[Reply to Reviewer #1]

We appreciate the constructive and beneficial comments by the reviewer.

Q1: In the introduction, the authors emphasized the importance of transiency to explain the glacial-interglacial carbon cycle. I would like to see more discussion of the time scale of the response of the carbon cycle. How fast do carbonate sediments change in response to changes in sea level? How do the time scales of carbonate sediment expansion and contraction differ from other processes that consist of the ocean carbon cycle (solubility, biological pumps, and ocean circulation)?

A1: We will re-organize the introduction to a great extent, and will include the description about the timescale of various processes relevant to the carbon cycle. We will thereby make the concept of this study clearer.

Q2: Although the authors focus on shallow-water carbonate sediments, carbonate is thought to be equally buried in the deep ocean (Cartapanis et al., 2018). I would like to see a clearer separation of these two contributions in the discussion.

A2: This comment by the reviewer is totally reasonable. We will include relevant descriptions in a reorganized introduction, and will extend the discussion in Section 4.1 by including deep-ocean processes. As the reviewer pointed out, in the mass balance calculation to connect the glacial and modern states, the outflow of CaCO<sub>3</sub> through deep-sea burial over the entire periods of time since the LGM should be also taken into account. At the same time, we will also mention a possible importance of the inflow by the land weathering, because, depending on the magnitudes of the outflow and inflow, they would have worked as another source or sink of alkalinity as a whole.

Q3: In the LGM experiments, whole ocean alkalinity is increased to adjust the atmospheric CO2 concentration to the ice core data. The magnitude of this increase does not seem to be explicitly stated (not shown in Table 2), but is it appropriate?

A3: The magnitude of alkalinity adjustment is already described in the right-most column of Table 1, and appears also in the first paragraph of Section 4.1. See also A6 below.

Q4: In section 4.1, the authors explained the change in alkalinity is related to the changes in shallow-water coral reefs but is it necessary to consider the effect of carbonate compensation, including deep-sea carbonate sediments (e.g., Brovkin et al., 2012; Ganopolski and Brovkin, 2017; Kobayashi et al., 2021)?

A4: As the reviewer pointed out, processes related to the deep-sea carbonate sediments should be included in the discussion about the compatibility of alkalinity inventory between LGM and PI in Section 4.1. As answered in A2, we will extend the section to include the discussion about the flux of alkalinity by deep-sea carbonate burial and that by the land weathering as well based on the quantitative estimates for each of them (rather than the mechanism of carbonate compensation itself) because the imbalance between them would matter in the context of the discussion.

Q5: It is reported that PMIP models tend to simulate lower ocean carbon sequestration and higher atmospheric CO2 if they had a lower ocean volume at the LGM (Lhardy et al., 2021). Do the ocean bathymetry and volume change in this study? It is expected that a greater change in alkalinity would be required if a lower ocean volume is adopted.

A5: Yes, we changed the bathymetry for the LGM, and the effects of sea-water volume change are included in the simulations. As described in Section 2.2, we applied the volume-change effects following the PMIP4 protocol (Kageyama et al., 2017). The volume change alters the concentration of alkalinity and other tracers, but does not affect the inventories.

Specific comments:

Q6: P1/L6: "The increase..." I am not convinced by this statement because of the lack of information on the inventory change in alkalinity.

A6: This question is closely related to Q3. Although relevant information is already available as we answered in A3, we will also specify the required increase of alkalinity inventory in the main text of a revised method section.

Q7: P2/L15: How much carbon (PgC) does the change in DIC in the deep ocean (µmol/kg) between the LGM and the present day correspond to?

A7: It is reported that the changes in concentration correspond to 730–980 PgC (Sarnthein et al., 2013) and 687 PgC (Skinner et al., 2015) in the respective papers. We will add those numbers in PgC (or GtC) to the introduction.

Q8: P2/L16: I would like to know more about the coral reef hypothesis and the subsequent study's discussion of the impact of changes in shallow-water carbonate sedimentation on global carbon cycle changes.

A8: As answered in A1, we will reorganize the introduction and will add more description of the coral reef hypothesis.

Q9: P2/L20: I would like to know the explicit statement about the impact of changes in DIC and alkalinity on atmospheric CO2 concentrations. In other words, the increase in DIC in the deep ocean during glacial periods and the increase in alkalinity throughout the ocean both contribute to a decrease in atmospheric CO2.

A9: The increase in the deep-water DIC storage should be the outcome of combined two factors: the change in the vertical structure of carbon storage and the change in the mean concentration. The increased vertical contrast contributes to the decrease in  $pCO_2$  as the increase of alkalinity does, while the increase of mean DIC concentration has the opposite effect. The adjustment of DIC inventory in the entire ocean of this study will provide the net effect of the two factors that is consistent with the estimated deep DIC storage. We will add these description to the introduction.

Q10: P3/L21: Would you explain a little more about the advantages of using SolveSAPHE?

A10: SolveSAPHE is based on a more robust algorithm to provide better numerical stability than the original pH solver, while it can offer comparable accuracy of pH calculation. As far as we experienced, the original solver was frequently highly sensitive to the chemical composition of seawater, so that it often caused numerical instability especially in a spin-up phase. We therefore adopted SolveSAPHE and continued to use it for consistency.

