Response to Interactive comment by Anonymous Referee #2

Comments from the referee are printed in black. Authors' responses are printed in red.

The manuscript presents and discusses an interesting analysis based on virtual (numerical) experiments on the TTD in small catchments / hilsllopes. The work is interesting and well done and it touches a relevant topic, namely the identification of the leading components and parameters in the definition of TTDs. The approach is rather "classic" in the sense that the analysis is somewhat based on the concept of time invariant TTD, while recent approaches have shown the importance of other metric, like e.g. the backward TT distributions, for a comprehensive description of water age and contaminant dynamics. Still, the analysis is useful and instructive.
We want to thank referee #2 for the assessment of our manuscript and a detailed and thoughtful review that led to a significant improvement of the study. We would like to point out that in our opinion the concept of 'time variability' is implemented in this study since factors causing time variability of TTDs are either changes in catchment storage (e.g. antecedent soil moisture) or changes in atmospheric forcing (like precipitation amount). Of course, there are other/more factors causing time variability we have not explored yet (e.g. erosion, vegetation, different precipitation patterns).

Perhaps the manuscript is too long and involved at times, with plenty of text (with some verbosity) and figures. See for instance the long Conclusion section (and it is the first time I see a subsection there…). I think that this might be detrimental to the work as the reader can easily get lost in the many details and miss the important aspects. Thus, I suggest further distilling the principal results, moving the details that are not important for the storyline in the supplementary material and concentrate on the main results that the Authors want to convey. This would strengthen the message of the work and its diffusion.
A very valid observation. We have struggled and continue to struggle with exactly the problem the referee describes. In the revised manuscript we are going to condense the conclusion and move more of the details to the supplement.

With so many fine details, I miss a description of the physical processes, as observed in the model runs, which determine the TTD. What is the impact of subsurface stormflow? Saturated and unsaturated flows? Groundwater? This is important in order to explain the impact of the parameters examined.
We have tried to always include explanations of the physical processes that play a role in shaping the TTDs for the different scenarios. Apparently that effort was insufficient in certain places. We are going to add more details on the description of the physical processes where necessary in the revised manuscript.

In the following a few specific comments.

- Line 38. I would also cite the pioneer works by Niemi (1977) and Nauman (Residence time distribution theory for unsteady stirred tank reactors, Chemical Engineering Science, 1969).
Thanks for the additional references. It is very hard to get a comprehensive overview of the pioneering work. Niemi is already cited, we will add Nauman.

- Line 55-57. Here the introduction moves to the field of groundwater hydrology, where the issue of the BTC tailing (power-law or not) has been the subject of intense discussions in the last 2 decades or so; this short text and citation does not even scratch the surface and it looks quite superficial here.
In order to avoid the surface scratching, we are going to do some more research on groundwater breakthrough curves and add some more references. It would be great if you could recommend/point out the most important studies so that we are not going to miss them.

- Line 57: The sentence of the "great" underestimation of mass is very much debatable, in most cases it's a tiny fraction of the total mass. It may be important for risk assessment of highly toxic compounds, but uncertainty is anyway very large there.
Agreed 100%. We will make clear that it might not be relevant from a mass balance perspective (but possibly when conducting a risk assessment).

- mTT: please define it (I guess it's mean TT)
You are correct. We will define it at the first mention (line 48).

- Line 94-95. This sentence is repeated in other parts of the manuscript. By definition such approach cannot "completely" erase differences. The question is whether the approximation is good enough for applications. The study by Ali et al (A comparison of travel-time based catchment transport models, with application to numerical experiments, JoH 2014) shows that in many cases it does the job, also considering the several sources of uncertainty, including for instance the estimation of ET (not done here).
We will add the reference to Ali et al. (2014) and discuss your point.

- Lines 137-139. Unfortunately the effective hydraulic conductivity cannot replace the dispersive effects of the distributed macropores because it only impacts the mean velocity. I would delete this sentence as it is not needed: the exclusion of such component is legitimate and meaningful in my view because of the important role of macrodispersion in the TTD determination.
Thank you for the constructive comment. We will proceed as suggested.

- Line 159. vertical or hortogonal to the slope? I guess the latter.
It is indeed vertical and not orthogonal to the slope (but that makes only a small difference).

- Line 163. 5m of dispersivity is quite a lot, even more so for the vertical one. Why the choice? In this case the inclusion of Dfree looks irrelevant.
The longitudinal dispersivity and lateral dispersivity were estimated with regard to the length scale of the model catchment (100 m). $\alpha L \approx 5$ m were estimated using the relation between the longitudinal dispersivity and length scale described in Gelhar et al., 1992 and Schulze-Makuch, 2005 (regression $\alpha = 0.085*L^{0.81}$). We agree that the free-solution diffusion is significantly smaller than the dispersion and could have been neglected. We will clarified this in the manuscript adding the references [Gelhar et al., 1992] and [Schulze-Makuch, 2005].

References:
Gelhar, L.W., Welty, C., Rehfeldt, K.R., 1992. A critical review of data on field-scale dispersion in aquifers. Water Resources Research 28 (7), 1955–1974.
Schulze-Makuch, D. (2005). Longitudinal dispersivity data and implications for scaling behavior. Groundwater, 43(3), 443-456.

- Lines 174-175. What head is provided in the boundary condition? Where is the water table located? This is quite important.

