Response to Reviewers

Time lags of nitrate, chloride and tritium assessed by Dynamic Groundwater Flow Tracking in a Lowland Landscape

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Anonymous Referee #3

General comments:

The authors clearly put effort into the revision of the manuscript. I think this is a state that allows for publishing this after some minor revisions. I don't believe in the scenario analysis the authors implemented but don't insist on changing this. I am still a bit more critical with the discussion as you may see below. But in the end, this is all satisfying and I really appreciate the management implications.

RESPONSE: We would like to thank the reviewer for again taking the time to review our manuscript and providing us with constructive feedback. We are happy to hear that the reviewer acknowledges the effort that we put into the Revisions. We will address the reviewer's comments below:

Specific comments:

Abstract

- L83ff: You explore the potential reasons for time lags in addition to the physical groundwater transport component captured by the travel time model, right? Can you make that explicit here?

RESPONSE: Yes, agreed and rewritten.
Revised text: *"In this follow-up paper we used these previously calculated dynamic travel time distributions and the accompanying spatially explicit groundwater flow paths to explore the potential reasons for time lags in agricultural contaminants delivery towards a stream. For this purpose, we added transport of tritium, chloride and nitrate towards the stream and compared simulated with observed concentrations in the Dutch Springendalse Beek stream from 1969 until present."*

- L85ff: The idea of adding information from Tritium and Cl is introduced after all the potential biogeochemical reasoning for time lags in nitrate response. For me it makes sense to introduce that the other way round - you verify/ challenge the model with conservative tracers and then discuss the additional reasons for time lags. This is also stated that way in line 95ff.

RESPONSE: Agreed, we moved the sentence on the reasoning for time last towards the end of the paragraph.
Revised text: *"In our study, tritium, chloride and nitrate were measured. …*

… Subsequently, we exploit the strong physical basis of particle based flow paths to explore…"

- L170ff: It would be helpful if you state a mean and / or median calculated travel time here. Maybe even for the agricultural fields compared to the entire catchments. This can be taken up in the presentation of the initial water quality runs.

RESPONSE: Both were stated in the paragraph, but we agree with the reviewer that they deserve more emphasis. We have included them earlier in the paragraph. We also added a sentence on the different travel times from agricultural fields.
Revised text: *"Using this model approach, Kaandorp et al. (2018a) showed travel time distributions with an exponential- like shape with a mean travel time of approximately 11 years and a median travel time of around 4 years."*

"Interestingly the mean travel time from agricultural fields is approximately 13 years in the upstream catchment compared to 11 years for the total catchment (median TT 8 vs 4 years)."

- Line 256ff: I am not insisting here on changes anymore but would like to state for the records that I am not convinced about this scenario. To show that the addition of 5 years delay in the unsat zone will add a delay to the total travel time, no model or scenario is needed. In addition it is clear that the shallow groundwater table in this catchment does not allow for such long delays. For me this is useless as your aim should be to explain time lags observed in you catchment, not in general.
- Line 277f: The former comment is also true for this scenario. Again, I state that there is no scenario needed to multiply the groundwater TTD by a factor of five. I would rather just state and discuss the time lags between modelled and observed (e.g., conservative) constituents and carefully state potential reasons. Surely, an underestimation of groundwater TTs is one of them.

RESPONSE: We agree with the reviewer that some of the 'scenarios' are rather simple and may not deserve the label 'scenario'. E.g. the delay of 5 years does not require a new run of the model, which would be expected when applying a scenario. However, we want to present and discuss these simple 'scenarios' to be able to compare the effect of different model changes, including the more complex scenarios.
Revised text L264: *"In our model exploration we applied a rather simple modification of a time delay of 5 years for all particles in our model,"*
Revised text L278-282: *"In Set 2, we explored the effect of the saturated zone travel time in a rather simple way by applying a multiplication factor of 5 on all the calculated travel times of all flow paths. For this, the calculated travel times at the moment of discharge to the stream of all individual particles was multiplied by this factor during postprocessing."*
We have also rewritten the introduction of the scenarios in lines 250-254 to also better include the simple model modifications:
Revised text: *"It was not further calibrated for solute transport, but instead we explored the model behaviour through comparing the effect of some relatively simple changes in model parameters, input pre-processing, or output post-processing. These model scenarios enabled us to compare the effect of the following processes on the breakthrough of solutes in the upstream part of the catchment:"*

- Line 383ff: Maybe I missed something here: Is an unsaturated zone delay affecting tritium at all? During the unsaturated passage, there is still atmospheric exchange possible? Does the tritium-clock starts ticking when the rain hits the ground or when the water reaches the groundwater surface?

