Author comment

We thank both reviewers for their supportive comments on our manuscript.
Both of them have raised the issue that the figures in Section 4 were difficult to understand because they only have units (e.g., "mol m^{-3}") as axis label. We will add the name of the geochemical species and fluxes in revised figures. We also agree with the suggestion of Reviewer #1 to plot fluxes going out of the Arctic as negative in Fig. 11, and we will update the figure accordingly. Lastly, we agree to summarize the mechanisms explaining Arctic oxygen variations with a schematic.
As for Revierwer #2's comment on "multi-million years", it was meant to refer to the timing of the evolution of "long" geochemical cycles (oxygen and sulfur), rather than the geodynamical evolution of Earth, such as continental drift or oceanic basin opening. These processes will require further model development to be taken into account.

Specific reviewers comment (written in purple and indented) are addressed hereafter.

Reviewer #1

Main comments:

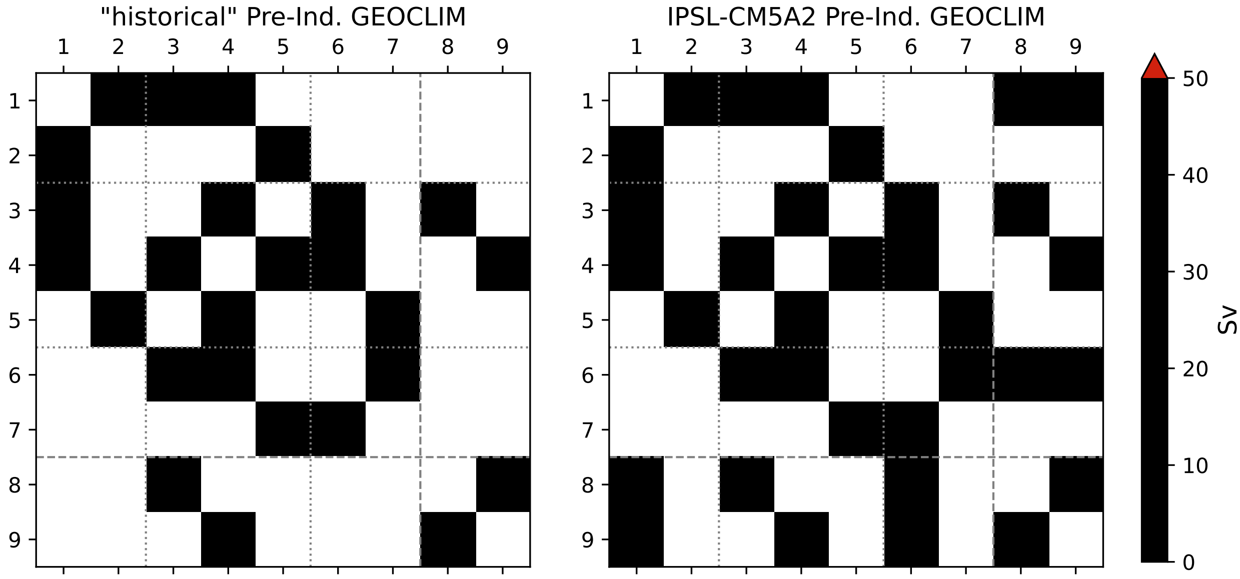
> In Sec. 2 (model description) and Sec. 3 (boundary conditions), the authors outline equations and interpolation methods used in the model. I would like to see a clearer indication of what components/equations are new to GEOCLIM7 and what components were previously published, and where. The sections that contain descriptions of pre-existing GEOCLIM equations lack referencing. I recommend adding citations to papers and/or GEOCLIM versions where the components are first described, tested, and applied. Such complete referencing will not only serve as a historical record of GEOCLIM development but also to track the steps that have been taken to validate the model and evaluate its performance.

We agree with the reviewer. We propose, for Section 2, to add a paragraph at the beginning of each sub-sub-section, indicating if the equations presented are new to GEOCLIM7, and if not, giving the references. For Section 3 (boundary conditions), we propose to do so within the text, when elements are introduced.

