We thank the reviewer for these comments on our manuscript. Below we respond (in bold type) to the reviewer's comments (in normal type).

The authors have developed a MATLAB script and an R script that estimates new water fractions and transit time distributions (TTDs) based on Kirchner's 2019 method. The method has been extended in this manuscript to provide robust estimations when outliers are present. I believe that this manuscript can serve as a good manual for potential users of that script. They have also provided some thoughtful analyses that would help users understand the potential limitations of the method. The manuscript is well-written and mostly ready for publication. I only have some minor comments to help increase readability. Also, as a potential user, I have a few questions for the authors on how to use the method correctly.

1. Required number of samples

Could you suggest, at least, a rule of thumb number for the minimum number of samples required to perform an analysis using this method? I do not think that there would be a definite answer, and I guess it would depend on which analysis a user wants to do (among many others). Still, any suggestion would help potential users design their sampling strategy for their analysis of interest.

The required number of samples will indeed depend on many other factors beyond just what analysis the user wants to do (new water fraction vs. TTD). Some of those factors include:

- a) how variable are the tracer concentrations in precipitation, and over what timescales?
- b) how variable are the tracer concentrations in streamflow, and over what timescales? (Note that this will depend not only on the answer to (a), but also on the timescales of catchment storage in other words, what the transit time distribution is.)
- c) how stationary vs. non-stationary (time-invariant vs. time-variant) is the catchment's transit time distribution?
- d) how large are the measurement uncertainties in the tracer concentrations? Are the measurement errors serially correlated, and by how much?
- e) what errors or uncertainties in the results (Fnew and TTD values) are acceptable?

Many of these factors will be unknown in advance (for example, the sample size needed to estimate a TTD will vary, depending on what that TTD is, which will not be known – that's the whole point of estimating the TTD in the first place). Thus it is difficult at this stage to provide a rule of thumb, until we (and the community) gain more experience with real-world applications.

In the meantime, the most informative approach is to generate benchmark data sets under a range of assumptions, and then test how the sample size affects the accuracy of the inferred Fnew and TTD. Unfortunately we cannot recommend a way to short-cut that process.

2. New water fractions and TTDs estimations when the TTDs are humped

The authors showed that the method overestimates uncertainty associated with the estimated averaged TTD when TTDs are humped. They argued that nonstationarity (time variability) of the

TTDs might have caused the overestimation problem. If that is the case, is it possible to get better uncertainty estimations when one estimates TTDs for each subset (assuming that the subsets are well constructed)?

We already tried this and unfortunately it doesn't work. Or rather, it might theoretically work, but not under conditions that are likely to be encountered in the real world. Consider a simplified nonstationary system that has two different states, "wet", and "dry", with a different (stationary) TTD in each of these two states. If the "wet" state lasted long enough that the catchment stayed wet between the time the tracer entered the catchment in rainfall and exited in streamflow, and likewise if the "dry" state lasted long enough that the catchment stayed dry between tracers entering in precipitation and exiting in streamflow... and if one could cleanly split the data set between the "wet" and "dry" subsets, then yes, the strategy described by the reviewer could potentially work.

But this would require that the timescales over which the catchment switches between wet and dry conditions were much longer than the timescales over which the catchment stores tracers, which will rarely be the case. In the messy real world, by contrast, many different precipitation events, and many changes in catchment conditions, are overprinted on each other between the time that tracers enter in precipitation and leave in streamflow.

The authors also showed that the method overestimates the new water fraction at the daily time scale when the TTDs are humped. While they have shown that the issue can be resolved at the weekly time scale, I think that there is a way to get a good estimation at the daily time scale. Some of their explanations about the overestimation of the new water fraction and the results that are shown in Figure 6 imply that the method could estimate Fnew pretty well at the daily time scale if one estimates TTDs first (probably with m about 7 days in this case, and for each subset to alleviate the uncertainty overestimation issue) and then use Beta_0 for QFnew?

We already thought of this and already tried it, and the reason we didn't describe it is because it didn't work. (Indeed if we mentioned every intuitive-sounding idea that doesn't work, and explained why it doesn't work, the paper would be many times its present length.)

