Comments to the first revision of “Nutrient transport and transformation in macrotidal estuaries of the French Atlantic coast: a modelling approach using C-GEM” by Xi Wei et al.

In my previous review my main criticism was that the manuscript does not provide a description of the simulated processes that may give readers a mechanistic understanding of the biogeochemical dynamics in the studied estuaries. The authors responded by adding texts about which processes were or were not incorporated, adding the reaction network in the supplement, and elaborating on the numerics in the response to reviewers. However, this only describes the methods that are used. The results and discussion still do not elaborate on the rates of processes, which are resolved by the model.

We are thankful to the reviewer for acknowledging the additions and answers we provided in the previous round of reviews. Based on this second evaluation, we now understand that the reviewer was not actually asking for further explanations and description regarding the model implementation and formulation (which was actually what the other two reviewers were requesting) but rather a more in-depth analysis and discussion of the biogeochemical process simulated by our model and a quantification of those biogeochemical transformations. We certainly understand the interest of such discussion and provide in the updated manuscript a several additions to strengthen that aspect of our study. We would also like to point out that we did already provide quantifications of several biogeochemical processes in a new table during the previous revision but we acknowledge that we did not discuss these values in the manuscript.

In the following document, you will find point by point answers to comments of the reviewers and several additions to the manuscript and supplementary material that, hopefully, addresses the desire of the reviewer for more mechanistic analysis of our model results. The key additions to the manuscript include:

- A totally new section in the results entirely devoted to the quantification of the main biogeochemical transformation occurring in the studied estuaries.
- An updated of table S2 of the supplementary material following which is now better connected to the main manuscript as it supports the new section mentioned in the previous point
- Some simulations performed without the biogeochemical module have been carried out, following a reviewer’s suggestion and a new figure was added to the supplementary material which compares mixing curves for TOC and NO3 between simulations ignoring or accounting for biogeochemical transformations.
- Time series for upstream and downstream boundary conditions for the main biogeochemical state variables as well as the fresh water discharge are now provided for all systems in order to provide a better feel for the magnitudes and characteristic time-scales of the variations of the conditions at both edges of the estuarine systems.
- The discussion of the limitations of the model and, in particular, the absence of explicit benthic module has been improved following the reviewer’s remarks.

However, we felt like some of the reviewer’s requests and suggestions, while certainly interesting, were getting past the scope of our study.

I tried to give suggestions to elaborate on the biogeochemistry. One suggestion was to conduct a sensitivity analysis. The authors refer for this to another paper (Volta et al., 2014) where the same model was applied, but as the forcing through the boundary conditions and the geometries are different, the response to changing parameters in a non-linear model is likely also different. I also suggested a simulation with the biogeochemistry turned off, as this would allow the authors to visualize in Fig 4 to what extent temporal variations driven by changing boundary conditions are modulated by the internal biogeochemistry. The boundary conditions at the ocean and the river side are constantly changing. This forcing based on data extracted from ECO-MARS3D, REPHY, and NAIADES is not shown in the manuscript. The manuscript only presents a table with the annual mean values for boundary conditions (Table 4), but it does not show the temporal variability (e.g. tides and seasons). This additional simulation without biogeochemistry may also allow for a better assessment of the model validation when data is only available near the boundaries (see comments RC1.9, RC1.10, RC1.12). The authors may still want to consider other ways to visualize the biogeochemistry that takes place within the model; for instance, showing reaction rates, presenting mass balances with the effect various processes on nutrient concentrations, etc.

We acknowledge that the reviewer provided a number of suggestions to investigate the internal biogeochemical processing of carbon and nutrient within the simulated estuaries. One of the suggestions was to perform a sensitivity analysis. Here, the reviewer suggests that this sensitivity analysis could be carried out by varying the boundary conditions applied to the different systems as it would allow assessing the effects of changing upstream and downstream constraints on the internal biogeochemical processing within the estuary. While we cannot deny that such analysis would certainly be of interest, we feel like it could also constitute an entire study in itself. We agree however that the previous version of the manuscript only provided annually averaged values for the boundary conditions used to constrain our simulation and this was not enough for the dedicated reader to evaluate the effect of, for example, varying river discharges on the biogeochemical dynamics of the model.