> The flexible ocean boxes are a novel addition in which ocean circulation is obtained from GCM output and converted into exchange fluxes between GEOCLIM boxes using a new tool. Can you show that the tool is indeed able to accurately reconstruct the large-scale ocean circulation, in particular the vertical component (WV) that is indirectly calculated? Perhaps a figure that compares the ocean fluxes in the GCM with that of an older standard GEOCLIM configuration and that using the new version? Would also be useful to reference Section 4 (where the ocean boxes specification is tested) in Section 3.2.

Indeed, the "cutting" of the oceanic boxes within the oceanic grid of IPSL-CLM5A2 have been carefully designed so that the main characteristic of oceanic circulations are retained.
Here follows a comparison of the "historical" exchange matrix, used in previous GEOCLIM versions, and the exchange matrix computed from IPSL-CLM5A2 outputs. Note that these matrices can be found in the GEOCLIM repository: "INPUT/COMBINE/historical/exchange_2.dat" and "INPUT/COMBINE/IPSL-PI-ref/exchange_2.dat".
In previous GEOCLIM, oceanic circulation was empirically parameterized. For instance, the high-latitude deep water formation was set at 20 Sv in both northern (box #1 to #2) and southern (box #6 to #7) hemisphere. With the current method, these flux are 10.3 Sv and 8.9 Sv (respectively), wich

is consistent with the values observed in IPSL-CLM5A2 pre-industrial simulations, though somewhat attenuated.



Exchange matrices representing oceanic circulation between GEOCLIM boxes. Fluxes are oriented from COLUMN *i* to LINE *j*. The ordering of boxes is: #1-2: N hemisphere high-latitude (surface and deep), #3-4-5: mid-latitude (surface, intermediate and deep), #6-7: S hemisphere high-latitude, #8-9: coastal oceans (surface and deep).

> Another innovation is the seafloor sediment routing scheme (Sec. 2.3.2). A schematic representation of the routing scheme would be helpful to visualize the mechanics of transport and deposition fluxes, or if this methodology was developed in a different study, cite the appropriate paper. The manuscript is missing a test case scenario that demonstrates the implication of this innovation. How does this improve the model?

A schematic representation of this routing scheme would be a good addition. We will include one in our revised manuscript. Also, part of this scheme was already developed by Maffre et al. (2021), which we hinted in the last paragraph of the Section (2.3.2). In the revised manuscript, we will explicitly mention it in the the beginning of the Section.

> If I understand correctly, two tools are designed to (1) convert ocean water exchange fluxes from GCM to GEOCLIM (BC_generator.py), and to (2) generate a river routing scheme (basinmap_editor.py). Great that these are made publicly available along with the GEOCLIM download. The scripts are clear and well annotated, but it is not clear from the manuscript text how one would use these to re-grid their own GCM outputs. As highlighted by the authors, the main improvement of GEOCLIM7 is that boundary conditions from any GCM (not just FOAM) can be used so a step-by-step outline of how users would go about this seems indispensable. For instance, you could provide a readme file or short manual along with these scripts to improve user experience? Or indicate what scripts are needed to perform each step in Figure 3?

The reviewer understanding is correct. We agree with both suggestions. We intend to write a step-by-step script explaining how to generate GEOCLIM boundary conditions file from IPSL-CM5A2 pre-industrial outputs, and reproduce the pre-industrial control run of GEOCLIM presented here. This script will be included in GEOCLIM repository.
Indicating on Fig. 3 the names of the Python scripts needed for each step would be helpful. We will think on how to update the figure without overloading it with information. One option would be to add it in the Figure caption.

