July 10, 2020

Dear Dr. Yool,

Following the reviewers' comments and your recommendation, we have made technical corrections in our manuscript entitled "A multirate mass transfer model to represent the interaction of multicomponent biogeochemical processes between surface water and hyporheic zones (SWAT-MRMT-R 1.0)".

Attached please find our response to the reviewers' comments.

Sincerely,

Yilin Fang and Co-authors

Response to review comments

Anonymous Referee #2

My only further suggestion is related to the results of section 4.4, and specifically on the estimated differences on nitrate removal (+3% vs -40%) depending on the modeling assumptions of multiple exchange zones. Beside stating the results, it would be useful that the authors provide some comments on the relevance of these findings: for instance, if the flux-weighting is thought to be more correct, the results imply that other approaches tend to overestimate nitrate removal; if it is unclear which approach is more reliable, then the results show how uncertain it is to estimante nitrate turnover and demonstrate the need for more research on this aspect. It is not required to give a definite answer in the manuscript, but a comment should be provided.

Response: We thank the reviewer for the further suggestion! We added the following comment in section 4.4 of the revision:

"These results show that the assumption of the exchange flux associated with each sub-storage zone can have a significant effect on the estimation of HZ nitrate removal and there is a need for more research on the reliable estimation of these exchange fluxes."

There is also a misprint in the title of fig. 7a ("Nitriate").

Response: Corrected.

Anonymous Referee #3

Manuscript Number: gmd-2019-301

Title: A multirate mass transfer model to represent the interaction of multicomponent biogeochemical processes between surface water and hyporheic zones (SWAT-MRMT-R 1.0)

Authors: Yilin Fang, Xingyuan Chen, Jesus Gomez velez, Xuesong Zhang, Zhuoran Duan, Glenn E. Hammond, Amy E. Goldman, Vanessa A. Garayburu-Caruso, and Emily B. Graham

Type: Second Submission (First review by this referee)

Recommendation: Publish

Overview: The manuscript describes the synthesis of three previously described models (NEXSS, SWAT and PFLOTRAN) into a coherent framework (SWAT-MRMT-R) that tracks hydrologic and biogeochemical processes at the watershed scale (and possibly larger) with reach scale resolution. Hyporheic processes, both transport and reactive, are explicitly included. The capabilities of the SWAT-MRMT-R model are demonstrated by its application, in a numerical study, to the Hanford watershed of the Columbia River. The fate of nitrogen solute is evaluated under several different scenarios. General comments: The study of solute fate at watershed and basin scale or even global scales has been an area of increasing interest of late and the Fang et. al. manuscript is a significant contribution to that effort. Overall, the manuscript is will written and well organized. Objectives are clearly stated, and the research has met the stated goals. Conclusions are well constrained by the observations and data. In fact, I would like to congratulate the authors for resisting the temptation (if such temptation was ever present) to describe this work as a predictive model. I see a bit of an alarming trend within our community to do a purely numerical study and call it a predictive model. This is, in my view, a significant overreach. Your work is a significant contribution as it stands. And with some development and field validation could evolve to a predictive model that has real-world impact.

My recommendation is to publish. I feel that the manuscript could be published as it stands, but I do have some suggestions and comments that are worth considering.

Response: We appreciate the reviewer's recognition of our work and helpful comments. We have included the comments in our revision.

NEXSS is not explicitly integrated into your modeling framework. However, NEXSS or something quite similar is essential to the successful implementation of the model. NEXSS does not seem to be readily accessible (maybe I am looking in the wrong place). If your goal if to deploy this framework to other researchers, then it would be quite useful to either make NEXSS more readily available or to provide a bit of guidance as to how the necessary information could be synthesized for large scale studies.

Response: NEXSS will be made available soon.

Overall you do a good job of describing the limitations of your modeling framework. There is one other factor that I think deserves some mention, even though it may seem completely obvious. The way that this model is parameterized with multiple reaches each possibly with multiple storage zones, transfer rates and reaction rates, means that, at the outlet, there are possibly a multitude of parameterizations that can yield the "right" answer – most of them for objectively the wrong reason. As such, if this model is to be used in real-world, large-scale studies then validation at multiple points within the study area should be strongly considered and recommended.

Response: We added the following in the conclusion:

"It is worth noting that this model has a multitude of parameters at reach scale. For real-world, large-scale studies model validation at multiple points within the study area is necessary."

I have some minor suggestions which are listed below.

Specific comments (need to be addressed in the narrative or explain why there is no need to revise):

1. Author listing – I believe that Velez should be capitalized.

Response: Corrected.

2. Line 15: "A two-step reactions for denitrification and an aerobic respiration reaction are assumed to represent…" There is a mismatch in tense. Could be modified as: "A two-step reaction sequence for denitrification and an aerobic respiration reaction is assumed to represent…" or something similar.

Response: Revised as suggested.

3. Lines 92 – 95: the wording is a bit ambiguous. I believe that you are saying: - solute reaction equations are solved by the Newton-Raphson method - hydrologic transport is solved by operator splitting Please clarify for readability.

Response: We clarified as follows:

"the operator-splitting approach in which the solute transport and the reaction steps are solved separately in SWAT"

4. Section 2.4: These are questions/suggestions about future efforts that likely arise from choices that were necessarily made to facilitate the present study. Does SWAT surface water module include denitrification include denitrification? Recent studies have shown this to be significant in high-order, turbid streams. For future iterations of the model, it would be interesting to include NO3 - -→ N2O↑ as this is a significant reaction in large rivers, mostly in the water column but also in the HZ.

Response: SWAT surface water module does not include denitrification. It will be considered in the future iterations of the model.

5. Line 160 – 169: The transition from the discussion of SWAT to PFLOTRAN is a bit abrupt. Suggest the following revision:

In SWAT, dissolved nutrients are transported with the water and those sorbed to sediments are allowed to be deposited with the sediments on the bed of the channel (Neitsch et al., 2011). PFLOTRAN is an open source, massively-parallel reactive multiphase flow and multicomponent transport code. It has well-established documentation (https://www.pflotran.org/documentation/). Nutrient transport and reactions in SWAT are solved sequentially. We modified the explicit time-stepping algorithm in the original code for instream chemistry so the resulting nonlinear system of equations from the transformations taking place within the stream water and storage zones are simulated simultaneously with the implicit time stepping through the Newton Raphson method in batch mode (i.e., no transport) of the PFLOTRAN (Lichtner et al., 2017) model.

Response: Revised as suggested.

6. Line 223: Replace "[L] is the elevation of the free surface elevation at" with "[L] is the free surface elevation at"

Response: Replaced.

7. Line 277- 279: For the seasonal flow scenario, the application of NEXSS is clearly defined. For the BASE and MRMT is seems a bit ambiguous. Is it annually averaged over the same period? Maybe I missed the it earlier in the paper, but it would be useful to make a clear distinction at the same point in the manuscript.

Response: BASE case does not use NEXSS. We clarified the application of NEXSS for MRMT as "The mass transfer parameters were estimated by NEXSS using the long-term average flow conditions." at the same point.

8. For Figure 4 which compares the BASE scenario to the MRMT scenario, it would be useful to add markers to one of the traces so that it would be visually obvious that the two traces follow essentially the same path.

Response: BASE case line width in Fig4a is increased for a better visual effect.