We thus created a large new figure with time series for the concentrations of the main biogeochemical state variables applied at the upstream and downstream boundary conditions for all systems. This figure was added to the supplementary material and can also be found at the end of this answer. We believe this will be very useful for the reader to evaluate the effect of the seasonal variations in fresh water discharges and nutrients concentrations on carbon and nutrients dynamics summarized on figures 3 and 4 of the main manuscript and completed by figures S1-S3 of the supplementary material. Indeed, we provided several longitudinal profiles and time-series for all the main biogeochemical variables simulated by the model and we believe that the (now possible) comparison between the temporal evolution of time-series extracted within the model domain with the seasonal variations of the upstream boundary condition will provide an insight to the reader regarding the biogeochemical dynamics within the estuary.

We would also like to point out that our study purposely investigates very different systems characterized by very different upstream constrains, residence times and geometries. While this does not in itself qualify as a sensitivity analysis, this strategy was nonetheless designed to assess the effect of these differences in boundary conditions on the resulting fate of carbon and nitrogen within the simulated estuaries.

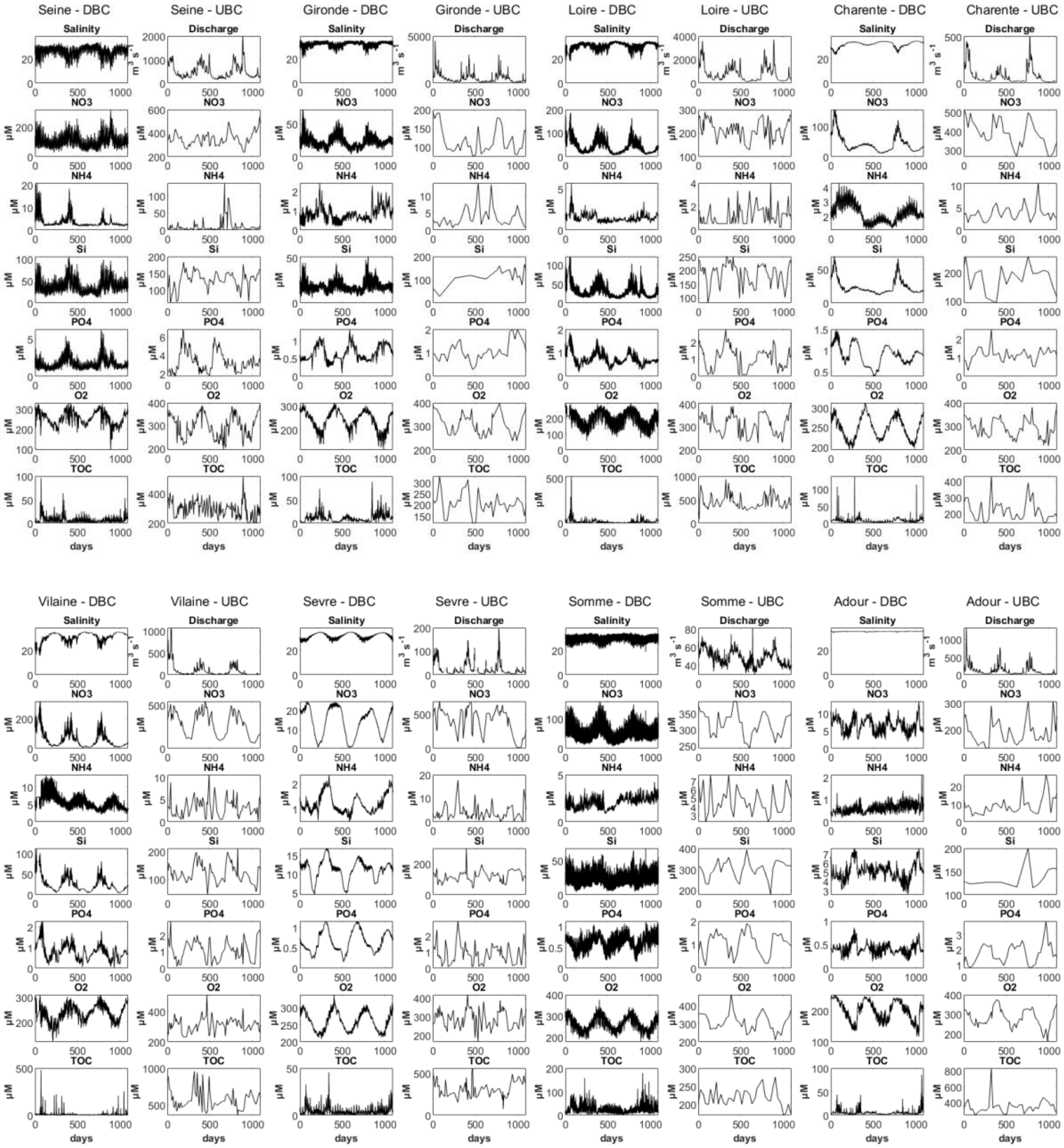

Figure S-4 Temporal variations for salinity, fresh water discharge, nitrate (NO3), ammonium (NH4), dissolved silica (Si), phosphate (PO4), dissolved oxygen (DO), and total organic carbon (TOC) concentrations from 2014 to 2016 at the downstream (DBC) and upstream boundary conditions (UBC) for all the estuarine systems simulated in this study. Salinity is only provided at the DBC while fresh water discharge is only provided for the UBC.

AC3.1 does not address my comment, which was about showing reaction rates and elaborating on the modeled rates. It was not about which processes are included in the model or the kinetic rate expressions.

We believe we now better understand what the reviewer was really asking for during the previous round of reviews. An entire new section was thus added to the results with a focus on the description and quantification of several biogeochemical processes (TOC degradation, denitrification, Si uptake) in order to better report the internal biogeochemical transformations simulated by our model. The new section is also supported by a table summarizing these fluxes and reporting them to the riverine loads in order to better assess the respective magnitudes of the lateral transport and the biogeochemical transformations within the system.

