

Response to Review#1

General Comments

This paper assesses the effect of riverine nutrient and C inputs on C cycling in global oceans, and notably on CO₂ outgassing in a pre-industrial state. The work includes quantification of riverine inputs of different elements under inorganic and organic forms, regional coastal shelves analysis, and global ocean biogeochemical modelling. Finally, the authors derive a global land-ocean-atmosphere pre-industrial budget, and discuss the effect of riverine inputs and gaps that need to be further addressed in the future (e.g., including volcanic emissions and the effect of shale organic oxidation). I believe this is a substantial piece of work, improving the current understanding of global C cycling, and totally fitting Biogeosciences' scope.

Response: We would like to thank the reviewer for the comments and very helpful contributions to improving the manuscript.

Major Comments

1) In general, the paper is extremely dense and would benefit from being shortened and making it more to the point. Major conclusions should be better highlighted, and repetitions avoided. Also, consider deleting sections with discussions out of the main scope of the study and already addressed in previous work (e.g., on weathering). A few suggestions to improve this point are also listed hereafter, in the specific comments.

Response: We think that the suggestion to streamline and shorten the manuscript is a good point that has been mentioned by both reviewers, and we will therefore consider this for the revised manuscript.

We suggest a revised manuscript with a shortening of the introduction (section 1), the elimination of repetitions in the methods section (section 2), and the merging and shortening of sections 3. and 4.

2) Further details should be provided on the construction of 2 ocean simulations, RIV and REF. The understanding of the differences between these 2 simulations is key to understand the paper's major conclusions.

Response: While this is partly done in paragraphs in the methods (section 2.2.3) and in the results and discussion (section 5.1), we also see the need to further highlight the main differences between REF and RIV, and discuss the analysis strategy. These are two things: Firstly, the geographical differences of inputs to the ocean (riverine inputs vs inputs to the open ocean), and secondly the increased carbon inputs to the ocean due to the transfer of carbon from land to ocean.

We will more clearly state these points in the revised manuscript. We also believe that the improvements from comment 1) will help improve the overview of these differences in the revised manuscript.

Minor comments

L7-8p1. “Thirdly, we quantify the terrestrial origins. . . in the framework, . . .” -> It is not clear from this sentence if this is purely a modelling exercise of if you are assessing global oceanic C budgets using the coupled land-atmosphere-ocean model.

Response: Here, we solely budget for the carbon fluxes from the “stand-alone” models, which are forced with the same pre-industrial atmospheric state. The models do not however give a feedback, which alters the atmospheric state, since the atmospheric CO₂ concentration would decline due to the carbon imbalance of the atmosphere (Figure 11). On the terrestrial side, the carbon fluxes are derived from the weathering and the organic carbon export models. On the ocean side, they are derived from the the ocean biogeochemistry model HAMOCC. We therefore do not take into account feedbacks in the system, as would be done in a fully coupled land-atmosphere-ocean model, but if so, one would need to consider further carbon outgassing sources (long-term volcanic emissions, shale oxidation), in order to have a balanced budget for the pre-industrial atmosphere.

We will clarify this sentence in the revised manuscript.

L11p1. “leads to a global oceanic source of CO₂” -> “leads a net global CO₂ emission to the atmosphere of”. To be consistent with the following sentence, the “source” would be 183+128 TgC/yr (Fig. 11).

Response: We will implement this correction in the revised manuscript.

L14p1. It is not clear what a sink due to a model drift is without reading the whole paper in depth. . .

Response: This is a good point, and we think that the word “model disequilibrium” is more understandable for the reader.

Introduction. The introduction provides a lot of information, some of it not totally relevant to the focus of the study. It should be shortened and re-organized to better highlight current state of knowledge, gaps and how they are addressed in this work.

Response: We will improve the structure and points of focus of the introduction in the revised manuscript. We will shorten the paragraphs p2.l4-p2.l18 and p3.l7-p3.l28 and increase the focus of the paragraphs.

L15p2. “these knowledge gaps” -> all knowledge gaps are presented in paragraph 7.

Response: We agree that this part of the sentence might be redundant and therefore will remove it.

Paragraphs 2-6. These present in detail processing in watersheds, gaps in knowledge on tDOM and POM degradation and transfers, etc. These could be substantially shortened, since these are not points tackled in the paper, which waters down the main message/focus of the paper. Parts of it (e.g., uncertainties on degradation of different OM forms, desorption of P as it enters saline waters etc.) could however be used in the Discussion (e.g., subsection 7.2).

