the bottom row. How can particle numbers  $N_p$  be negative?

Reply EZ: This is indeed misleading. The numbers are normalized with the total number of particles - the maximum number is thus one (all particles past through). We will explain this in the revised manuscript

Reviewer MH: p.11,l.304-306: ok, purely technically seen the mean(T) are larger. But given that the difference is  $\leq 1\%$ , how significant/relevant is it?

Reply EZ: Well observed, one would indeed be inclined to classify this as not significant.

Reviewer MH: p.11,l.326 and elsewhere: the term “average” is somewhat ambiguous as it can refer to any measure of central tendency, i.e. mean, median or mode. Thus, please replace accordingly for clarity.

Reply EZ: When using the term average, we generally refer to the arithmetic mean as an estimator of the expected value (the first central moment). We will be precise about that in the revised manuscript,

Reviewer MH: In addition, it would be good to provide the actual values for the different tracers and show the TTDs – either in Figure 4 or in the supplementary material.

Reply EZ: Brilliant idea, we will do so.

Reviewer MH: p.12, p.333ff: given that the TTDs of the individual tracers in figures 5-7 are largely indiscernible and plotting on top of each other, it may be good to also show them with log-scale axes to better grasp the differences. In addition, the exclusive focus on mean(T) may conceal other effects. Thus, perhaps it is interesting to also report and discuss differences in young and old quantiles or the medians.

Reply EZ: Excellent idea, we will do so in the revised manuscript.

Reviewer MH: p.13,l.358: how was the  $RT_{hyd}$  exactly calculated here? Is it based on the flow weighted averages of the upper and lower layers? Does it make a difference whether it is flow weighted?

Reply EZ: This was calculated by dividing the total storage volume in the pore space by the volumetric flow rate. This assumes perfect mixing. It makes a difference when doing this separately in the high and low conductive zone.

Reviewer MH: p.14,l.395-396: ok, but for context it would also be good to mention that this is only  $\sim 2\%$

Reply EZ: absolutely correct, we will stress this.

Reviewer MH: p.17,l.429-433: also here, reference to the results of Wang et al. (2023) is required to provide a full picture.

Reply EZ: absolutely correct, we will stress this. At the end the relative difference matters

Thank you for this interesting contribution!

Best regards,

Markus Hrachowitz

Thank you very much again for the insightful comments,

Erwin Zehe

References:

Elhanati, D., Zehe, E., Dror, I., and Berkowitz, B.: Transport behavior displayed by water isotopes and potential implications for assessment of catchment properties, *EGUsphere* [preprint], <https://doi.org/10.5194/egusphere-2025-3365>, 2025, in press for HESS.

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Wang, S., Hrachowitz, M., Schoups, G., and Stumpp, C. (2023): Stable water isotopes and tritium tracers tell the same tale: no evidence for underestimation of catchment transit times inferred by stable isotopes in StorAge Selection (SAS)-function models. *Hydrology and Earth System Sciences*, 27(16), 3083-3114.

## Reply to the review by Mike Stewart

Reviewer MS: This excellent paper ('On the relevance of molecular diffusion for travel time distributions inferred from different water isotopes', E. Zehe et al.) reports on the travel time distributions expected for different self-diffusion coefficients of  $^1\text{H}^2\text{H}^{16}\text{O}$ ,  $^1\text{H}^3\text{H}^{16}\text{O}$  and  $^1\text{H}_2^{18}\text{O}$  in varied environments. While transport variations with different water isotopes might have been expected, this is the first study to quantify the effects in various situations. The paper is structured as a modelling study simulating the transport of the isotopic molecules in three scenarios representing different hydraulic conductivity fields. The paper is concise, logically constructed and well written.

The abstract is clear and complete. The Introduction focuses the contribution appropriately. I am not qualified to judge whether the three scenarios considered adequately explore the different transport effects to be expected for the isotopic molecules, but relative effects of up to 10% in travel times are demonstrated. The figures could be improved, I found them difficult to read and unpredictable in content. Referencing is full and complete.

Reply EZ: We are extremely grateful to Mike Stewart for his insightful assessment of our work and his constructive recommendations. We will naturally improve the figures, as already stated in our reply to Markus Hrachowitz.