Q11: P3/L30: Changes in carbonate sediment burial result in changes in whole ocean alkalinity (Kobayashi et al., 2021). Is it difficult to investigate the changes in carbonate burial by using the sediment model?

A11: The amount of global carbonate burial was not available in principle in the application of MEDUSA in the remapped (1deg x 1deg) domain, because it did not cover all the ocean floors of the POP2 domain. For a revised manuscript, we coupled MEDUSA simply or directly to every bottom grid cell of POP2 as in the preceding study (Kurahashi-Nakamura et al., 2020) to involve the whole ocean floors and carried out similar simulations again. This new method has enabled us to provide the global sum of MAR of CaCO<sub>3</sub> for each experiment, which are 0.094 GtC/yr (expPI), 0.14 GtC/yr (expLGM), 0.12 GtC/yr (expLGMws), and 0.087 GtC/yr (expLGMss). Although the modeled modern MAR is ~25% smaller than the estimate (~0.13 GtC/yr) by Cartapanis et al. (2018), the LGM values approximate the +-2-sigma range of the estimate for the glacial period (the mean: ~0.11 GtC/yr) by the same study. Another advantage of the new method is that now we can provide global continuous maps of MAR. We will introduce new plots for CaCO<sub>3</sub> MAR in a revised manuscript.

Q12: P3/L30: I would like to see information on carbonate burial fluxes and sediment distribution throughout the ocean as calculated by the sediment model in expPI. p7/L8 includes a brief description, but I do not think it is sufficient.

A12: As answered in A11, in a revised manuscript we will introduce a new coupling scheme between MEDUSA and CESM to cover the global ocean, so that we will be able to provide further MEDUSA-related results.

Q13: P4/L13: Would you cite a reference for the Ruddiman belt?

A13: Although here we show a relevant reference "W.F. Ruddiman, Late Quaternary deposition of ice-rafted sand in the subpolar North Atlantic (lat 40 to 65N), Geol. Soc. Am. Bull., 88 (1977), pp. 1813-1827", we will delete the words in a revised manuscript following a comment by the other reviewer.

Q14: P4/L15: Why did you choose 0.25 Sv for the freshwater input to the Southern Ocean?

A14: We did test runs in which we tried various different amounts of fresh water for the additional forcing, and empirically found 0.25 Sv to realize a shallower-but-stronger AMOC structure. That is to say, we needed to subtract that amount of freshwater for the southern-sourced water to counteract the strengthened northern-sourced water.

Q15: P4/L34: What is the total change in alkalinity adjusted for "second-guess"? Would you add the information to Table 1?

A15: The information is already available in the right-most column of Table 1, and appear also in the first paragraph of Section 4.1.

Q16: P5/L14: Would you consider showing the stream function as well?

A16: We will add stream-function plots as well in a revised manuscript.

Q17: P5/L19: From my understanding, LGMss was an experiment in which freshwater was removed from the LGM in the North Atlantic and fresh water was added in the Southern Ocean, resulting in a stronger north-south density gradient at the sea surface. However, why do we see a somewhat weaker AMOC relative to the LGM?

A17: We assume that this is a misunderstanding by the reviewer. Freshwater was removed also from the Southern Ocean as shown in the text and in Table 1.

Q18: P5/26: How about comparing the modeled deep-sea salinity to paleo records (e.g., Adkins et al., 2002; Insua et al., 2014; Homola et al., 2021?

A18: We will add new plots to show the comparison between the model results and the data given by the three studies.

Q19: P6/L9: Can we assume that this change is consistent with estimates of changes in terrestrial carbon storage (e.g., Peterson et al., 2014; Jeltsch-Thömmes et al., 2019)?

A19: Yes, as discussed in Section 4.1 by referring to Kemppinen et al. (2019), our model results show the growth of terrestrial carbon storage of 340--390 GtC, which is within the range of estimates by various previous studies including Peterson et al. (2014). Although Jeltsch-Thömmes et al. (2019) is not included in the compilation of previous work, their estimate (450 to 1250 GtC) is also within the range of the uncertainty, which does not affect our discussion. We will add the latter article to the reference in the revised manuscript.

Q20: P6/L9: Is the total amount of carbon in the atmosphere, ocean, and terrestrial reservoir the same for all experiments? It is difficult for me to understand the experimental design.

A20: No, it isn't. Each experiment has a different total amount of carbon stored in the entire (atmosphere-ocean-land) system. In our experiments, instead of fixing the total amount, we tuned the size of the atmospheric reservoir to a specific amount (i.e. ~190 ppm), which practically governed the terrestrial reservoir's size, and also tuned the size of the deep-ocean reservoir as well. The size of the shallow-ocean reservoir varied among different experiments depending on the vertical gradient of DIC concentration in the ocean, hence the ocean circulation. The difference in the total carbon inventory in the entire system can be therefore interpreted as the uncertainty of the total carbon inventory that arose from the uncertain ocean circulation. By adding a similar description, we will modify the manuscript to better explain the experimental design.