Response: We understand the point of view of the reviewer, and also think the paragraphs can be shortened. However, some of the information within these paragraphs is also vital in order to understand the assumptions chosen later in the methods section. For instance, due to the previous degradation of tDOM within rivers, we assume it is less reactive once it reaches the ocean than oceanic DOM.

In the revised manuscript, we will shorten these paragraphs and increase their focus.

L2p5. "Pre-industrial" could be defined here (state in 1850).

Response: We will define pre-industrial here in the revised manuscript.

L5p5. "spatially explicit quantification of global riverine loads" -> "spatially explicit quantification of riverine exports to global coasts". It is not clear otherwise if the loads within watersheds are quantified as well.

Response: The loads are indeed quantified for every watershed. We agree this might be somewhat unclear here and will correct this in the revised manuscript.

L10p5. "We first briefly..." -> Description of the Method's content is already described in the previous paragraph. These introductory sentences could be most of the time removed to make the paper shorter and more to the point.

Response: We will remove this sentence since it is a repetition.

L12p5. Define what you consider like alkalinity here.

Response: Alkalinity (which is carbonate alkalinity in the case of our derived riverine loads) will be more clearly defined here in the revised manuscript.

L20-32p5. This is described in detail in the subsections; it could be deleted to avoid repetitions.

Response: We believe it is important to give a brief overview of all derived loads before addressing the individual elements. Otherwise we fear the reader will be missing the "big picture" if we start with individual details of every compound load. We will however remove some parts from this paragraph, which might be unnecessarily repeating information in the revised manuscript.

Fig.1. Precise that the C from weathering sources is DIC (OC assumed to originate only from the uptake of atmospheric CO₂).

Response: This will be added to the figure in the revised manuscript.

L12-14p7. Doesn't river damming affect POM loads between pre-industrial and the 1970s? This point could be considered later in the discussion on river loads.

Response: We assume in the whole study that the organic matter loads have remained constant globally since the pre-industrial time period. While increased organic matter supplies to catchments have been suggested in literature (for instance in Regnier et al. 2013), increased retention and remineralization (for instance due to damming, Maavara et al., 2017) has also been reported. There is however yet a study to spatially quantify these combined effects, while comparing the pre-industrial time-frame to the present day. This is briefly already discussed in 4.1. (125p20.), and we believe a more comprehensive discussion of these combined anthropogenic influences escape the scope of our study.

L13p8. "Pre-industrial runoff..." -> this is already explained L1-5p7, not necessary. This repetition occurs several times throughout the paper.

Response: We will remove this repetition in the revised manuscript.

L24p8. Are the soil types listed here those with typically low erosion rates? Is the 0.1 factor also used for wetlands and areas with a high groundwater table? If yes, what definition did you use for "high water table"?

Response: The soil types listed here are indeed the ones assumed to have very low weathering rates. The wetland areas are the Gleysols. These (along with the other soil types) are defined in the Harmonized World Soil Database (Fao et al., 2009). The description found in the database is that gleysols are soils with permanent or temporary wetness near the surface. The derivation of the soil shielding factor is extensively described in Hartmann et al. (2014). In our opinion, further details escape the scope of our study.

L4p9. What do the 1.6 TgP/yr used here correspond to? Fertilizer P in surface runoff reaching rivers? Hart et al. (2004) report an annual fertilizer consumption of 873 TgP in 1913.

Response: This number was wrongly cited. The 1.6 Tg P to catchments was derived from Figure 3 in Beusen et al. (2016), which on its side derived from inputs described in Hart et al. (2004), as is cited in the corresponding technical manuscript (Beusen et al., 2015). Assuming 873 million tonnes of P inputs of fertilizer for the year 1913 (Hart et al., 2004), this relatively high value seems plausible.

L7-9p9. Why is it reasonable to assume P equilibrium in soils at pre-industrial state (besides that state-of-the-art models usually use this initialization)?

Response: We agree this assumption is a limitation of our study. For instance, Filipelli et al. (2008) suggest an increase of P inputs to catchments due to increased soil erosion caused by deforestation and land use change. This likely already had an impact on land-ocean fluxes prior to industrialization, although it is completely unknown how large these fluxes could be. This point goes hand-in-hand with our assumption that the organic matter fluxes remain near constant, which is at least consistent. In the revised manuscript, we will add this point to 7.1 when discussing limitations and aspects that could be potentially improved.