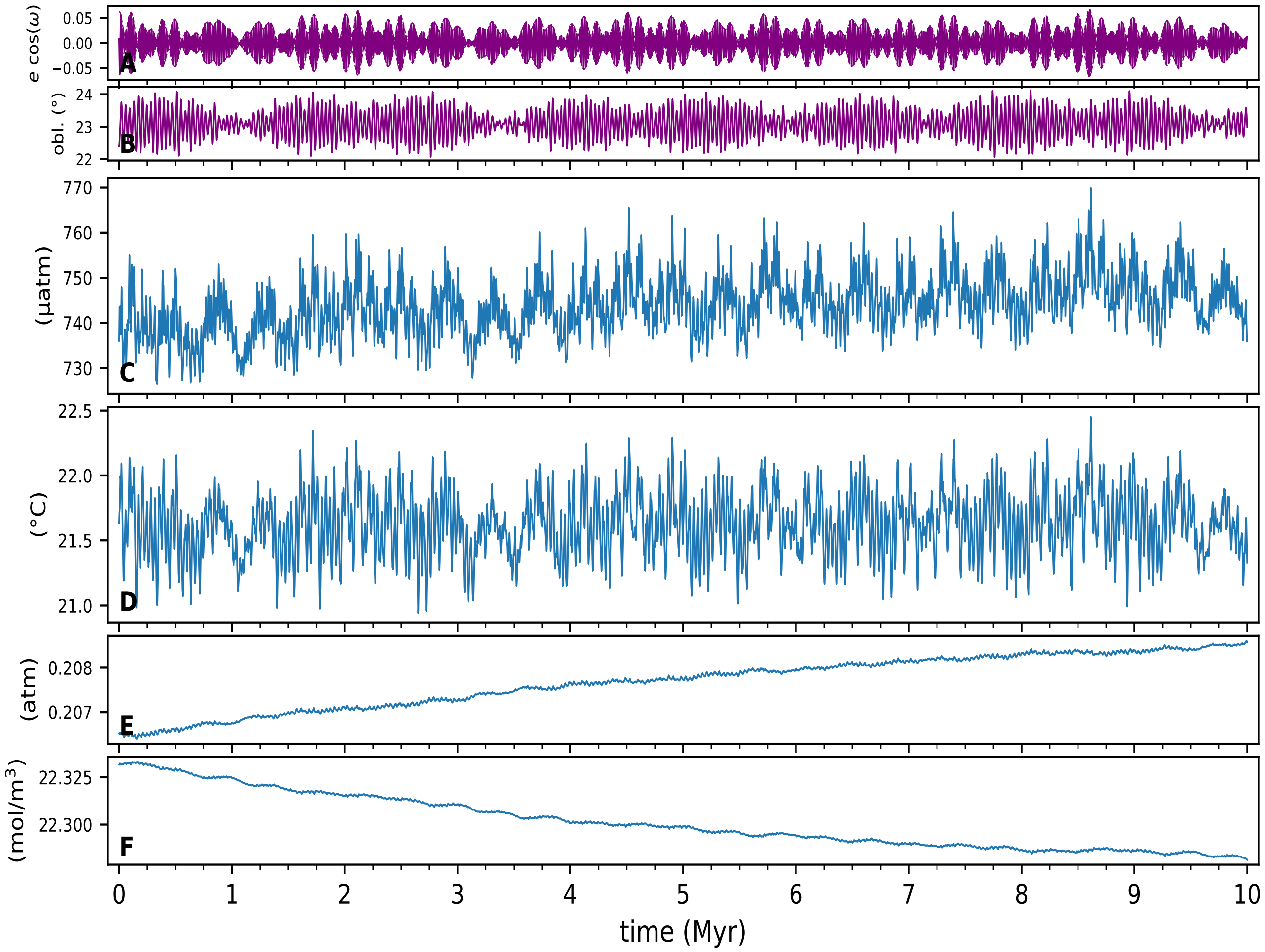
> Is the lithological information (Section 3.4.2) a new addition to GEOCLIM7? If it is, this calls for a sensitivity test to check how sensitive results are to choice of lithology, especially important for paleo-configs in which lithology is often unknown. If this is not a new addition of GEOCLIM7, cite the appropriate manuscript in which it was developed and tested.

Lithological information is not a new addition of GEOCLIM7. It has been implemented and tested in “GEOCLIM-DynSoil-steady-state” (i.e., without the ocean-atmosphere geochemistry module) by Park et al. (2020), and already used in “full” GEOCLIM (v5.3) by Maffre et al. (2021), with the exception of strontium isotopic signatures (see our answer to line-by-line comments below). We will add this information in the revised manuscript.

