Manuscript revision (2nd round)

A fresh look at the pre-industrial air-sea carbon flux using the alkalinity budget

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We have carefully addressed the reviewers' comments in our revision and would like, once again, to express our gratitude to both reviewers for their careful and constructive evaluations. Over the course of these two rounds of review, their insights have greatly contributed to improving the quality and clarity of our work. Below, we provide a detailed point-by-point response to all reviewers' comments.

In addition, we have updated Fig. 4b, Fig. B2 and Fig. B3c using a colour palette from https://sronpersonalpages.nl/~pault/, to ensure that readers with colour vision deficiencies can correctly interpret our findings.

Reviewer #1

The authors have made a remarkable job revising their ms. (I enjoyed reading this new version !) and at this stage, I only have a few very minor comment (technical corrections):

- Table 1: If I recall well, the underlying assumption in Jacobson et al. (2007) was that the distribution of outgassing mirrored the regional inputs/outputs (no inter-hemispheric transport). Thus, why 'not applicable' ? Please check.

You are absolutely right. However, this Table is simply a summary regarding the GCB manuscripts. At the time (from GCB 2013 to GCB 2017), the distribution of the air-sea carbon outgassing from riverine/burial carbon and alkalinity fluxes was not used. In fact, there was no regional distribution of the ocean anthropogenic carbon flux shown back then. For clarity, we have changed '*Not applicable*' to '*Not considered at that time*' in the Table.

- Line 67: Fig1a should be 2a (please check references to figures throughout the text)

This has been corrected. References for the Figures and Tables have been checked throughout the manuscript.

- Figure 3 (caption): "and the colors of the arrows is consistent with what is used throughout the manuscript (e.g. Fig. 2)". Not sure to what this statement refers to.
Remove ?

This has been removed.

- Figure 3 (caption): "deviations" with respect to what ?

We have added two sentences in the first paragraph of the Methods to make it as clear as possible:

"We refer to a 'deviation' in a given variable when the system is in steady-state, but a persistent trend is identified for that variable (e.g. the global carbon inventory). In contrast, we use the term 'drift' when a trend in a variable disrupts the steady-state."

- Line 122: add "ocean" to "a non-conserved global Alk inventory"

This has been added.

- Line 186: explain in a few words (for the broad readership) what an "Alk restoring scheme" means

This has been added: *"periodically restoring the global Alk inventory to a reference value by adding/removing the required amount, either uniformly or in a weighted manner"*

- Line 200: Here (or somewhere else in 2.2.2 or 2.2.3 if you find it more appropriate), I suggest adding a reference to Liu et al. (Nature Geoscience, 2024) to highlight that your river input in your std simulation is on the low side.

Such a reference has been included in Appendix B2.1: *"It is worth noting that the global values and latitudinal distribution of riverine inputs are based on (Ludwig et al., 1996) and have recently been revised (Li et al., 2017; Liu et al., 2024) although the human imprint on these fluxes cannot be removed."*

- Lines 237&238: add "ocean" next to inventories ?
This has been added.

- Line 271-72: Should these numbers not be reported with a negative sign ? Add that the difference between 0.27 and 0.31 is due to the slight "residual component" ?
Since we are referring here to a net global outgassing (i.e. 'a net global outgassing of …'), we report the value without the negative sign. The positive or negative sign is used only when the values are presented in brackets (see previous paragraph in the manuscript).

Regarding the clarification of this residual component offset, we have decided not to include the reference at this point in the manuscript, as it is already explicitly mentioned at the end of the previous paragraph in the manuscript. Moreover, it is too early in the Results section to introduce the expected value of 0.27 PgC/yr.

- Line 277: Should be Fig.4d (not 2d)
This has been corrected.

- Section 3.2.3: Add a reference somewhere to fig.5c ?
This has been added.

Reviewer #2

In their revised manuscript, Planchat et al. strongly improved their manuscript, especially in their restructuring and improving the contexts and explanations of their results, and their work remains significant and a huge effort. Both introduced frameworks are much clearer, and I especially appreciate that the second theoretical framework was introduced into a new section, which helps differentiate the results from the simulations obtained directly from the model, to those derived from making offline corrections to the simulations through combination of sensitivity experiments. The introduction of the second framework is also mostly clear through the step-wise description of the approach. I would still have a few points to clarify, but would otherwise recommend the manuscript for publication.