L9-11p9. The assumption that spatial distribution of P river loads is the same in pre-industrial and 1970 is quite strong. For example, agriculture was probably much more developed in North America and Western Europe than in Asia at the beginning of the 20th century. Implications of this assumption should be discussed later in the discussion on river loads. Why not use load distribution from models describing earlier states (e.g., Beusen et al., 2016)?

Response: This is a good point, we however are not aware of available data from which such a differing distribution of the anthropogenic inputs from the present-day could be constructed. It is also not clear from the Beusen et al. (2016) study if and how the study assumes differing anthropogenic input distributions to the present day. We however suggest shortly investigating the differences between the approach chosen here and the approach chosen in Beusen et al. (2016) in a supplementary information section, if this data is available. We will also discuss this uncertainty in the discussion section 7.1.

L23-24p9. Molar C:P ratios are already provided earlier. You can just say L19 that P is incorporated in organic matter accordingly to C:P molar ratios for tDOM and POM.

Response: We will correct this in the revised manuscript to avoid repetition.

L6-12p10. Precise here that DIN export was calculated by subtracting the part contained in organic matter. Was DFe export equal to Fe inputs to catchments?

Response: The Fe composition of the organic matter is also subtracted from the Fe inputs to the catchments, which results in DFe. We will clarify this in the revised manuscript.

L11-16p11. This is not totally clear. Do you assume that only HCO₃⁻ affects alkalinity and that DIC is only HCO₃⁻ as well (Alk:DIC = 1:1)?

Response: We assumed that only HCO₃⁻ release from weathering affect the alkalinity and that the Alk:DIC ratio is 1:1. This is stated and justified in the paragraph, but we will attempt to make the paragraph more understandable.

Sub-subsection 2.1.5 Silica. Why not use the same weathering model type as for P (Hartmann et al., 2011)?

Response: In theory, this would be possible, but the model would first need to be calibrated for Si. Since the Beusen et al. (2009) model is already calibrated for Si, we used this one out of simplicity.

Sub-subsection 2.2.1 Ocean biogeochemistry. A scheme with model state variables and processes would be helpful here (as well as a description of biogeochemical processing equations and parameters as Supplementary Information).

Response: We will add a scheme which illustrates the main processes represented in HAMOCC in the revised manuscript. Regarding the equations, this is too extensive in the case of this manuscript. We refer to Ilyina et al. (2013) for the model description, equations and parameters, with more recent model developments described in Mauritsen et al. (2019). If necessary, we can provide a list of changed parameters in comparison to the Ilyina et al. (2013) study in a Supplementary Information document.

L22p13. How did you choose the 0.003 d⁻¹ value for tDOM degradation? 0.008 d⁻¹ for the oceanic DOM is also in the literature range for tDOM degradation.

Response: The assumption chosen here was that the tDOM entering the ocean at the river mouths is less reactive than the “freshly produced” oceanic DOM, since tDOM is reported to have been strongly degraded previously during its transport in rivers. This is already stated in paragraph p13.l20-p13.l24.

2.2.3 Pre-industrial ocean biogeochemistry model simulations. Please provide more details here, since the distinction between the REF and RIV simulations is key to understanding the paper's results. In the REF simulation, are input fluxes (per surface area?) globally homogeneous? Or do they compensate sediment losses to reach equilibrium locally (at the cell scale)? In the RIV simulation, are there also open ocean surface inputs in addition to river inputs?

Response: In short, the REF inputs are added globally homogeneously (per surface area) to the ocean surface. This represents oceanic inputs by (to a major part) passing the coastal ocean and does not at all represent geographic riverine inputs. Their magnitudes are dictated by losses to the sediment: every few hundreds of years, the averaged global burial loss of biogeochemical compounds was computed, and these were the fluxes added to the surface ocean. They are

indeed homogeneous per surface area. In the RIV simulation, there is no open ocean surface inputs, since these were replaced by riverine inputs, derived as shown in the previous sections of the manuscript.

We will discuss and clarify these points extensively in the revised manuscript.

L1-6p14. What do you call quasi-equilibrium? How were the lengths of the different simulation chosen? Why did you perform a succession of 3 runs for the RIV simulation? Does “standard simulation” (L4) refer to REF?