Reviewer MS: The novel approach adds to a current controversy on whether the use of seasonally variable tracers (e.g.  $^{18}\text{O}$ ,  $^2\text{H}$ ) leads to truncation of catchment travel times compared to the use of radioactive tracers (e.g.  $^3\text{H}$ ) (Rodriguez et al., 2021; Stewart et al., 2021). While Rodriguez et al. (2021) obtained similar results for  $^2\text{H}$  and  $^3\text{H}$  for the Weierbach Catchment, Luxembourg, using SAS-function models, the mean transit times for both were only about 3 years. This suggests that a possible explanation for the concurrence of the results with deuterium and tritium is that there was simply not much older water in the Weierbach Catchment (i.e. no long tail of older ages) which if present could have been detected by tritium but not deuterium (Stewart et al., 2021).

Reply EZ: We agree that the absence of very old water in the Weierbach likely explains the moderate difference in the average travel times of  $^2\text{H}$  and  $^3\text{H}$  reported by Rodriguez et al. (2021). Our recent work suggests that these differences are physically based and can be explained by the different self-diffusion coefficients of both molecules in combination with anomalous transport. The Weierbach is in fact highly susceptible to preferential flow phenomena both in the partially saturated zone (Sprenger et al. 2016) and as well as in the weathered shist layers controlling rainfall runoff generation in this setting (Loritz et al. 2017). We will add this point in the revised manuscript.

On a broader context, we note that the main reason for this study was the discussion between the reviewer Mike Stewart and Julian Klaus, when Mike Stewart (<https://doi.org/10.5194/hess-25-6333-2021>) commented on the paper of Rodriguez et al.. Our argument was basically that HDO and HTO are two times the same molecule, so there is no physical reason why travel time distributions of HDO and HTO may be different. In our recent study we show they are not the same! There is a difference in mass and in the (self-)diffusion coefficient, and these differences may in case of imperfect mixing, even cause difference in average travel times. This is because the different diffusion coefficients matter, particularly in the case of a long tailing, which is a manifestation of anomalous transport (Elhanati et al., 2025).

Reply MS: However, Wang et al. (2023) reported mean transit times of 11-17 years with the SAS-function method for  $^{18}\text{O}$  in the Neckar Catchment, Southern Germany, even longer than those inferred using  $^3\text{H}$  (11-13 years), showing that the truncation effect noted by Stewart et al. (2010, 2021) does not apply to isotope data analysed using SAS-function models. Instead the truncation effect noted by Stewart et al. (2010) is attributed to the methods used to infer the mean transit times from isotope data in earlier studies (sine wave and convolution integral methods for  $^{18}\text{O}$  and convolution integral method for  $^3\text{H}$ ).

Reply EZ: We absolutely agree with MS that our findings need to be reflected against the very interesting work of Wang et al (2023). What we understood from this work is that the water ages inferred from  $^{18}\text{O}$  and  $^3\text{H}$  with SAS were largely consistent even in the presence of old water, while related differences were substantial, when using a convolution model in combination with a sine wave approach. Obviously, we must be very precise about the methods which have been used to infer travel times, otherwise one might compare apples with oranges. As far as we understood the study, it relies on a smart, HRU based hydrological model structure. While this makes much sense, such a structure is per default not sensitive to differences arising from different diffusion coefficients, which requires a spatially distributed velocity field. We will further elaborate on this in the revised manuscript.

#### References reviewer

Rodriguez, N.B., Pfister, L., Zehe, E., and Klaus, J. (2021): A comparison of catchment travel times and storage deduced from deuterium and tritium tracers using StorAge Selection functions. *Hydrology and Earth System Sciences*, 25, 401-428.

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Wang, S., Hrachowitz, M., Schoups, G., and Stumpff, C. (2023): Stable water isotopes and tritium tracers tell the same tale: no evidence for underestimation of catchment transit times inferred by isotopes in StorAge Selection (SAS)-function models. *Hydrology and Earth System Sciences*, 27(6), 3083-3114.

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#### References authors

Elhanati, D., Zehe, E., Dror, I., and Berkowitz, B.: Transport behavior displayed by water isotopes and potential implications for assessment of catchment properties, *Hydrology and Earth System Sciences*, 29, 6577-6587, 10.5194/hess-29-6577-2025, 2025.