Thanks for catching that. I thought I would have written it somewhere. We will add information on the location of the head (it is equal to the surface elevation).

- Line 204. What is the "subsequent precipitation amount"?

Will be clarified (essentially a measure of the amount of precipitation after the delivery of the tracer).

- Line 214. I guess that mm/a means mm/y

Yes, HESS officially prefers this abbreviation.

- Line 214. Please provide more details on the rainfall time series, e.g. regime, climate etc. As a matter of fact TTD depends also on the rainfall regime, not only the total rainfall per year (e.g. Botter et al 2010).

We agree it is correct that the TTD also depends on the distribution of rainfall. We investigate the influence of different precipitation event frequencies. The precipitation time series we used has the following properties: Average interarrival time: 2.64 days; Average event duration: 3.17 days. The climate in the north west of Germany can be described as maritime temperate (Cfb in the Köppen classification) Maximum precipitation falls usually in June (65 mm), minimum in February (28 mm). We are going to add this information to the manuscript.

- Line 338. I don't like the definition, I would rather speak of "The Inverse Gaussian distribution, with parameters D, …, that is a particular solution of the Advection Dispersion Equation". AD is misleading, as ADE can have several different solutions.

We would like to follow your suggestion. If we reformulate the description in the following way, would it be correct?

1) The inverse Gaussian distribution with dispersion parameter D (dimensionless) and mean mTT (d) that is a particular solution of the advection dispersion equation (Eq. 6):

- Line 401. This discussion is based on log-log plots, which many times are misleading. The convergence of curves at large time can be an artifact of the plots.

It is correct that log-log plot can make large differences at large times appear smaller. However, they also exaggerate small differences at short times. In this particular case we are interested more in the short time differences because we expect the largest differences at the beginning of the TTDs. At late times, differences are averaged out more and more.

- Line 408-409. Differences seems larger to me. Again, the log-log plot does not help.

We double-checked the numbers and they are correct. The fact that the differences seem larger is probably due to the very high resolution of the log-log plot for short and very short times.

- Section 3.3. Some of the (interesting) conclusions here are very similar to those of Fiori et al (Stochastic analysis of transport in hillslopes: Travel time distribution and source zone dispersion, WRR 2009) which I think is important for this work. There, the different parts of the Gamma distribution pertains to different mechanisms and parameters (soil, bedrock, etc.). The main difference is that they identify the important role of KBr in the behavior of the tail, which is the exponential part of the Gamma, which in turn is related to groundwater discharge. The aquifer volume, which depends on water table, thickness and slope, has an important role here.

Thank you for pointing us to this reference. It is indeed a very interesting study that we were not aware of yet. In the revised manuscript we are going to include it and discuss the similarities/differences we found in our work.

- Line 490. I don't see the power law.

We are aware of the fact that straight lines in log-log plots are necessary for identifying power laws but insufficient as evidence. So you are right, we cannot be sure whether they are actually power laws just from this graphical analysis. Would you have a recommendation on how to call these tails instead?

- Line 510. How is the fitting done? What inference methods? How one can say that a distribution performs better than another? Any statistical test?

In Section 2.4.3 (Fitting) we describe the procedure. It was done by the least squares method on the cumulative distributions.

- Line 668. I don't agree with this analysis, the presumed power-law tail covers less than one logscale. Also, identification of power law tails is not simple (see e.g. Pedretti and Bianchi, Reproducing tailing in breakthrough curves: Are statistical models equally representative and predictive? AWR 2018), the emergence of a (short) straight line in a log-log plot may not be enough. At any rate, I would not say that the inadequacy of the distributions in fitting the TTD is because of the tail, that by the way involves a tiny fraction of the mass, which is magnified by the log-log representation. I think that the issue of powerlaw tails is too much emphasized here.

We agree with your comment. We would like to find a better description of the TTD tail behavior (maybe we should just describe the fact that the tails begin with a sudden break in the slope of the TTD and continue from there on as straight lines on a log-log plot). It's also clear that the tails are not relevant in terms of the total mass balance and will hardly be noticed for most solutes – with the exception of highly toxic pollutants. We will make sure to stress this in the revised manuscript.

- Section 4.2. This part is not entirely convincing, I can't see the validity of the prediction based on F. By the way the latter does not include other relevant ingredients, like e.g. KBr.

We understand your concerns. This section is not meant to represent to full and complete truth about TTD shapes. It is rather an attempt to find some structure in the way TTD shapes change with certain parameters, an attempt to explore overarching principles. Many of the potential shape-controlling parameters are still excluded from this analysis (like KBr). We will try to put more emphasis on this interpretation of our results in the revised manuscript.

- Line 750. Again, the method cannot erase “all” differences, but perhaps is adequate for many applications.
Agreed. We are going to add this remark to the revised manuscript.

- Conclusion section. It is too long, one cannot see immediately the main results of the work. It’s a pity because there is a lot of interesting material, that however needs to be better distilled and conveyed.
There is definitely room for improvement in the conclusion section. We agree with your criticism and we will do our best to further condense the conclusions in the revised manuscript.

- Line 754-755. “…it is possible to predict the change using the dimensionless flow path number F.”. At the third line of the Conclusion section this seems the major conclusion of the work. Is it so? It does not seems like after reading the text.
This can indeed be considered the main conclusion of our work. We need to make sure that this outcome is conveyed better in the revised conclusion section.