Dear Editor

We are grateful to you and the three reviewers for the helpful comments on the previous version of our manuscript. We have addressed all the comments made by the three reviewers, as follows. Generally, we revised the manuscript and conducted additional experiments (abiotic experiments) according to reviewer's comments.

5 This is because the comments of all reviewers are very useful for improving our manuscript and strengthening the interpretation of our model results. Native speaker has performed proofreading of our manuscript and corrected errors and inappropriate expression in English sentences We hope that the revised version of our paper is now suitable for publication in Biogeosciences.

10 Response to Reviewer #1 (N.C. Swart) (Our response highlighted gray.)

General comments

This paper seeks to understand the source of carbon climate feedbacks arising in the ocean on multi-centennial timesscales. This is an important question in the Earth System Modelling community, including for understanding future climate change, and interpreting carbon budgets. The authors use a well thought out experimental design to quantify the sensitivity of different aspects of the ocean carbon cycle (e.g. biology, circulation, solubility etc) to climate change. The approach is based on previous work, but fairly novel in this particular application. The paper is well organized and written, and the results, including the graphics are clear. Most uncertainties are addressed and the results are placed in the context of previous work. I thoroughly enjoyed this paper. Almost every time I had a question it was answered in the follow sentence or section. Overall I assess the quality as very high, and I recommend publication. I don't have any major issues. I do have some comments which I think could help to clarify the paper and address the few lingering questions that I did have.

Response: We appreciate the positive evaluation and many thoughtful comments from the reviewer. Referring to the comments, we will carefully revise the manuscript. Specific replies are as follows.

30 Specific comments

- The authors describe a decreasing ocean CO2 uptake under global warming, and attribute this in large part to a reduction in export production. However previous literature (e.g. de Vries et al. [2012], Marinov et al. [2008] and references therein) has shown that ocean CO2 uptake is not directly tied to export production (as one might guess), but rather to the so called "efficiency of the biological pump". Please clarify how export production,
- 35 biological pump efficiency and carbon uptake relate in this study. Specifically, is it really export production which is important and if so why is this different from the above literature?

Response: Thank you for a useful suggestion. In our simulation, globally averaged preformed PO₄ increases from 1.15 mmol/m³ in the pre-industrial condition to 1.40 mmol/m³ at the end of the simulation. Export production decreases from 8.1 PgC/yr to 6.3 PgC/yr. Considering the previous literatures pointed out by reviewer, the reduction of oceanic CO₂ uptake due to global warming would be attribute to decrease in biological pump efficiency rather than EP reduction in our simulations. We added the changes in biological pump efficiency to page 9, lines 7-11 in the revised manuscript (page 22, lines 7-11 in this response). We also described the importance of reduction in biological pump efficiency to decreasing CO₂ uptake under global warming in the abstract and conclusion

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• The authors force the offline ocean biogeochemical model with monthly mean fields from the AOGCM (including for insolation, velocity, temperature, salinity etc). This means that much variability is being averaged over, including the diurnal cycle, synoptic scale variability and so on. There is a known sensitivity of ocean model response to forcing frequency. Obviously forcing at a higher frequency means more data, and is more expensive. But please discuss how the results might be sensitive to the forcing frequency. I don't necessarily need any more experiments, just a clear caveat on this point.

Response: As mentioned below and manuscript, we compared passive salinity tracer in the offline model to online salinity in the AOGCM. There were no significant differences in the salinity distribution between two simulations. Therefore, the short-term processes have limited impact on our results. We added this information to page 5, lines 1-5 in the revised manuscript (page 18, lines 1-5 in this response).

• Circulation plays a small direct role, but a large indirect role through nutrient transport. The circulation changes are large (and mostly consistent with expectations). In various parts of the manuscript, the authors do a good job of comparing their results to those from CMIP and other studies. If possible it would be interesting to know how the MIROC simulated circulation changes under 4xCO₂ compare to other CMIP models. More generally a comment on how sensitive the results are to uncertainties, for example in the climate model response to increasing CO₂, would be helpful. (I note the authors do discuss the need for similar studies using different models, but the reasons for this could be fleshed out).

