Response to Andreas Schmittner (Referee) (Our response **highlighted gray**.)

General comment:

Yamamoto and co-workers present a nice modeling study of glacial ocean oxygen and carbon changes. The manuscript is well written (except for a few typos) and nicely illustrated. I think the main new finding is that glaciological iron sources from Patagonia are particularly important for lowering atmospheric CO2. Although similar suggestions have been made previously with simpler models (e.g. Brovkin et al., 2007) this study is the first to my knowledge that cleanly separates glaciological from other (desert) dust sources.

However, I have a few concerns that require revisions. Some of those concerns result from a study by Khatiwala et al. that is currently in review with Science Advances. We hope that it will be published soon so that the authors can access it and consider it in their revision.

Response: We are grateful to Professor Andreas Schmittner for careful review and useful comments. The reviewer's comments are helpful for us to improve our manuscript. Referring to the comments, we will carefully revise the manuscript. The specific replies are as follows.

Comment #1

Khatiwala et al. use a data-constrained model of the LGM to decompose the carbon cycle. They show that using the AOU approximation to calculate respired carbon leads to large errors (even the wrong sign) in LGM – PI simulations. This conclusion is supported by previous studies who have demonstrated the errors in the AOU approximation (Russell et al., 2003; Ito et al., 2004; Duteil et al., 2013). For this reason, I would advise not to use it and remove the corresponding parts of the manuscript (e.g. in section 3.2).

Response: We were not aware of these previous studies and agree that AOU contains errors. In the revised manuscript we will not remove section 3.2, but add the following annotation after line 202. We will refer Khatiwala et al. if their manuscript will be accepted during the revision of our manuscript.

"It is important to note that the AOU is different from true oxygen utilization due to air-sea disequilibrium which is on the order of 20 mmol m⁻³ in deep-water formation regions (Russell and Dickson, 2003; Duteil et al., 2013). Changes in surface ocean disequilibrium between PI and LGM simulations might lead to large errors in AOU changes."

Comment #2

It is OK to refer to the iron fertilization effect as increasing the efficiency of the biological pump, but not that the LGM biological pump was enhanced. Khatiwala et al. show that the biological pump was not enhanced, but that airsea disequilibrium was increased, which caused the glacial ocean carbon inventory to be larger.

Response: According to the reviewer's comment, "enhanced biological pump" and/or "biological pump was enhanced" will be removed in the revised manuscript.

Comment #3

Air-sea disequilibrium was enhanced in the LGM not only for carbon but also for oxygen and radiocarbon. Khatiwala et al. show that in their best fitting model the ideal age of the whole ocean is younger, while the whole ocean c14-age is older due to the increased disequilibrium (or increased preformed c14-age). This is relevant for the discussion at the end of section 3 (lines 280-287) and the corresponding parts of the abstract (lines 22-24). Thus, ideal age and c14-age cannot be compared and there may not be a discrepancy here between modeled younger ideal age and older (observed) c14-age. I also think that one quantitative oxygen reconstruction from the Southern Ocean alone (Gottschalk et al. 2016) is not enough to indicate that the model is wrong. Reconstructions have errors and therefore I would not overemphasize this apparent discrepancy.

Response: Thank you for sharing the manuscript. We will add the discussion about the effect of air-sea disequilibrium on ideal age and c14-age if Khatiwala et al. will be accepted during the revision of our manuscript.

Comment #4

Another concern is the discussion of nutrient inventory changes. Somes et al. (2017) have considered this and shown that existing nitrogen isotope data provide no constraints on this effect. I'm also not aware of other observations supporting it (including evidence provided in this manuscript). For this reason, I think this effect remains unconstrained by observations and thus highly uncertain. I'd encourage the authors to reflect this uncertainty more in their discussion of this effect and to cite the above paper, which has also examined its effects on oxygen.

Response: Thank you for your useful suggestion. As reviewer said, the discussion of nutrient inventory changes is necessary for our manuscript. We will add the following discussion to the revised manuscript.

