

## Response to Reviewers

# Aged streams: Time lags of nitrate, chloride and tritium assessed by Dynamic Groundwater Flow Tracking

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**Anonymous Referee #2** Received and published: 31 January 2020

**OVERALL RESPONSE:** First we would like to thank the reviewer for taking the time to review our manuscript and for their comment that our manuscript is well-written. Based on the reviewer's comments, we will rewrite large parts of the text, change some of the structure and add some extra analysis.

The large changes to the manuscript are:

- We will clearer articulate the innovative method that we use: this paper is one of the first to apply dynamic Travel Time Distributions to explore and interpret the processes driving observed water quality and isotopes dynamics.
- The sensitivity analysis has been renamed to an 'exploration of the model behaviour under different scenarios', which better reflects the aim of the analysis. Parts of the manuscript have been rewritten for this.
- The measurements of concentrations in-stream will get a more prominent place in the manuscript. They will now be presented in a new paragraph in the Results: "3.1. Water quality measurements of surface water" and more detail will be added on e.g. the time of observations, the resolution of the data and the amount of measurements. We will discuss the seasonal variability shown in the data and calculated by the model in more detail. Likewise, the constructed input curves are now added to the Results section.
- The way that the unsaturated zone was incorporated in the method is clarified and more extensively discussed.
- The discussion will be extended and rewritten to focus on the hydrological and chemical processes that occur in the breakthrough of agricultural solutes and the associated time lags, especially for nitrate. For instance, an extra scenario has been added in which a distinction is made between directly-available nitrogen and organic N that is more slowly leached towards the groundwater. We also intend to better define the concept of 'time lags' to avoid confusion over this term.
- Paragraph 4.4 on 'Improving and use of the model' for the study catchment, which included Figure 10 with the 'Best model fit' will be removed. This part of the manuscript was confusing and distracted from the overall aims of the manuscript. Relevant parts from this paragraph have been added elsewhere in the paper.

We will discuss the points raised by the reviewer step-wise below.

### **General comments:**

In terms of the question of novelty and relevance to an international audience, I was unable to determine this from the abstract and conclusions. For instance, the abstract sets out what was done but doesn't really articulate any major contribution to wider hydrological understanding in terms of enhanced model development or physical processes. The main conclusions are: [list]. None of these findings are particularly insightful – rather they demonstrate that the model produces “as expected” behaviour. Which leads to the question: how such conclusions provide a meaningful contribution to the international literature?

**RESPONSE:** We agree with the reviewer that the listed conclusions do not reflect the innovative work done. This paper is a first attempt to couple measurement data of water quality and isotopes to Travel Time Distributions calculated using a groundwater model. Even though many recent papers suggest that this method holds a lot of potential, but the papers which attempt to do so are absent or at least very scarce. Furthermore, much confusion exists about the effect (and importance) of the rate of change of the input (legacy nitrate) on the lag in the breakthrough. Which we demonstrate and explain in the paper. Lastly, the conclusions on the effect of location of fields and riparian zones are interesting for water managers. We will focus on rewriting the Discussion, Conclusions and Abstract to better reflect these points.

In terms of the model application, an existing groundwater flow model was predesigned for a different purpose (the representation of water levels and flows rather than to represent solute transport: lines 180-185) and this was then applied to the case of solute transport and subject to a sensitivity analysis to consider potential uncertainty in the resulting outputs. This seems rather odd. Surely a study of solute transport needs to rely on a model that is built and calibrated to do just that? There are many such models available and there isn't a sensible justification why this was not attempted. It would be interesting to see a comparison of the approach taken in this manuscript with a bespoke model (calibrated for the purpose) to demonstrate how sensitivity analysis may be used for the purpose described here, but that hasn't been presented.

**RESPONSE:** Goal of the manuscript was to explore and test the use of a groundwater flow model, particle tracking and Travel Time Distributions to calculate the breakthrough of solutes. In this groundwater driven system, a correct representation of groundwater flow paths and travel times is needed and provided by groundwater models such as the one we used. This method has been discussed in several papers, but so far, the papers which attempt to do so are absent or at least very scarce. We agree that the wording in lines 180-185 was poorly chosen and this has been modified. We agree with the reviewer that comparing this method to other models build for solute transport could be interesting, but this was not in the scope of the current paper. The focus of the current paper is on: 1) testing if the results of the TTD method are realistic by comparing them to measurements of solutes, 2) explore the important processes in the method, and 3) explore the effect of the different parameters on the breakthrough and time lags of different solutes as a way forward in a) model calibration and b) management of (ground)watersheds. We will make sure to further clarify the goals and novelty of the manuscript in the text.

A second major issue here is the function of the unsaturated zone. A large body of work in the international literature has outlined the complexity of unsaturated zone processes in some detail, and particularly their impact on solute transport. It therefore seems that a useful model linking land-based solute inputs to stream solute responses would require a detailed representation of the unsaturated zone processes in order to provide the sort of management decision-support that this manuscript purports to give. The unsaturated zone assumed in the model was of uniform thickness and did not account for any macropore flow or chemical processes. This would seem to have essential processes omitted and, therefore, automatically limits the utility of the model. I am therefore struggling to see how this can be used as a reasonable representation of the catchment processes.

**RESPONSE:** In this modelling study we focus on the groundwater system and highly simplify the unsaturated zone. The unsaturated zone is included in the method in two ways: 1) the groundwater model is coupled with an unsaturated zone model (MetaSWAP, see also Kaandorp et al., 2018 WRR) to provide a realistic groundwater recharge based on e.g. land-use; 2) the input curves include part of the unsaturated zone processes: before 2000 by using a nitrate transformation factor of 0.85 and after 2000 by using the concentrations of the shallow groundwater to construct the input curves. Thus, we do take into account many of the unsaturated zone processes, just not the delay.

Furthermore, the relevance of the unsaturated zone also depends on the research area and the temporal scale of interest. In this case, the research area is a lowland groundwater-driven catchment with high groundwater levels and the focus is on seasonal changes in flow paths and solute concentrations in the stream. This can be well simulated with a simple representation of the unsaturated zone. In a more hilly catchment, or if we would want to simulate the concentration response to individual events, a more detailed representation of unsaturated zone hydrology would be needed.

We will make both these points clearer in the manuscript text.

The sensitivity analysis was particularly thorough and seemed to confuse sensitivity analysis with uncertainty analysis. It is therefore unclear as to what this evaluation actually provides in terms of confidence in the model structure, parameterisation or predictive capacity.

The sensitivity analyses described seem to be very simplistic: the state of the art in model assessment for parameter, structural and data sensitivity are far more advanced than is being attempted here. Given the sensitivity analysis is being used to justify the validity of a model calibrated against water levels and flows as a way to represent solute transport, it would need to be much more comprehensive to provide confidence that the approach was appropriate and robust. I also found the framing of the sensitivity analysis rather confused (line 185). Is this a sensitivity analysis or is it an uncertainty analysis? From what is provided in the manuscript it is not at all clear how uncertainty has been accounted for.

**RESPONSE:** We agree with the reviewer, the term 'sensitivity analysis' was misleading for what we tried to do. We have changed this term throughout the manuscript to an 'exploration of the model behavior under different scenarios', to better represent the actual method and goal. Goal of the exploration of the model behavior under different scenarios is to demonstrate the effect of the different parameters that are included in the model. Using these results, we discuss in what way different processes are important for (the time lags in) the breakthrough curves of solutes. We will further clarify the goal of the analyses that is done.