

Dear reviewers and editor,

please check our responses to your constructive comments (marked in blue). Unless specified, all the line numbers in our responses refer to the line numbers of the TRACK CHANGE version of the manuscript. Thank you!

Response overview:

We have made updates to the manuscript, main updates are: (i) more details about calibration were added, and (ii) updating the introduction by including more literatures. We would like to acknowledge the efforts from the editor and the reviewers for the MS.

RC1:

The main goal of this paper, a resubmission of a manuscript I have already reviewed, is to investigate the impacts of topographic slope on subsurface flow, water age, and nitrate export at the catchment scale. Compared to the previous version, the paper has been significantly improved and all my major comments have been satisfactorily addressed. One minor request that I would make is to list the specific ranges for those zonal values in Table 1.

Response #1:

Thanks for the positive comment. We updated the Table 1 by including the two ranges for the zonal hydraulic conductivity and porosity values, respectively (line 223).

RC2:

I have appreciated the changes made by the authors to the Ms. in response to the comments of the referees. I think the paper is now improved as compared to the original submission, and the scope is certainly broader owing to the addition of observational data. However, the broader scope implies more issues to face. In particular, I suggest to provide more context / justification / details about the calibration procedure - the range of values for each model parameter explored (prior distribution), the possible definitions of the objective functions (e.g. using only Q data for the calibration of the hydrologic params, and then use C data for reactive transport params vs. all params being calibrated at once with a global obj function related to C and Q), the choice of fixed params vs calibrated params, the ensuing posterior distributions, the potential uncertainty. While I understand a full uncertainty analysis could be unfeasible in this case, the impact of operational choices done by the authors in their calibration exercise need to be better assessed/discussed. Moreover, the literature about nitrogen modeling in soils is huge, and more refs could be added to the Ms. Overall I congratulate the authors for their efforts.

Response #2:

Thanks for the suggestions. We added more details to the calibration procedure, and

discussed several impacts of the calibration exercise, accordingly. The main updates are:

- We clarified how the objective function in this study was defined using the data sets for transport as “PEST uses the Marquardt method [Marquardt, 1963] to minimize a target function by varying the values of a given set of parameters until the optimization criterion is reached. We used the measured C_Q and N surplus as the target variables for comparison with the simulated ones. The N surplus, which is the annual amount of N remaining in the soil after consumption by plant-uptake, was estimated as $48.8 \text{ kg ha}^{-1} \text{ yr}^{-1}$ [Yang *et al.*, 2021). As two different data sets (C_Q and N Surplus) were used, a weighting scheme was used such that the defined multi-objective function was not dominated by one data set” (lines 304 – 309).
- We clarified that our calibration actually followed a procedure of two steps: first for flow, and second for transport. The potential effect of choosing this two-steps procedure instead of calibration all at once was also mentioned and discussed. These clarification was done by adding “Note that the entire model calibration (for flow and transport) actually followed a procedure of two steps: first for flow, and second for transport. Alternatively, the flow and transport parameters can be calibrated at one step by defining the multi-objective function using all the data sets (discharge, groundwater levels, C_Q and N surplus). The potential effect of the two different calibration procedures on the modeling results should be further explored, however, being out of the main focus of this study. We consider the two-step calibration procedure to be acceptable, because our result showed that it was sufficient to reach an acceptable model performance for both flow and transport (described later)” (lines 310 -316). We also added “As the flow parameters (e.g., hydraulic conductivity and porosity) were already calibrated in Yang *et al.* [2018] using data sets of discharge and groundwater levels. In this study, the calibration was only performed for the transport...” (lines 301-302) at the beginning such that the readers can be clearer with the calibrations.
- We clarified that several parameters were fixed and others were adjustable for calibration by adding “Several transport parameters were fixed at the values selected according to prior information, such that the degree of freedom in the calibration can be reduced as much as possible (Table 2). In total eight parameters were adjustable and calibrated, because they were the key parameters to determine the N fluxes in soil and groundwater” (lines 317-319).
- The ranges of the adjustable model calibrations were listed in table 2 and described in text as “Their adjustable ranges were selected according to the literature or to cover the values that the parameters can realistically reach (Table 2)” (lines 319 -321).
- We emphasized the potential model uncertainty in the simulated N loads and fluxes in the discussion section 4.5 as “While the numerical model provided general insights, there was potential uncertainty in the simulated results. Firstly, the aforementioned simplifications may introduce model structural errors. Secondly, the model calibration was only constrained by limited data sets, which may lead to the non-uniqueness in the model parameters. Both of the aspects may introduce uncertainty in the simulated N loads and fluxes. Future work should be devoted to better constrain the model parameters, either by enhancing the concentration data quality through more frequent measurements or by providing additional data sets related to the N pool” (lines 628-633).

Response #3:

Thanks for the pointing that out. We added more literatures studding the nitrogen dynamics in catchments to the introduction, as “A number studies focused on numerically simulating the nitrogen fluxes (or loads) in soil and groundwater [*Smith et al.*, 2004; *Rivett et al.*, 2008; *Lindström et al.*, 2010; *van der Velde et al.*, 2012; *Van Meter et al.*, 2017; *X. Yang et al.*, 2018, 2019; *Kolbe et al.*, 2019; *Knoll et al.*, 2020; *Nguyen et al.*, 2021, 2022]. For example, *van der Velde et al.* [2012] constructed a lumped numerical nitrate transport model for the Hupsel Brook catchment in the Netherlands. *Lindström et al.* [2010] developed HYPE water quality model allowing for simulating the nitrogen fluxes in soil. *Van Meter et al.* [2017] investigated the two-centuries nitrogen dynamics in the Mississippi and Susquehanna River Basins using a TTD (transient time distribution) based transport approach. *X. Yang et al.* [2018] developed the coupled mHM-Nitrate model, which can provide valuable insights int the spatial variability of water and nitrate fluxes in catchment scale. *Nguyen et al.* [2021] further updated that model to the mHM-SAS model by implementing the SAS-function based solute transport module [*Harman*, 2015, 2019; *Rinaldo et al.*, 2015; *van der Velde et al.*, 2012], allowing for simulating the nitrate export from a Mesoscale Catchment. However, most of these works provided little information on the spatially-explicit details (such as the flow field) for interpreting the nitrate dynamics” (**lines 91 -104**).

The reference list was updated accordingly.