Major Comments

- Limitations: The manuscript would, especially regarding the theoretical framework, benefit from a brief limitations paragraph. While the methods introduced are definitely interesting and computationally efficient, they are not correcting for all physical and biogeochemical biases of the model (I don't think it is really what the authors mean, but it is not really stated otherwise). While this will likely work for corrections of model river inputs and potentially burial fluxes, structural model errors present in all simulations, which affect the redistribution of the carbon in the ocean, will not be corrected from combining the sensitivity simulations together to fit (globally evaluated?) observational values.
Regarding the global pre-industrial riverine/burial-driven air–sea carbon flux, it can now be assessed without relying on models, based solely on theoretical considerations (see Sect. 4.1). The physical and biogeochemical biases of models have no impact on these results. What remains limiting is our knowledge of, and confidence in, the values of riverine and burial fluxes of carbon and Alk (see Sect. 4.1.3).

As for the spatial distribution of the flux (Sect. 4.2), we explicitly discuss the limitations associated with model use, particularly in the final paragraph of Sect. 4.2.3 (also in Conclusion and perspectives). It is true that the approach (Sect. 4.2.1) does not resolve the potential impact of

biases in model physics or biogeochemistry on the spatial distribution of the flux. These biases are inherent to the use of a single model. However, if we aim to mitigate at least part of these model-intrinsic biases, an intercomparison exercise is needed, and our approach (Sect. 4.2.1), being highly adaptable, is particularly well suited for that purpose.

- Importance of terrestrial organic carbon discussion: The (equal?) importance of terrestrial organic carbon inputs for riverine-induced outgassing, which is really only very coarsely taken into consideration here, is still not clearly enough stated. Estimating the terrestrial organic contribution by correcting the simulations with burial observations as presented here would, in my opinion, not be robust as the regional outgassing is not necessarily coupled to regional burial fluxes associated with terrestrial organic carbon inputs (as one could make the case for terrestrial alkalinity/DIC). It is fine to not address this in the manuscript, but limitations regarding especially spatial patterns need to be acknowledged clearly (I am aware there is was single sentence introduced in the discussion section on this).

We fully agree on the importance of terrestrial organic carbon and on the need to better understand, represent, and constrain it in the future. This is, in fact, a central theme throughout the manuscript. From the introduction, we emphasize the significance of riverine organic matter and its lability. In our set of sensitivity simulations, we included a panel of four experiments specifically designed to investigate and illustrate the consequences of changing the partitioning of the riverine input (rivorg and rivinorg), its lability (rivref), and the magnitude of the total riverine flux. The assumptions related to riverine discharge, particularly its organic component, are at the heart of the present study.

We detail the influence of these assumptions on the flux distribution in Sect. 3.3.2, and discuss it in the penultimate paragraph of Sect. 4.2.3. We further highlight the critical need to improve our understanding of the fate of terrestrial organic matter in the 'Conclusion and perspectives' section.

While we do not resolve the open questions surrounding riverine terrestrial organic matter, we do illustrate the range of its potential impacts through our sensitivity simulations. Furthermore, we propose a flexible framework (Sect. 4.2.1) for combining sensitivity simulations, which can readily accommodate future updates reflecting revised understanding of the fate of this terrestrial organic carbon flux (e.g. magnitude, lability).

- Clarifications regarding the theoretical framework: The framework is now much more clearly explained! Could you however clarify in the text if the corrections applied are based on global observations, or are you taking into account the broad region definitions addressed in the manuscript (North-Tropics-South)? If I understand correctly, it is the latter, but if the former, I am wondering if the corrections applied might actually be introducing regional biases due to biases in the river inputs and modelled spatial distributions of burial fluxes. I am also wondering to which degree the application of the different correction steps are overriding each other (if I understand correctly, it is the air-sea CO2 flux that is being corrected again from steps 2-4?). Some further clarifications could be helpful here.