Response: According to the reviewer's comment, we compare our results to other AOGCMs and EMICs under high CO₂ scenario (e.g. 4xCO₂ and RCP8.5). The weakening of AMOC and AABW formation in the first 140 years of our simulation are consistent with the results of CMIP5 models under RCP8.5 (Weaver et al., 2012; Heuzé et al., 2015). However, the longer-term responses of AMOC and AABW formation are very uncertain. In our simulation, AMOC shutdown continues to the end of the simulation without recovery. Partial or full AMOC recovery to the pre-industrial level has emerged in other long-term AOGCM and EMIC simulations [Schmittner et al., 2008, Weaver et al., 2012, Li et al., 2013]. AABW formation recovers and overshoots after 1000 years in our simulations. These responses have not been reported in previous multi-millennium simulations [Schmittner et al., 2008, Li et al., 2013]. The uncertainties of circulation changes would have impacts on millennial-scale CO₂ uptake. We added the description of uncertainty of circulation change and its impact on long-term carbon cycle to page 16, lines 1-10 in the revised manuscript (page 29, lines 1-10 in this response).

Technical comments and typos (by pg and ln) pg 1 / Abstract:

35 In 8: "accelerate an increase in CO2" - Is there really an "acceleration". I'm not sure that this is the right word. I think just "decrease oceanic carbon uptake and therefore increase atmospheric CO2 and global warming" would sound better and be more accurate.

Response: According to the reviewer's comment, we modified this sentence in page 1, lines 8-9 in the revised manuscript (page 14, lines 8-9 in this response).

In 14: "...first 140 years (at year 2000)" - the meaning of this because clear later when reading the methods, but this could be a little confusing in the abstract, because readers do not know at that point what experiment you are conducting. For example, on first reading I was thinking "calendar year 2000".

Response: We agree the reviewer's comment. Following the comment of reviewer #3, we changed from "at year 2000" to "after 2000 model years" in page 1, lines 15 in the revised manuscript (page 14, lines 15 in this response).

5 In 19: "...gradient of DIC substantially" - add a comma after "DIC"

Response: We added "and" instead of comma in page 1, line 19 (page 14, line 19 in this response).

ln 23-4: "uptake through natural carbon cycle" - suggest removing "natural carbon cycle". I don't think this is

Response: We removed this in the revised manuscript (page 14, lines 24, right column in this response).

pg 2:

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- 15 In 5-6: "...long-term evolution of climate systems with slow response times..." -> "...long term evolution of climate system components with a slow response time..." (i.e. there is only one climate system, which is made up of many components).
- Response: We revised this sentence following the reviewer's comment in page 2, lines 5-6 (page 15, lines 5-6 in this response).
 - In 10: "accelerating the rate of CO2 accumulation" again I'm not sure if "accelerating" is accurate? Maybe just "increasing CO2 accumulation in the atmosphere".
- 25 Response: We agree the reviewer's comment. We modified this sentence in page 2, lines 10-11 (page 15, lines 10-11 in this response).
 - In 13: "primarily alter"...delete "primarily". There are only the natural and anthropogenic CO2 cycles.
- 30 Response: We removed "primarily" in the revised manuscript (page 15, line 13, right column in this response).
 - In 15-16: Another good study to reference is Randerson et al. (2015). They show that ocean carbon feedbacks become larger than land carbon feedbacks, but only on very long time scales. There is a nice tie in with this work.
- Response: Thank you for the nice suggestion. We added Randerson et al. (2015) to the reference in page 2, line 12 (page 15, line 12 in this response).
 - In 15-20: I suggest mentioning here that you will explain later why those studies came to that conclusion (and are different from yours).
 - Response: We added these information to page 3, lines 7-16 (page 16, lines 7-16 in this response).
 - $\ln 25$ "However the contributions"...suggest deleting "However". This sentence is not really a continuation of the previous sentence.
- Response: We removed "However" in the revised manuscript (page 15, line 30, right column in this response).