'The reaction process rates calculated by the model along the estuaries over the 3 years were integrated and provided as annual average, in ton of total organic Carbon, N-NO3, dissolved Si or phosphate per year for organic matter degradation, denitrification, net primary production either from diatoms or the from all the algal community, respectively (see Table S-1 in supplementary material). These fluxes are highest in the largest estuaries with the largest carbon and nutrients loads as well as the highest residence time. Reported in terms of percentage of the import fluxes, these values allow comparing the intensity of the biogeochemical processing simulated by the model between the different estuaries as well as with carbon and nutrients retention rates discussed above. Overall, Total organic carbon degradation and NO3 denitrification percentages were the highest in the Gironde (98 % and 26%, respectively) and the lowest for the Vilaine (10%, 3%, respectively). Other systems displayed intermediate values falling in the 24-39% and 3-14% ranges for TOC degradation and denitrification, respectively (Table S-1). This intensity of organic carbon processing is consistent with the global figures suggested at the global scale by Bauer et al. (2013) as well as previous modeling studies performed with C-GEM in Europe and along the East coast of the US (Volta et al., 2016, Laruelle et al., 2017, 2019). The integrated denitrification rates are also consistent with the compilation performed by Nixon et al (1996). Biogeochemical reactivity regarding organic matter degradation and denitrification appeared greater for the most retentive Gironde, with its longest residence time. This trend was also illustrated by the calculation of mixing curves for the Seine, Loire and the Gironde (the 3 systems with the longest salinity gradient) computed for the reference simulation and simulations in which the biogeochemical model of C-GEM was deactivated (Figure S2 of the supplementary material). The difference between the refence simulation and the simulation without biogeochemistry provides a visual representation of the intensity of the biogeochemical processing within the system and is significantly more pronounced in the Gironde. Interestingly, silica uptake percentages, which are entirely sustained by diatoms primary production were also the highest in the Gironde, the Seine and the Somme (16%, 13% and 12%, respectively), in accordance with the ones for phosphates (16%, 43%, 76%, respectively). These results revealed however a large range of autotrophic activity. The percentage PO4 uptake flux was particularly high in the Somme because of its proportionally low specific P riverine loads from its upstream boundary. Overall biogeochemical phosphates fluxes were rather well balanced with those of silica, according to the Redfield ratios, indicating mostly a development of diatoms (Table S-1).'