"The changes in nutrient inventory during the LGM have large uncertainties. Previous studies estimate that the oceanic PO₄ and NO₃ inventories could have been 15–40% (Tamburini and Föllmi, 2009; Wallman et al., 2016) and 10–100% (Deutsch et al., 2004; Eugster et al., 2013; Somes et al., 2017) greater during glacial than interglacial periods, respectively. Moreover, Somes et al (2017) shows that sedimentary δ¹⁵N records provide no constraints on this effect. Future simulations should test the biogeochemical sensitivity to changes in nutrient inventory."

Comment #5

The authors claim that their model fits reconstructions of export production by Kohfeld et al. (2005), which show not much change in the Pacific sector of the Southern Ocean. However, there are some newer data from that region by Studer et al. (2015) and Wang et al. (2017) that indicate increased nutrient utilization there as well. This suggests

that the model underestimates iron fertilization in the Pacific sector of the Southern Ocean.

Response: Thank you for information on new data. In addition to suggested references, we also found Kohfeld et al (2013) which include many reconstructions of export production in the Pacific sector of the Southern Ocean. We will add these data to figure 3 and also revise lines 182-188 as follow.

"In the model, EP changes also have an east-west dipole pattern; slight increases of EP are found in the South Pacific Ocean and significant EP increases occur in the South Atlantic and Indian Oceans. We found that this pattern is attributed to iron fertilization by glaciogenic dust. Glaciogenic dust derived from Patagonian glaciers is transported to the South Atlantic and Indian Oceans by the southern westerly wind, but is unable to reach the South Pacific (Fig. S2). Proxy data show no clear east-west dipole pattern, suggesting that the model underestimates iron fertilization in the Pacific sector of the Southern Ocean. However, proxy data in the South Pacific are still sparse and quantitative comparison of EP changes between South Atlantic and South Pacific is limited. Therefore, further proxy data in the South Pacific is required for a comprehensive understanding of the glacial EP changes and iron fertilization."



Figure 3. Model-proxy comparison of EP change from the PI to LGM. EP difference from the PI for (a) LGM_clim, (b) LGM_glac3%, and (c) LGM_all. Circles show proxy data (Kohfeld et al., 2013). Solid (dotted) lines refer to the glacial sea ice fraction of 0.1 in August (February). (d) Zonal mean changes in surface EP from the PI for LGM_clim (black), LGM_glac3% (red), and LGM_all (blue).

In any case, given the uncertainties in existing paleo data and iron models and solubility of iron, it is not fair to say that the upper limit of iron fertilization is 20 ppm as claimed here in lines 214-215. Khatiwala et al. suggest an iron effect of 35 ppm. Here I also disagree with Fortunat's suggestion to mention the CO2 limit in the abstract. I don't think it is a robust result. However, the idea that the effect of iron fertilization is limited and that increasing fluxes will have a smaller effect at high fluxes than at low fluxes is robust and agrees with previous results (Muglia et al., 2018). The latter paper suggests this limitation is due to increased scavenging rather than reduced regions of iron limitation. Both seem plausible explanations.

Response: Thank you for this discussion. We will add the following discussion about the uncertainty of upper limit of iron fertilization to the revised manuscript.

"The simulated upper limit of CO₂ reduction due to iron fertilization would not be a robust result because present iron models have large uncertainty. While Parekh et al (2008) show upper limit of 10 ppm, other simulations show CO₂ decrease by more than 20 ppm (Oka et al., 2011; Muglia et al., 2017). To obtain a deeper understanding of the impact of iron fertilization on glacial CO₂ decrease, the variability of upper limit among iron models should be investigated in the future study."

Minor and technical comments

#1

Line 16-17: I suggest to remove "(e.g. more sluggish ocean circulation)" because no such attribution was done in the paper. Khatiwala et al. suggest no CO2 effect from ocean circulation changes.

Response: According to the reviewer's comment, we will remove this part.

#2

Line 17-18: I suggest to remove "enhanced efficiency of the biological pump" here for the above mentioned reasons.

Response: In the revised text "by enhanced efficiency of the biological pump" will be changed to "by iron fertilization and an increase in whole ocean nutrient inventory".

#3

Line 21: this sentence is awkward. I suggest to rephrase to "glacial deep water was a more severe environment for ... than the modern ocean."

#4

Lines 24, 26: again, I'd suggest to rephrase to avoid using the term "biological pump" because it has not been quantified how much CO2 change was due to biological pump changes. Perhaps better to use "iron fertilization and/or global nutrient increase".