> The Turonian experiment nicely demonstrates the applicability of the new model components. However, I’d like to see an explanation or discussion about the multilinear interpolation between climate fields of variable pCO2 and orbits (as mentioned in Section 3.4.3). In particular, because the relation between pCO2 and global climate response is not linear and only two end-member CO2 values are tested. Considering this, it would be useful to know if (and by how much) the pCO2 in the ‘All processes’ experiment changes, i.e. how do modelled variations fall in the range of 560-1120 uatm?

In a first draft of the article, we intended to present the following figure (top of next page), but decided not to do so, in order not to lengthen the article too much. We propose to keep panels A to D of this figure (with the names of the variables indicated on the y axis labels, as requested for Figs. 9-11). This new figure can be added it between Sect. 4.4 and 4.5.

We also realized that an important information was missing in the manuscript. The multilinear interpolation is performed with respect to the “raw” values of orbital parameters and of “log(pCO2)”, to account for the logarithmic sensitivity of climate to CO2. We will add this information in the revised manuscript, and hope it answers the reviewer’s concern.



A-B: orbital parameters. C: atmospheric pCO2, D: global mean surface temperature, E: atmospheric pO2, F: Mean oceanic sulfate concentration.

> I'm surprised to see how insensitive the system is to orbital forcing in the 'Cont. fluxes' experiment. The authors explain the weak response by the low absolute P weathering flux but this, in my opinion, is an incomplete assessment. Is it possible that orbital changes to the terrestrial P flux are subdued because of the annual averaged surface temperature and runoff fields, unable to capture nonlinear seasonal behaviours of weathering (e.g equations in Sec. 2.4)? Can you address the absence of seasons in the model and how that may impact the orbital results, for weathering but also ocean circulation?

The reviewer raised a point worth of discussion, although it could only be speculative. There is indeed no guarantee that weathering behaves linearly, and that integrated weathering flux throughout a seasonal cycle matches the flux computed with the anually-integrated runoff flux. However, one cannot say if existing non-linearities will increase (convex runoff-weathering relationship) or reduce (concave runoff-weathering relationship) the annually-integrated weathering flux. In other words, if stronger seasonality, at same annual mean runoff, would increase of reduce the weathering flux.
When examining concentration-discharge relationships in time-series of monitored rivers, a dilution behavior has often been observed (e.g., Ibarra et al., *GCA*, 2016, 10.1016/j.gca.2016.07.006, Ibarra et al., *Acta. Geochim.*, 2017 10.1007/s11631-017-0177-z), pointing toward a concave runoff-weathering relationship. A more complex behavior, with a hysteresis loop throughout the annual cycle has also been observed (Moquet et al., *Environ. Sci. Pollut. Res.*, 2016, 10.1007/s11356-015-5503-6), but not pointing either toward a convex relationship. On the other hand, some studies have argued that stronger seasonality promotes higher weathering fluxes (e.g., Wirchern et al., *Clim.*

Past, 2024, 10.5194/cp-20-415-2024; De Vleeschouwer et al., *Nat. Rev. Earth Environ.*, 2024, 10.1038/s43017-023-00505-x).
We will add a discussion about this in the revised manuscript, indicating that it is unclear how it would affect our results.

As for the oceanic circulation, since the mixing fluxes are U*[X] (where U is the water flux and [X] the concentration of a species); a non-linearity would only exist if U and [X] covary throughout the year. The residence time of all species species is determined by the residence time of water in each box “volume / Σ U”, which, in GEOCLIM simulation, is between 1.5 and 300 years for open-ocean boxes. One should thus not expect significant variations of concentration within a year. In coastal boxes, on the other hand, this water residence time is often less than a year (down to 0.1 year in the smallest boxes). We cannot exclude that a non-linearity exist in the exchanges fluxes between coastal and open-ocean boxes. Yet, it is difficult to say how it would affect our results.

Other comments:

Here follows a selection from line-by-line comments from reviewer #1. All the remaining comments are suggestions of rephrasing, referencing, further explanations on equation terms, and typos. They will be addressed in the revised manuscript.

> Figure 1: Add number to each box to reflect the definitions on previous page

This is a good suggestion. We will add it on the revised Figure 1.