Response: The lengths of the simulations were chosen accordingly to the model state, meaning the simulation were performed until no strong drifts of model variables was remaining. Since biogeochemical processes in the ocean, and especially in the sediment, likely need larger simulation time-periods to perfectly equilibrate than is feasibly possible with current computing resources, there will however always remain a small drift in model variables in such simulations. Therefore, this is a state of quasi-equilibrium and not a perfect equilibrium state.

The 3 sequential simulations for the RIV simulation were done in order to achieve a more stable state in the ocean sediment. The motivation here is that it is much less expensive in terms of computing power to simulate the sediment separately from the ocean, and since the time-scales of processes taking place in the sediment are much longer than in the water-column, it makes sense to perform simulations for the sediment alone once the oceanic water column has reached a state close to equilibrium. We therefore firstly performed a simulation of 4'000 years including both the water column and sediment biogeochemistry, until the global particulate fluxes from the water column to the sediment were approximately stable. Then simulations of the sediment component of the model, while being given the stable global particulate fluxes from the previous simulation. Finally, the sediment state was re-coupled to the ocean water-column for the final 2000 year simulation.

We believe the full description of the model spin-up mentioned above are too technical for the scope of this journal, and would substantially unnecessarily lengthen the manuscript.

Are the 100-year means (output results) calculated on the last 100 simulated years for each simulation? What are the simulation timesteps?

Response: The last 100 year mean of both REF and RIV were used to the analysis. Using a 100 year mean prior to this would not affect the results much. The simulation timestep is one hour.

It is mentioned that water loads vary inter-annually based on runoff inputs from OMIP. Are the ocean physics' inter-annual variations modelled with the same patterns for every simulated year?

Response: The parameters within the physical circulation model MPI-OM remain the same. We would like to point out that the model simulates the response of the ocean physics dynamically to the atmospheric state, from which the information is given to the model for every time-step. There is therefore inter-annual variation in the physical features of the ocean, and even variation at every model time-step.

It escapes the scope of our study to completely explain mechanisms of such a circulation model in the scope of our study, but this has been done before and we would like to refer to the cited references (Junclaus et al., 2013 and Mauritsen et al., 2019).

L20-24p16. For which periods were these Si loads from the literature estimated?

Response: These estimates are for the present day. We will mention this in the revised manuscript.

L26-32p16. Comparison with Mackenzie et al. (1998) does not seem necessary here, especially since they assess only the TIC load.

Response: We agree and will therefore remove this comparison in the revised manuscript.

Subsection 3.1 Runoff, precipitation and temperature patterns. Hydrology model performance is not the scope of the paper; this section seems to water down once more the message/goal of the study. The scaling of the MPI-ESM runoff should be detailed earlier, in the Methods section. How is the factor 1.59 chosen? Why is the runoff not scaled to the OMIP one, to be fully consistent with the freshwater inputs?

Response: In order to shorten the manuscript, we will merge sections 3 with section 4, while shortening both with only the most central information for the next parts of the manuscript remaining. Factor 1.59 was chosen in order to scale the average of Fekete et al. (2002) and Dai and Trenberth (2002) (l24.p15). In the revised manuscript, this will be more clearly stated.

Subsection 4.1 Global loads in the context of published estimates L2p19. Does the Fe-P load given in Table 3 only corresponds to the fraction that is desorbed when entering the estuary?

Response: The Fe-P loads refer to desorbed P loads both for the given modelled estimates (Model. Global load), as well as for the Compton et al. (2000) values. This will be stated in the Table 3 Caption in the revised manuscript.

Table 3p19. Precise for the POP comparison that the 5.9 value (for 1970) is for PP, and not only POP.

Response: This will be added in the revised manuscript.

L20-23p20. Doesn't the potential increase in Si retention at the end of the 20th century mainly concern particulate forms?

Response: This is not correct. Global reservoirs have also been suggested to retain a significant amount of DSi (Lauerwald et al, 2013; Maavara et al., 2014). This is due to in-reservoir formation of biogenic silica, which is then in turn also increasingly retained.

L20-26p21. Please better organize text.

Response: We will improve the organization of the text in the revised manuscript.

L4p22. Could the discrepancies also be due to the fact that weathering formalisms are less adapted to Arctic regions?