AC3.2 does not address my comment. Here I asked for a sensitivity analysis of the model outcomes with regard to the biogeochemistry occurring within the model domain. It was not about the numerical implementation.

Again, we acknowledge that we did not initially understand what the reviewer was expecting but we believe the that new section of text presented above now addresses the desire of the reviewer for a better description of the inner biogeochemical dynamics of the model. We also note that the reviewer suggested to perform a fully blown sensitivity analysis of the model and we already justified why we feel like such an extensive analysis was not justified to and falls outside of the scope of this particular paper. We did however follow the suggestion of the reviewer to perform simulations in which the biogeochemical module of our model was deactivated. These simulations allowed us calculating mixing curves for TOC and NO3 for the Seine, Loire and the Gironde (the 3 systems with the longest salinity gradient) which were added to the supplementary material and referred to in the manuscript.

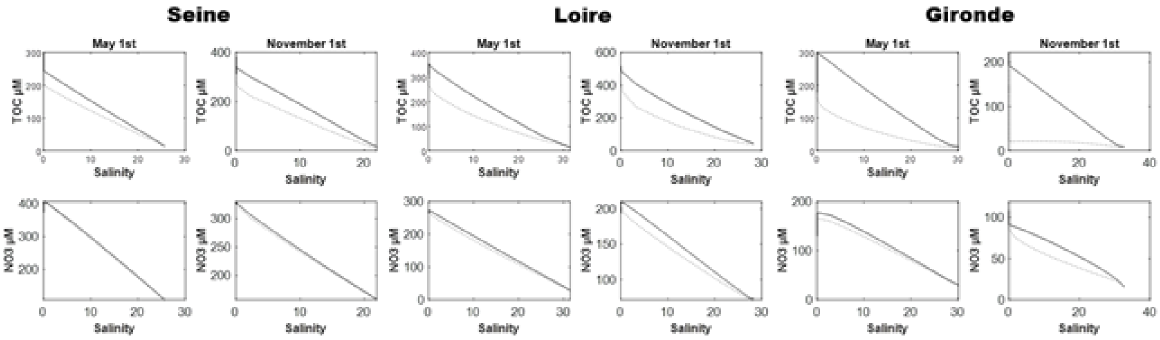

Figure S-5 Mixing curves extracted from the model outputs for total organic carbon (TOC) and nitrate (NO3) along the Seine, Loire and Gironde estuaries on May 1st 2014 and November 1st 2014. Continuous lines correspond to simulations performed without the biogeochemical module of C-GEM while dashed lines correspond to the reference simulations.

AC3.3 and RC/AC3.9:

The text at lines 514-524 does not provide a compelling argument not to include a benthic module.

Following the reviewer’s comment, we modified this section of the discussion in the updated manuscript and tried to better express the fact that we do not argue that benthic processes should be ignored altogether but, rather, that the model set-up used for our study was first step (with a first order representation of burial) towards better representation of carbon and nutrient removal within estuaries, starting from generic model without any interaction with the sediments. The updated text now reads as follows:

‘*With its current set-up, the lack of explicit benthic biogeochemical module obviously limits the depth of mechanistic understanding the model can provide of nutrient cycling, particularly regarding interactions between pelagic and benthic compartments which can significantly influence the intensity but also the timing of nutrients and organic matter cycling in estuaries (Laruelle et al., 2019). In that context, future developments of the model should include the implementation of several benthic processes this task comes with a number of hurdles. For instance, while the addition of a full diagenetic module at each grid cell of our model would be possible, it would also increase its calculation time by one order of magnitude and likely require a very long spin-up to generate initial conditions for the benthic species. There exist simpler benthic modules of lower complexity, which would limit the computation cost of adding an explicit representation of benthic processes (Billen et al., 2015; Soetaert et al., 2006) but those would nonetheless significantly increase the data demand of the model to be properly calibrated. Indeed, the increase in complexity of the model will involve the use of field data to constrain and calibrate the newly implemented processed. While measurements of estuarine benthic processing of nutrients and carbon do exist, they are still relatively scarce. In the present study, the simple representation of particulate matter burial that was implemented and applied to phytoplankton and TOC to provide a first-order representation of the process, which is necessary to evaluate the retention of carbon and nutrients within the system. We believe this addition, coupled with denitrification provides a first insight on the main pathways removing nutrients from estuaries and allows calculating carbon and nutrient retention rates that can be compared with previously published estimates.’*