Q21: P6/L14: Would you show us d13C paleo records (e.g., Peterson et al., 2014) in the figure? It would make comparisons with other studies easier.

A21: We will introduce new figures of the modelled  $\delta^{13}C_{DIC}$  with overlaid plots of paleo records by Peterson et al. (2014) and Yu et al. (2020).

Q22: P6/L22: The impact of changes in the distribution of export production on nutrients and AOUs should also be considered. How about conducting sensitivity experiments with fixed biological fluxes?

A22: We will add discussion about the topic by introducing new plots for PO<sub>4</sub> and AOU. Although we find the sensitivity experiments suggested by the reviewer intriguing, we judged that they are beyond the scope of this particular work because the model configurations currently available do not allow us to do that and demand further substantial technical development.

Q23: P6/L25: The decrease in ideal age in the Southern Ocean in Fig. 4 is caused by changes in AABW flow or changes in local convective mixing. Please describe.

A23: The younger ideal age in the Southern Ocean of the LGM experiments is caused by the changes in local convective mixing rather than by the changes in AABW flow. expLGM has a very similar magnitude and geometry of AABW to that in expPI, but nevertheless the ideal age in the Southern Ocean is significantly younger. On the other hand, in expLGMws and expLGMss, more vigorous AABW does not convey the comparatively old water at the depths of 2000-3500 m in the Atlantic to the south of ~40S. In addition, the LGM experiments have a deeper mixed layer depth in the Souther Ocean, which would contribute to the better ventilated Southern Ocean. We will add a similar description to a revised manuscript.

Q24: P6/L26: Some studies have reconstructed carbonate ions from B/Ca (e.g., Rickaby et al., 2010; Yu et al., 2013, 2020). Would you compare your modeling results with them? As stated in the discussion, the increase in alkalinity seems to be overestimated in the current setting.

A24: We will update the plots for the carbonate ion concentrations with overlaid plots of the data by Yu et al. (2020) to facilitate the model-data comparison. We will also refer to the other two papers that have very sparse data points in a discussion section for further comparisons.

Q25: P6/L31: How about showing the reconstructed changes in export production (Kohfeld et al., 2005)? The characteristics of the changes appear to be well reproduced in the model. Also, I would like to see a discussion of the effects of sea ice distribution and iron fertilization on export production.

A25: We will refer to Kohfeld et al. (2005) to update the discussion about the modeled changes of the export production fields. We will add description and discussion to the main text because Kohfeld et al. (2005) only provides the qualitative changes. We will also update the plots for the export production with overlaid sea-ice extent in respective LGM experiments and will discuss the effects of sea ice distribution and iron fertilization on export production with some additional references such as Kurahashi-Nakamura et al. (2007), Sun and Matsumoto (2010), and Gupta et al. (2020).

Q26: P7/L12: Would it make sense to compare this model-data comparison of CaCO3 MAR for other ocean regions (the Southern Ocean and the Pacific Ocean)? It makes the model validity and shortcomings clearer.

A26: As mentioned in A11, we will introduce new plots for CaCO<sub>3</sub> MAR in a revised manuscript to show the model-data comparison in the global ocean.

Q27: P8/L15: The pyrite oxidation showed here as negative feedback on atmospheric CO2, but was insufficiently studied to constrain its quantitative contribution to the glacial-scale carbon cycle. Is my understanding of this correct?

A27: Yes. The pyrite oxidation accompanied by  $CO_2$  release is expected to occur (at least get triggered) during the lowstands of sea-level changes under the glacial low- $pCO_2$  environments, and therefore would potentially work as a negative feedback in the  $pCO_2$  variations in that sense. However, to specify the actual role of the pyrite oxidation in the glacial-interglacial cycles, not only the amount of the associated  $CO_2$  release but also its timing need to be better constrained.

Q28: P8/L32: A recent modeling study of Kobayashi et al. (2021) also used d13C to constrain their ocean carbon cycle fields in the LGM.

A28: We will add the reference in a revised manuscript to the line specified.

Q29: P9/L32: I asked a similar question about the export production, but does the change in sea ice coverage affect the sinking flux of CaCO3 in this region?

A29: Unlike the total export production, the export of CaCO<sub>3</sub> is sensitive to the sea-ice distribution, because, in the BEC model, CaCO<sub>3</sub> production is scaled by the difference between local seawater temperature and the freezing point of seawater (Moore et al., 2004). We will add this description to a revised manuscript.

Q30: P10/L7 Typo? The references are not listed correctly.

A30: We will correct it.

Q31: P10/L13: The maximum values of ideal age in the Pacific appear to be getting younger in expPI, expLGM, and expLGMss (expLGMws). Is this related to the increased AABW-related deep-water flow?

A31: Yes, as the reviewer pointed out, the ideal age of Pacific water is younger in the LGM runs because the fresher southern-sourced deep water is more dominant. In revised Section 4.3, however, we will totally modify the sea-water age and relevant discussion by taking the reservoir age of surface water into account.

Reference:

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