Response: This is indeed very likely, with weathering mechanisms in the Arctic being likely more complex than is modelled here (for instance due to permafrost, Hartmann et al., 2014).

5.1 Ocean state – An increased biogeochemical coastal sink L14-15p25. “There is a higher carbon load originating from organic matter, since tDOM C:P ratio is higher than the oceanic DOM C:P ratio” -> This explanation is not straight-forward, since the estimation of DOC is not related to P in the model.

Response: This is correct but the DOP is coupled to the DOC estimation. In (long-term) model equilibrium, for every mol P exported to the sediment, 122 mol C is also exported. For the river

inputs however, for every mol P 3583 mol C are added. Therefore there is an accumulation of DIC when tDOM is mineralized and POM/DOM is subsequently produced by the oceanic biology with the released DIP, which leads to higher pCO₂ and outgassing. This is explained from l20p42, but we will attempt add clarify to the statement on L14-15p25.

L15p25-L2p26. Comparison with other studies was already discussed earlier.

Response: The central point here is that the carbon inputs are increased in RIV comparison to REF., in agreement with previous estimates. This is why carbon outgassing takes place and therefore we believe this is central information here, even if it has been stated before.

L17-19p26. Isn't the effect of large oxygen minimum zones on DIN concentrations also visible in WOA dataset?

Response: This is indeed the case and we can mention this in the revised manuscript.

What explains the major differences in DIP, DIN and DSi concentrations in the Southern Ocean?

Response: This is likely due to the relatively poor representation of the complex ocean circulation of the Southern Ocean in global ocean circulation models. An improved representation would require high enough resolution to better represent vertical mixing taking place in the Southern Ocean and a better representation of the effects from storms.

Table 5.p27. Do N inputs include N₂ uptake from the atmosphere in the two simulations?

Response: Both simulations consider dynamical N₂ fixation by cyanobacteria as well as (natural) nitrogen deposition. This is stated in the methods p12.l11: "The changes were made to incorporate dynamical nitrogen fixation through cyanobacteria (Paulsen et al., 2017), .."

L12-14p28. Comparison of Arctic concentrations to the WOA database could be presented earlier in text, with the rest of the comparisons.

Response: We agree and will correct this in the revised manuscript.

7.1 Rivers in an Earth System Model setting. Paragraph 2 (L12-17p36). These improvements could further avoid strong assumptions, such as globally constant N:P ratios for river inputs, and on the spatial distribution of non-weathering sources.

Response: These are both very good points and we will add these to the subsection in the revised manuscript.

L11-14p37. Is the need for a runoff scaling factor a major conclusion of this work?

Response: Although this is a more technical conclusion, this is quite significant: in order to have dynamically changing riverine loads in a fully coupled (land-ocean-atmosphere) model, the runoff would first need to be addressed.

L17-21p37. "Even for present-day..." -> comparison with literature is already discussed earlier, this could be removed.

Response: We agree and the sentence will be removed in the revised manuscript.

L26-27p37. “which have been identified to be more strongly controlled by extreme hydrological events than other C species” -> Was this considered in the study? What does it imply? It seems that this belongs to some discussion (that should be more developed if added).

Response: The NEWS2 model results, which we derive POC exports from, calculate the year-means of these exports. Therefore, these extreme events are not at all grasped in the model. This will be shortly discussed additionally in the revised manuscript.

L17p38. “despite previously being sinks” -> be more explicit by explaining that this is without accounting for river inputs, and not “previously” in a temporal way.

Response: We will correct this statement in the revised manuscript in order to improve clarity.

L27-29p38. Interhemispheric C transfers are already mentioned earlier in this section (L1-3p38). Please focus in this last paragraph only on the major points that this study shows have to be included in ESMs to better assess land-ocean-atmosphere C transfers.

We will remove this second discussion interhemispheric C transfer and add an outlook in the conclusions.

References (excluding cited references in the manuscript):

Beusen, A. H. W. Et al.: Coupling global models for hydrology and nutrient loading to simulate nitrogen and phosphorus retention in surface water – description of IMAGE–GNM and analysis of performance, *Geosci. Model Dev.*, 8, 4045-4067, <https://doi.org/10.5194/gmd-8-4045-2015>, 2015.

Lauerwald et al.: Retention of dissolved silica within the fluvial system of the conterminous. *Biogeochemistry*, 112,637-659. 2013.