> L. 193. Should 15/8Fsulw be xFsulw as in Eq.1 and 12?

The factor 15/8 is not related to xFsulw. It comes from the stoichiometry of the sulfide weathering equation:
$(1/2)\ FeS_2 + (15/8)\ O_2 + (7/4)\ H_2O \ \text{-->}\ (1/2)\ Fe(OH)_3 + 2\ H^+ + SO_4^{2-}$
i.e., for each mole of S oxidated, 15/8 moles of O_2 are consumed. The factor xFsulw represents the fraction of generated H^+ that is neutralized by dissolving silicates (instead of carbonates), in a second reaction step.
In the revised manuscript, we will indicate “the factor 15/8 comes from stoichiometry of sulfide weathering reaction”.

> L. 215. Add reference to origin of this equation

There is actually no reference for this equation. This formulation was unchanged from the first published version of GEOCLIM (Godderis & Joachimsky, 2004), whose strontium model comes from François & Walker, *American Journal of Science*, vol. 292(2), pp. 81-135 (1992).
However, neither Godderis & Joachimsky (2004) nor François & Walker (1992) indicated the mathematical functional form of the carbonate burial Sr sink.
Eq. 13 should be seen as a tuning equation: $[Sr]_{ref}$ is the pre-industrial mean oceanic Sr concentration. Therefore, given that the proportion of Sr input from the ocean is, approximately 72% from silicate weathering and 28% from carbonate weathering, setting the Sr:C ratio of PIC this way ensures that the mean oceanic Sr concentration making the carbonate burial Sr sink balance the input fluxes is $[Sr]_{ref}$.
We will indicate this in the revised manuscript.

> L. 223. Are Fadv and Fsink missing in Eq.15? They are described on L.224-225

Fadv and Fsink are implicitly included in Eq. 15 because they are both based on the same generic form described in Eq. 15 (i.e., $F(X)*(\delta^nX_{flux} - \delta^nX_i)$, cf Eq. 51 and 52. The same remark can be make for Eq. 16. In the revised manuscript, we will explicitly state that Fadv and Fsink falls within the general form of Eq. 15 (and 16), and refer to Eq. 51 and 52.

> L. 266-270. Eq.21-22. Just checking, no isotopic fractionation associated with dissolution and remineralization?

The $\delta^{13}C$ of the new productivity is indeed slightly lighter than the $\delta^{13}C$ of the sedimentary organic carbon. But the difference is small (about 1.5 ‰). It appears that this difference may fluctuate, for instance when non-photosynthetic producers are incorporated within the sediments, such as sulfate-reducing bacteria. Such conditions cannot be simulated within our large scale model. For this reason, the difference between primary productivity and sedimentary organic carbon is assumed to be constant, and is thus incorporated within the biological fractionation.
Similarly, the $\delta^{13}C$ of sedimentary carbonates can be slightly different from the $\delta^{13}C$ of CO_3^{2-}. Again, this is a small effect, and fluctuations cannot be simulated.
Ref: J.M. Hayes, H. Strauss, A.J. Kaufman. The abundance of ^{13}C in marine organic matter and isotopic fractionation in the global biogeochemical cycle of carbon during the last 800 Ma. Chemical Geology, 161, 103-125, 1999.

> L. 276-280. Is there an upper limit to the dependence of primary productivity on P flux/inventory? E.g. in environments where the P inventory is extremely high, is there still a dependence on P input flux?

This equation is an approximation, as only one nutrient (P) is represented in GEOCLIM. Hence, the behavior of phosphorus, in the model, may be viewed as the average behavior of all nutrients. Primary productivity in modern ocean is either limited by nutrient,or by light (at high latitude). Regions with high P inventory are either regions where another nutrient is limiting (which cannot be simulated by GEOCLIM), or high latitudes, limited by light (which is already parameterized in GEOCLIM).

> L. 698. So the terrestrial Corg export is independent of terrestrial productivity?

Indeed. This approximation is based on the work from Galy (2015), that identified the erosion and export of bulk material as the limiting process for organic C export. We will indicate it in the revised manuscript.