RESPONSE: Atmosperic exchange was neglected so the 'tritium-clock' starts ticking when rain hits the ground.
Revised text: *"We assumed that the tritium decay starts in the unsaturated zone directly after infiltration."*

- L411ff: This section would benefit, as mentioned above, from a description how computed travel times diverge between agricultural fields and the entire catchment.

RESPONSE: Agreed. Earlier on in the paper we added the difference for agricultural fields and the whole catchment (see comment on L170ff). For Scenario 4 we added this difference as well, following the suggestion of the reviewer.
Revised text: *"For the 200 m zone around the stream, the mean TT towards the stream is approximately 3 years, for the area outside this zone the mean TT is approximately 15 years (median TT 1 vs 12 years)."*

- Line 524-546: All these text introduced the idea of varying contributing areas in general. This belongs rather into the introduction. Only in lines 546-555 you refer to your catchment. This is quite unbalanced. Moreover, the specific discussion if and how the spatial arrangement of sources changes the output signal is discussed in line 513-522 already. So for me this section does not go significantly beyond what is already known. I suggest to start from the specific case made here and after that broaden the view to generalize a bit.

RESPONSE: We do agree with the reviewer that there is quite some introduction for the idea of varying contributing areas. Figures 9 and 10 are important figures generalizing the findings from the scenarios in section 4.1 and these figures require enough introduction and explanation. These figures go beyond the concepts of Rozemeijer and Broers (2007) by adding the 3D concept and including travel times, and this has not been presented before in literature.
Following the reviewers comment we considered moving parts of this paragraph to the Introduction. However, we have rewritten the Introduction and Discussion sections multiple times in previous review iterations and we are afraid that moving text towards the Introduction and changing its structure will weaken the Introduction as it is at this moment. We therefore decided not to move these lines. We have rewritten lines 524-546 to start with our specific case and findings and broaden from there.
Revised text: *"In the previous paragraph, we have seen that the spatial distribution of inputs and processes is an important factor in the breakthrough of agricultural solutes. In our model, particles have a starting point and ending point for which we introduce the terms 'groundwater contributing area' (GCA) and 'runoff contributing area' (RCA) (Figure 9). The groundwater contributing area (GCA) is defined as the area where the water that is actively contributing to streamflow at a certain moment of time through active flow paths entered the coupled groundwater-surface water system as precipitation. This is thus not the same as the catchment of a stream, which is the area in which all discharging water finally ends up in the stream and does not include a time-variable component. The runoff contributing area (RCA) is defined as the area where at a certain moment in time water is leaving the subsurface domain (catchment storage) to become discharge, and thus is the area where runoff is generated (Figure 10). Within this catchment, areas that are neither groundwater- nor runoff contributing areas do not actively contribute to streamflow at that specific moment in time. The concept of contributing areas can thus be used as a different way of describing the catchment and spatial processes and their effect on stream chemistry."*

- Line 614-617. In all the text on nitrate in your catchment, these are the only two sentences that try to explain why there is a time lag between nitrate and chloride. And this seems to be rather unrelated to all the scenarios. I would expect to see a speculation on all of what you know from the catchment and the scenarios, why there is a mismatch in time lag between model and conservative solute observations, between model and nitrate and by that also between observed chloride and nitrate. What do you think is best explaining this?

RESPONSE: In the initial submission of our manuscript we had a section 'Improving and use of the model' which discussed the points that the reviewer is referring to. Here, we tried to use the model to better describe the measurements and did so by adding an unsaturated zone delay and by making some changes in the spatial layout of agricultural fields which is acceptable given that the geology of

the ice-pushed ridge is known to be more complex than the conceptualization in the model and that land-use in the catchment has slightly changes in the past decades.
However, in previous review iterations reviewers recommended us to remove this section and keep the focus on the concepts of time lags. We followed the reviewers suggestions and removed this section.
To further answer the reviewers question: probably a combination of different factors (as shown in the scenarios) best explains the observations. One reason is that the geology in the catchment is highly complex which could mean that the volume of the groundwater reservoir is larger (thus longer travel times) and that the locations of the agricultural fields that contribute to discharge is different. Furthermore, the slow mineralisation of persistent N plays a role (Set 1b) and another thing that we did not considered in the scenarios is that spatial differences in the input exists due to different uses of manure and fertilizers (more Cl).