If including benthic processes leads to more uncertainty (lines 519-520) then ignoring these processes cannot make the model outcomes more reliable. Letting water-column processes account for sedimentary processes makes no sense. For instance, as the water-column is fully oxygenated it will not allow for denitrification and cannot compensate for missing benthic denitrification.

We feel like the comment of the reviewer ('…benthic processes leads to more uncertainty…') does not accurately reflect what we were trying to express in lines 519-520 but acknowledge that our text might not have been clear enough. Our point was that an increase of the complexity of the model associated to the implementation of new fluxes requires new data for the calibration of the model. In the case of estuarine benthic processes, we do not deny that such data exist but they remain quite scarce. In any case, the sentence singled out by the reviewer was removed.

Moreover, while we understand and agree with the remark of the reviewer that 'Letting water-column processes account for sedimentary processes…' is not an ideal solution, we would also like to point out that the practice of simplifying the conceptual representation of nitrogen processing within estuaries (or rivers, lakes or reservoirs) is not new and can be found, for instance, in simple mechanistic models such as those of Maavara et al. (2019). Again, these approaches do not accurately represent benthic dynamics but are a compromise and an intermediate step between totally ignoring the benthos and using a model too complex to be adequately calibrated with available data.

Maavara, T, Lauerwald, R, Laruelle, GG, et al. Nitrous oxide emissions from inland waters: Are IPCC estimates too high? Glob Change Biol. 2019; 25: 473– 448. https://doi.org/10.1111/gcb.14504

If the authors do not want to include a benthic module for this paper, they should try to assess the effect of omitting benthic processes. Typical rates for benthic processes may be taken from literature and be compared to simulated and measured rates in the water column. The importance of benthic reactions can also be estimated by calculating Damköhler numbers.

We understand the point the reviewer is trying to make and we believe that our discussion of the current level of complexity of the reaction network of our biogeochemical model demonstrates our willingness to openly discuss the limitations of our approach. However, we are not convinced that assessing or quantifying the effect of omitting benthic processes on the results of our simulations can be easily performed using any of the method suggested by the reviewer. While there indeed exist observation and model-derived estimates for benthic process rates such as denitrification, remineralization or burial in some well-studied estuaries, those are typically very site-specific and widely vary from a system to the next. As a consequence, evaluating the potential magnitude of the 'omitted' processes on the basis of such uncertain data would likely yield fluxes with very larges uncertainties resulting in limited insight. Bearing in mind the limits of our approach, we feel like comparing the carbon and nutrient retention rates simulated by our model with previously published literature (as we did in our discussion) and evaluating our model outputs against available water-column measurements is arguably a more concrete (yet indirect) way of evaluating the potential effect of missing benthic processes on our simulations.

AC3.8: The abstract states that one of the goals of the paper is to constrain chemical budgets, not the comparison of models or model development. So is it not better to just increase the grid resolution?

In C-GEM, changing the spatial resolution of the grid is not as trivial a task as the reviewer seems to suggest and we still believe that the implementation of such modification would only have added little insight regarding the accuracy of our simulations considering the very limited amount of data available for the smallest estuaries (Somme and Vilaine in particular). Besides, in spite of what the reviewer seems to understand from reading our abstract, one of our goals was indeed apply the same model set-up to several estuarine systems along the French Atlantic coast and take full advantage of the genericity of C-GEM with as few as possible modifications from site to site (besides forcings and boundary conditions). To take into account the reviewer's remark and better convey this objective of our work, the following statement was added at the end of the abstract: *'This study also demonstrates the ability of our model to be applied with a similar set-up to several estuarine systems characterized by different size, geometries and riverine loads.'*