Response: Thank you for pointing out. Following reviewer's comments, we will correct these two sentences.

#5

Line 31: the biological pump also includes the CaCO3 pump

Response: We will change "the biological pump" to "the soft-tissue biological pump".

#6

Lines 50-51: consider including Schmittner and Somes (2015) and Somes et al. (2017)

Response: Suggested reference will be added in the revised manuscript.

#7

Lines 51-52: Khatiwala et al. have explored oxygen changes in more detail

Response: We will refer the results of Khatiwala et al. if their manuscript will be accepted during the revision of our manuscript.

#8

Line 83: see above comments on "biological pump"

Response: "enhanced efficiency of biological pump associated with" will be removed in the revised manuscript.

#9

109-110: iron solubility is modified by transport in the atmosphere. This leads to increasing solubility at lower concentrations. This effect has been considered in Muglia et al. (2017; their Fig. 2). This suggests using a constant solubility is not correct. This should be discussed.

Response: We agree that a constant solubility is not correct, as was described in lines 121-123. We will add the discussion about iron solubility to the revised manuscript, as follow.

"Present observation shows generally lower Fe solubility at higher Fe concentration in aerosols and higher solubility at lower concentration. Thus, assumed constant iron solubility at 2% in all types of dust could lead to overestimation of total DFe flux from different types of Fe-containing aerosols in LGM (Muglia et al., 2017). On the other hand, much higher Fe solubility (1–42% of Fe solubility) is measured for the LGM aerosols in Antarctica (Conway et al., 2015), suggesting that assumed constant iron solubility at 1% for all types of dust could lead to underestimation of DFe flux in LGM."

#10

116-119: This is about a factor of 10 increase in the 3% experiments. Compare with Muglia et al. (2018) who only have a factor of 4 increase in their best fitting model, which is constrained by d15N and d13C data.

Response: We will add following sentences to line 118.

"This value is roughly 10 times larger than in PI simulation and is larger than a recent estimation, which suggest that quadrupling of global DFe flux is constrained by model proxy comparison of δ¹⁵N and δ¹³C (Muglia et al., 2018)."

#11

129-130: Muglia et al. (2017) shows the sea level effect to be important.

Response: "Muglia et al (2017) show this effect causes CO₂ increase by 15 ppm." will be added after lines 129-130.

#12

General comment on section 2: how was the effect of sea level lowering on benthic denitrification treated? Somes et al. (2017) show that this effect reduces N loss in the LGM ocean and leads to a larger N inventory.

Response: Benthic denitrification is not considered in our model. We will add this information to line 150, as follow.

"In our simulations, changes in benthic denitrification is not considered. Somes et al (2017) show that decrease in benthic denitrification due to sea level drop reduces NO₃ loss and thus leads to a lager NO₃ inventory in the LGM ocean."

#13

165: delete: "because dust deposition flux of the Southern Ocean is underestimated in LGM_dust"

#14

166: delete "in the" and "with iron limitation"

#15

167: delete "in the"

#16

182-184: see above comment on new data from the S. Pacific

Response: As mentioned above, we will revise lines 182-188 and compare our model with new data of the Southern Pacific.

#17

199-201: see above comments on biological pump. I doubt that this conclusion is true because of the use of the AOU approximation here, which compromises the results.

Response: As mentioned above, we will add the annotation about errors of AOU approximation.

#18

239: replace "is the one" with "may be one of the". Or even better remove this whole part due to my above comments.

Response: "is the one" will be changed to "may be one of the".

#19

243: typo: "whehre"

Response: Thank you for pointing out. We will fix typo in the revised manuscript.

References:

Kohfeld, K. E., Graham, R. M., de Boer, A. M., Sime, L. C., Wolff, E. W., Le Quéré, C., and Bopp, L.: Southern Hemisphere westerly wind changes during the last glacial maximum: Paleo-data synthesis, Quat. Sci. Rev., 68, 76–95, 2013.

Parekh, P., Joos, F., and Muller, S. A.: A modeling assessment of the interplay between aeolian iron fluxes and ironbinding ligands in controlling carbon dioxide fluctuations during Antarctic warm events, Paleoceanography, 23, Pa4202, doi:10.1029/2007pa001531, 2008.