We have revised the first paragraph of Sect. 4.2.1 to better clarify our approach:

"The set of sensitivity simulations conducted to validate our theoretical framework spans a wide range of assumptions regarding riverine and burial fluxes of carbon and Alk. This provides all the necessary tools to reassess the spatial distribution of the riverine/burial-driven air–sea carbon flux. As in our global estimate (Sect. 4.1), this reassessment strategy is grounded in the most recent global budgets of carbon and Alk. By logically selecting and weighting some of our sensitivity simulations, we construct a composite simulation whose riverine and burial fluxes match those reported in these global budgets (Table 3). In this way, the composite simulation also combines the associated air–sea carbon fluxes, both at the global scale and regionally. It is this regional aspect that enables a revised estimate of the spatial distribution of the pre-industrial riverine/burial-driven air–sea carbon flux."

Thank you also for the comment, which has invited us to discuss an additional limitation of our approach in Sect. 4.2.3, not mentioned previously:

"A limitation of our approach is that a substantial revision of the spatial distribution of a flux -- such as riverine inputs -- would require rerunning a simulation, as it cannot be addressed through our current framework alone."

Finally, and as expected, the effects of the successive steps on the global air–sea carbon flux, and its regional distribution, can accumulate or partially offset each other as the steps progress (see Fig. E3).

Specific Comments

L13 enabling efficient reassessment -> an efficient reassessment?
This has been modified accordingly.

L41 "literature estimates of riverine and burial fluxes" I would remove "literature" here, since some of these studies derive the river flux magnitudes themselves.
This has been modified accordingly.

L209 "Indeed, while at the global scale, the net air-sea carbon flux directly corresponds to the riverine/burial-driven air-sea carbon f lux (Eq. 2), at the regional scale (N, S, or I, Fig. 3b), the air-sea carbon flux (FC, air−sea nat. components: one associated with the functioning of the ocean carbon pumps (FC, air−sea pump riverine and burial fluxes (FC, air−sea riv./bur.), which is our primary focus:" Why not just call this the "natural air-sea carbon flux without/prior to river fluxes" or something like this? (I mean the carbon pump processes also affect riverine carbon?) I would also add "..to the theoretically-calculated riverine/burial-driven air-sea carbon flux?"
We agree that, at the regional scale, there is an interaction between the effect of the ocean carbon pumps and the riverine/burial carbon and Alk fluxes on the air-sea carbon flux. However, we chose to retain the terminology we originally used, as it helps to highlight the source of the driver: internal to the ocean for the carbon pumps, and at the system boundaries for the riverine and burial fluxes. We believe this distinction is both more relevant and more accessible for readers. That said, to take your comment into account, we have slightly refined some of the phrasing to be more accurate.
"Indeed, while at the global scale, the net air-sea carbon flux directly corresponds to the riverine/burial-driven air-sea carbon flux (Eq. 2), at the regional scale (N, S, or I, Fig. 3b), the air-sea carbon flux (FC,air−sea nat.) can be decomposed at first approximation into two components: one internal component linked to the functioning of the ocean carbon pumps (FC,air−sea pumps), and a boundary component associated with the riverine and burial fluxes (FC,air−sea riv./bur.) – our primary focus – :"

L222 Importantly, within the context of our study, the absolute values of the fluxes– whether they align with literature estimates or not– are not of primary concern. What matters are the relative differences between these values across simulations, which reflect the assumptions being tested. Could the authors maybe be precise here which assumptions are specifically being tested?
These are specified in the following paragraph, with further details provided in Appendix B2.1. We have clarified the end of this paragraph to ensure continuity for the reader.
"What matters are the relative differences between these values across simulations, which reflect the assumptions being tested and briefly outlined below (see Sect. B2.1 for further details)."

Table 3 This Table should be explained in more detail in the text.
We believe that this Table is already sufficiently explained in the main text. Going further would require detailing the numbers it contains (as in the first version of the manuscript), but this would be verbose and reduce readability. The Table thus serves to highlight in the text only the key elements and figures related to the approach (Sect. 4.1.1), the findings (Sect. 4.1.2), and the discussion (Sect. 4.1.3), without overwhelming the reader.