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## Reply to the anonymous reviewer

Reviewer: The paper investigates the relevance of molecular diffusion for travel times of water isotopes. The authors conduct a numerical study of flow and transport in different two-dimensional media at the continuum (Darcy) scale. A medium with a low-conductivity inclusion (scenario 1), a two-layer medium (scenario 2, flow aligned with the layering) and a multiGaussian medium (scenario 3) with three different logK variances (1, 3, 5) are considered. Transport is solved by particle tracking. In scenarios 1 and 3 only relatively minor differences are observed between the travel time distributions for tracers with different diffusion coefficients. The largest difference is observed in scenario 2, which is characterized by a high and a low-K layer. It is argued that the difference is physical and not "measurement errors".

While for scenario 2, the difference seems to be significant and quite clear based on the matrix-diffusion type scenario the authors chose, this is less clear for scenario 3. In the following I provide a few comments that may be useful to the authors.

Reply EZ: On behalf of all co-authors, I thank the anonymous reviewer for her/his insightful assessment of our work and her/his constructive recommendations. The main reason for this study was the discussion between Mike Stewart and Julian Klaus, when Mike Stewart (<https://doi.org/10.5194/hess-25-6333-2021>) commented on the paper of Rodriguez et al., which was co-authored by the main author these days. Our argument was basically that HDO and HTO are two times the same molecule, so there is no physical reason why travel time distributions of HDO and HTO may be different. After a while we realized, they are not the same! There is a difference in mass and in the (self-)diffusion coefficient, and these differences may in case of imperfect mixing, even cause difference in average travel times. Thus, our study is in a certain way a late acknowledgment that Mike Stewart had a point.

Reviewer: It is not clear why the authors go in such a length through the example of Taylor dispersion in a tube, first, because it has been extensively studied in the literature (references are missing, though) and second, because dispersion in porous media does in general not behave like Taylor dispersion, see the textbook by Bear (Dynamics of Flow in Porous Media), and the papers by de Jong and Saffman for example, among many others. If the authors want to make the point that the Taylor dispersion coefficient is inversely proportional to the diffusion coefficient (dispersion in porous media is not), I suggest to shorten this paragraph significantly, and state only Eq. 3 (corrected either by using the correct coefficients or by using a proportional sign, not an equal sign).

Reply EZ: We of course agree with the reviewer that dispersion in a porous medium is more complex than Taylor dispersion. In fact, we stated this in our manuscript. We also agree with the reviewer that Taylor dispersion is well documented in the literature. We added these details because we suspect that some members of the catchment tracer community may not be aware of this concept, nor the fact that the molecular diffusion coefficient affects in the case of perfect mixing the variance in the tracer travel time distribution, while it does not affect the mean travel time. In the revised manuscript we might move this part to the appendix, depending on the recommendation of the editor.

Reviewer:- At first it is surprising that the authors have not found a larger difference for the low-K inclusion scenario, which is also a matrix diffusion-type scenario. However, the conductivity contrast

may not be large enough so that advection in the inclusion still dominates over diffusion. The authors could check this.

Reply EZ: Good point, we'll happily check this, also by re-running simulations and checking the Peclet numbers.

Reviewer:- In general, for pure matrix diffusion, the residence time in the low-K or immobile zone is proportional to the characteristic diffusion time  $\tau = w^2/D$  (with  $w$  the size of the matrix, or inclusion or layer) and  $D$  the diffusion coefficient. Thus, the diffusion coefficient clearly has an impact on travel times. This can be seen when one considers breakthrough curves in fractured media, for example, that are characterized by a  $t^{-3/2}$ -decay (for instantaneous tracer injection) and a cut-off at  $\tau$ , which depends on the diffusion coefficient and determines the mean travel time, etc. .

Reply EZ: Very valid point. When using the 50% of correlation length to estimate "w" the diffusive time scale  $\tau$  ranges for the isotopes between  $2.07 \cdot 10^8$ s (2390 days) for  $^{18}\text{O}$  and  $2.44 \cdot 10^8$ s (2829 days) for  $^2\text{H}$ , which is a difference of order 20%. So, one would clearly expect a difference in average travel times due to the different cutoff times. We will happily add this point to the revised manuscript. However, we do not see that this point contradicts our main line of argument, as transport in fractured bedrock implies imperfect mixing due to preferential flow and a long tailing.

Reviewer: For scenario 3, a 3% difference is found in the mean breakthrough time that may as well be an effect due to the finite number of particles used in the numerical simulations. Have the authors conducted an analysis of the impact of the particle number on the estimate, for example, of the mean travel time?