> L. 724. What interpolation method is used for the climate fields? Add reference.

Good catch, this information was only given in section 3.4.3 (line 922). We will add this information here, and refer to section 3.4.3.

> Table 4 caption. Include meaning of abbreviations and explain that where one value is given, it applies to all lithologies (is that correct?)

The reviewer is correct. We will add this information in Table 4’s caption.

> L. 745. Since you provide a complete overview of GEOCLIM, can you also add information about the model run/computation time along with the info about time stepping?

Adding information on model run time at the end of Section 2.5 is a good suggestion. In a nutshell, GEOCLIM runs at 30 minutes per Myr in its lowest resolution, and up to 5 hours per Myr at the highest resolution tested here.

> L. 865-866. Reference the origin of these equations.

As stated earlier, we will indicated in the revised manuscript which equations were taken from previous version of GEOCLIM (it is the case for those two).

> L. 933. In addition to silicate weathering, and volcanic/anthropogenic CO2 outgassing, does fossil Corg weathering not contribute to the CO2 fluxes to the atmosphere?

It does contributes to CO2 fluxes to the atmosphere, though indirectly as it generates DIC first (see reponse to Reviewer #2). But the reason why it is absent from this calibration step is because the calibration is meant to achieve a steady-state of all geochemical cycles, which implies that fossil Corg weathering is balanced by Corg burial (with zero net C flux). Therefore, at steady-state, the silicate weathering flux should only balance the magmatic degassing flux.

> Figure 3. Clarify if ‘paleo-geology’ refers to the lithology or to the geological categories? Make sure to include both in Fig.3

Paleo-geology only refers to the geological categories (each of them would contain a set of different lithological classes). We will add this information in the revised manuscript.

> Figure 4. Can you add the modern mean profiles of these basins so readers can visually compare GEOCLIM pre-industrial output to the modern?

This is a good suggestion. This data is available in GLODAP and World Ocean Atlas databases. We will include it in revised Figure 4, for each corresponding box of GEOCLIM.

> L. 1020. Why not include tuning of δ13C? The equations are listed in Section 2.2.3 so it leaves the reader wondering how these are resolved.

Less attention has been paid to isotopic tracers, in this manuscript, as it is not its main focus. The tuning of Sr isotopic cycle was less a purpose than a necessity: with the addition of silicate lithological classes, a strontium isotopic signature has to be assigned to each class. Although the lithological classes were already added by Maffre et al. (2021), they did not update the strontium isotopic signatures in a way consistent with the global value from continental fluxes. The carbon isotopes, on the other hand, is not concerned by this issue since all carbon associated with silicate weathering comes from the atmosphere. All parameters associated with carbon isotopic cycle are unchanged from Godderis & Joachimsky (2004).
We will change this section in the revised manuscript, to indicate that the tuning of isotopic tracers was not the purpose of this manuscript.

> L. 1110. Has this acceleration technique been published or tested before to yield accurate results? If so, please cite reference. If not, include a comparison between an accelerated and non-accelerated run to demonstrate the accuracy of final results.

This acceleration technique as not been published before, though it was used for all steady-state simulations of Maffre et al. (2021), and likely other GEOCLIM studies.

It simply consists in applying a factor “1/A” before the time derivative in Eq. 7 (atmospheric oxygen) and Eq. 8 (oceanic sulfate). Were “A”, the acceleration factor, is typically 30 or 100, being only limited by the stability of the numerical resolution.
We argue that it does not require a demonstration of the accuracy, because it is mathematically exact. The steady-state value is not affected by the factor “1/A”, since the time derivative term is null at steady-state, this factor will disappear from the steady-state equation.
We will provide this information in the revised manuscript.

> L. 1127. Checking Laugie et al. (Fig.5), it seems like panel H yields the best match with Laugie et al. (2021)?

We do not agree with the reviewer. GEOCLIM has a resolution to coarse to simulate strong local anoxia like the vertical profile #5 of Fig. 5 of Laugie et al. (2021). Instead, Fig. 2 of Laugie et al. shows large horizontal variations in oxygen concentrations (GEOCLIM also cannot reproduce the anoxic layer at 700 m, Fig. 5C of Laugie et al., due to its coarse vertical resolution).
On average (horizontally and vertically), the difference of O2 concentration, in Laugie et al.’s simulation, between proto-North Atlantic and Pacific is ~ 100 mmol/m^3, which is consistent with panel G, and less consistent with panel H where that difference is ~ 200 mmol/m^3.