3. On the use of IRLS

The role of IRLS is a bit unclear. Their robust estimation method consists of two steps (the MAD-based filtering and the use of IRLS), but those steps' relative importance is not discussed. As the authors described in lines 173-178, IRLS could be an additional source of getting less accurate estimates. Would it be possible that, in some cases, the method estimates better TTDs and new water fractions when only the filtering is applied? Then, I think it would be great to provide an option to do the MAD-based filtering separately.

We really don't think this would be a good idea. MAD-based filtering and IRLS, like any other robust estimation procedures, will both reduce the accuracy of any results that rely on extreme values that are not actually outliers (but instead, for example, are simply the very long tails of an outlier-free distribution). If, on the other hand, the extreme values are indeed outliers, then these procedures will greatly improve the accuracy of the results (relative to those from non-robust analyses corrupted by outliers.

The only case in which it would make sense to used MAD-based filtering and not use IRLS would be if we knew that <u>all</u> of the outliers were big enough to be detected and removed by MAD-based

filtering, and <u>none</u> were small enough to get through the MAD filter. Such a situation seems highly improbable. Thus the decision to use robust estimation or not is, in our view, an either/or decision that the user should make.

4. Clarifications

L9, L65: I am not sure if the method can "measure" TTDs and new water fractions.

We can say "estimate" instead since the TTD and Fnew are not measured directly.

We will note, though, that many quantities that are actually estimated from proxies are typically called measurements instead. For example, an altimeter actually <u>measures</u> air pressure, and uses it to calculate (or infer) altitude. A GPS unit actually <u>measures</u> the relative arrival times of radio waves from GPS satellites, and calculates or infers the user's position. But most people have no problem saying that an altimeter measures altitude and a GPS measures one's location.

L58: It is hard to understand why the strongly biased outliers are harder to detect and eliminate.

Strongly biased outliers are harder to detect and eliminate because they shift the mean of the distribution, making it harder to distinguish between the outliers and the un-corrupted data. Data with strongly biased outliers may also be difficult to distinguish from the naturally skewed distributions that characterize many environmental variables.

L61: "Large enough" – Wouldn't it makes the outliers easy to detect?

That depends on the detection technique. The example shown in Fig. 2 involves some outliers that have so much leverage on the fitted line that it lies close to them – and thus they are harder to detect as outliers based on their residuals (which is why we can't rely on IRLS alone to do the job, since IRLS is based on identifying unusually large outliers).

L317: The authors have used the term "nonstationarity" frequently throughout the manuscript. If I understand correctly, I think it should be "time variability," not nonstationarity.

Nonstationarity refers, in conventional usage, to the time-variability of the statistical properties of a quantity (typically a distribution). Thus "nonstationary" and "time-varying" are typically used interchangeably. To make this equivalence explicit, we will add "(i.e., time-varying)" after one use of "nonstationary" and "(i.e., time-invariant)" after one use of "stationary".

L330: Perhaps better to provide the lag-1 serial correlation rsc for the non-humped TTD cases.

The goal of this analysis is to show how the non-stationarity of the humped TTDs leads to inflated standard errors. To show this, we compare results from stationary and non-stationary benchmark models that have similar average TTDs. The stationary and nonstationary benchmark models have the same parameter values, but one has constant precipitation (giving a stationary TTD) and the other has time-varying precipitation (giving a nonstationary TTD). If instead we compared benchmark models with different parameters, as suggested by the reviewer, we would not be able to demonstrate the role that non-stationarity plays in generating large standard errors.

Figure 2: CP and CQ notations here do not match with the notation used in the text. In the text, the double subscript notation is used.

That's a formatting issue in the plotting program, which doesn't allow double subscripts. We'll fix it by hand.

Figure 2b: Coloring the corrupted data point (using different colors for the corrupted CP and CQ) would make the figure easier to understand.

Good point. We will re-plot the figure with the same colors shown in Fig. 2a, and the outliers in black.

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