Reply EZ: We will double-check the dependence on the number of particles ( $10^5$ ). Note that all simulations were conducted until the last particle left the downstream domain outlet.

Specific comments:

Reviewer: line 149: The molecular diffusion coefficient decreases with increasing viscosity and not with viscosity.

Reply EZ: Indeed, thanks for pointing this out.

Reviewer: line 153: factor  $\rightarrow$  rate. - Eq. 3 is a proportionality and not an equality.

Reply EZ: We will add the correct coefficients.

Reviewer:- lines 213-216: It is not clear what the authors want to say here. They are considering linear transport throughout the whole manuscript. Non-Fickian does not mean non-linear.

Reply EZ: In the ADE advection and dispersion are independent terms, which implies that the molecular diffusion coefficients does not affect average travel times in the case of perfect mixing. We will revise this passage and remove the term "linear" as it is misleading.

Reviewer:- lines 221-222: What is meant by the "symmetry of perfect mixing" and how is it broken by non-Fickian transport?

Reply EZ: We simply meant that perfect mixing leads to a symmetrical, non-skewed travel distance distribution, which non-Fickian transport causes a skewed travel distance distribution. We will revise this passage accordingly.

Reviewer:- Section 2.2: Some more details on the numerical simulations are needed. For example: What scheme is used for the discretization of Eq. 5? What scheme is used for the interpolation of velocity? What is the size of the time step? How many particles are used?

Reply EZ: We happily provide these details in the revised manuscript. Velocities were interpolated to the particle positions from the for nearest neighboring grid points using inverses distance weighting (Roth and Hammel, 1996). Particle steps were conducted using a two point Runge-Kutta scheme suggested by Roth and Hammel (1996). Time steps set to the 10% of ratio of the grid size divided by the maximum darcy velocity or porosity. Particle number were  $10^5$ .

Reviewer:- line 258: A discretization of 0.2 m for a correlation length of 1 m seems to be quite coarse. Are the results for the flow field independent of the grid size, specifically for the large logK variance?

Reply EZ: We think that the discretization is appropriate to the problem. The underlying numerical simulations were already used and thoroughly tested in the studies of Zehe et al. (2021) and Edery et al. (2014).

Reviewer: line 259: Which equation is solved by "CATFLOW" and what numerical method is used?

Reply EZ: Sorry for omitting this. Catflow solves the 2d potential based form of the Richards equation on an orthogonal curvilinear grid, using a mass conservative Picard iteration (Celia et al., 1990). It may thus account address flow saturated as well as partially saturated media.

Reviewer: line 262: How did the authors upscale the medium of Elhanati et al. (2025)?

Reply EZ: Scenario 1 is motivated by the setup of Elhanati, however the hydraulic conductivity values of the media in the model were not taken from the experiment. We will explain this in the revised manuscript.

Reviewer: line 314: What is the value for the total pore volume and how is it determined?

Reply EZ: We multiplied the local porosity with the volume of each grid cell and summed this up over the entire domain. In the third dimension we assumed the same extend as in the vertical. We will provide these details in the revised manuscript.

Reviewer: lines 356-357: How is the hydraulic retention time defined here?

Reply EZ: We divided the pore volume by the averaged volumetric flow. The latter was calculated may averaging the volumetric flow in each grid cell.

References:

CELIA, M. A., BOULOUTAS, E. T., and ZARBA, R. L.: A General Mass-Conservative Numerical-Solution For The Unsaturated Flow Equation, Water Resources Research, 26, 1483-1496, 1990.

Edery, Y., Guadagnini, A., Scher, H., and Berkowitz, B.: Origins of anomalous transport in heterogeneous media: Structural and dynamic controls, *Water Resources Research*, 50, 1490-1505, 10.1002/2013wr015111, 2014.

Roth, K. and Hammel, K.: Transport of conservative chemical through an unsaturated two-dimensional Miller-similar medium with steady state flow, *Water Resources Research*, 32, 1653-1663, 1996.

Zehe, E., Loritz, R., Edery, Y., and Berkowitz, B.: Preferential pathways for fluid and solutes in heterogeneous groundwater systems: self-organization, entropy, work, *Hydrology and Earth System Sciences*, 25, 5337-5353, 10.5194/hess-25-5337-2021, 2021.