> Figure 11. It is difficult to interpret the results in this figure because the reader has to switch between the caption and the subpanels to figure out the variables that are plotted. Can you add the variable names to each subpanel to improve legibility? Plus, both the influx and outflux of an Arctic box are plotted as positive values. It would be more intuitive to plot influxes as ‘positive’ and outflux as ‘negative’ to reflex the result of the ‘net’ flux.

See our comment at the beginning of present document.

> L. 1183. Evacuate → ‘eliminate’ or ‘confirm’? This entire paragraph (L.1180-1205) needs some re-writing to present a clearer explanation, it is difficult to follow in the current state, partially because Fig.11 is not immediately intuitive. Perhaps a schematic of the proposed mechanism that drives Arctic [O2] changes can help, showing changing P fluxes in and out the Arctic boxes.

We will pay attention to make this paragraph clearer (in addition to Fig. 11 modification). A schematic would be helpful, and we will provide one.

Reviewer #2

Selection of most critical comments:

> Line 174 and 194: “Ffocw” - organic carbon weathering – is not included in the CO2 mass balance but should produce CO2? Some further explanation of the oxygen cycle mass balance would also be useful, e.g. how it differs from the GEOCARBSULF.

Indeed, fossil organic carbon weathering is assumed to generate DIC, but not directly gaseous CO2. It is therefore included in Eq. (1) (line 163), but not Eq. (2). The idea behind this representation is that this reaction occurs in river, or in water-saturated soils. Degassing of CO2 is delayed, and occurs when surface oceanic boxes exchanges with the atmosphere, as GEOCLIM does not represent rivers: continental fluxes are instantaneously transferred to the ocean.

> Line 295: “This 10-fold reduction in coastal boxes was tuned in order to avoid massive precipitation of carbonates in coastal surface boxes”. Does this hint that the formulation is not very accurate in the first place? Why is the initial prediction so much higher than in the real world?

The reviewer is probably right that the formulation is not very accurate in the first place. Setting a global, uniform fraction of carbonate producers is a simplification. Although it may be true that the fraction of carbonate producers (excluding reef bioconstructors) is indeed lower in coastal, estuarial and epicontinental environments than in pelagic environment, there are also some clues that GEOCLIM is missing some processes. Sulpis et al., *Nat. Geosc.* (2021, 10.1038/s41561-021-00743-y) showed that a significant part of carbonate particles dissolve in shallow waters, above the aragonite lysocline, driven by metabolic CO2 production within marine organisms or aggregates. A process that is ignored in GEOCLIM. That same study also showed that a large fraction of “deep” carbonate dissolution occurs at water-sediment interface. In GEOCLIM, all carbonate particles reaching seafloor are assumed to be preserved and buried. Carbonate is not tracked by the sediment advection scheme, the way organic carbon is. Therefore, GEOCLIM cannot represent the dissolution of carbonate particles produced in coastal environment, but exported and deposited in deep ocean seafloor.
In summary, this formulation is an approximation to compensate for processes that are either poorly constrained, or that cannot be represented in the current state of GEOCLIM, and are left for future development.

> Line 557: hydrothermal burial is with iron oxides?

This representation comes from Goddéris & Joachimsky (2004), meaning to simulate “the scavenging of phosphorus by ferric oxyhydroxides formed within hydrothermal systems” (quoting from Goddéris & Joachimsky 2004), process is described with more details in Benitez-Nelson, Earth Sci Rev (2000): 10.1016/S0012-8252(00)00018-0
Nevertheless, we agree that this formulation is too simple to meaningfully represent such a specific process. In the revised manuscript, we will refer to it as “net P sink due to hydrothermal processes”.

The remaining comments of reviewer #2 are suggestions of rephrasing, referencing, further explanations on equation terms, and typos. They will be addressed in the revised manuscript.