In 26-27: There are no studies doing this breakdown for CMIP5?

Response: We do not know this kind of study using CMIP5. We added "To our knowledge," to this sentence in page 2, line 30 (page 15, line 30 in this response).

In 28 "with AOGCM" -> "with an AOGCM"

pg 3:

0 ln 3 "using AOGCM" -> "an AOGCM".

In 13 "with MIROC 4m AOGCM" -> "with the MIROC 4m AOGCM"

Response: Thank you for pointing out. We corrected these in page 3, line 1; page 3, line 4; page 3 line 25 (page 16, line 1; page 16, line 4; page 16 line 25in this response).

In 27-28 "according to AOGCM climate simulations" - I got what you meant, but this could be clearer. Maybe something like "following the physical evolution of AOGCM climate simulations", or "forced by output from AOGCM climate simulations".

Response: We changed the sentence to the latter one in page 4, lines 7-8 (page 17, lines 7-8 in this response).

ng 4

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In 11: "setting flux" -> "settling flux"

Response: Thank you for pointing out. We fixed typo in page 4, line 25 (page 17, line 25 in this response).

In 16-18: "we confirmed..." - I found this confusing. At the bottom of page 3, it says that salinity is specified from the AOGCM simulations - but here you are saying you are using salinity from the offline simulation to validate against the AOGCM simulation. Something is missing. Do you simulate a passive salinity tracer in the offline model, to compare against the "online" salinity in the AOGCM? Please clarify.

Response: As reviewer said, we compared passive salinity tracer in the offline model to online salinity in the AOGCM. We added this description to page 5 lines1-5 (page 18, lines 1-5 in this response).

In 25-31: Just noting that the comparison is between a pre-industrial simulation and modern observations. This could have some impact. Are you using GLODAP estimated PI DIC and ALK to compare against? Not a big deal but worth clarifying.

Response: We compared a pre-industrial simulation with modern observations. We added this information to page 5 lines 15-16 (page 18, lines 15-16 in this response).

pg 5:

In 5-7 "This model does not include..." - it seems like these sentences belonged in section 2.2 to me. They are about the model, not the experiment.

Response: We moved this sentence to section 2.2 (page 4, lines 29-31) (page 17, lines 29-31 in this response).

In 9 "We conducted additional experiments"...these were only run for 500 years, right? Maybe worth mentioning here.

Response: The reviewer is right. We added this information to page 6, line 4 (page 19, line 4 in this response).

In 9-20: It is mentioned briefly below, but I think it is worth mentioning clearly here at the outset that the experimental design assumes linearity of the feedbacks.

Response: We agree the reviewer's comment. We added the assumption of linearity of the feedbacks to page 6 line 11 (page 19, line 11 in this response).

- In 23: "and oceanic interior temperature and salinity". When I thought about the experimental design as far as I can tell these interior T and S values are not used for anything in the offline model for this particular experiment, since the organic matter cycle is specified. The SST is, I believe, still be specified as GW. If this is all true, I would just remove the mention of "interior T and S values", since it is not relevant, and could be confusing. If these values are used for something, please clarify.
- 20 Response: This is our mistake. Interior T and S are not used in the sensitivity experiments. We just remove the mention of "interior T and S values" in the revised manuscript (page 20, line 5, right column in this response).

pg 6:

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in 12: "after the summary of the global mean" - a bit confusing as written. Maybe "...and ocean biogeochemical variables. A full summary of the global mean changes is reported in..."

Response: We modified this sentence in page 8, lines 3-4 (page 21, lines 3-4 in this response).

pg 7:

0 ln 2 / fig 1 e: I suggest you add the line for wind stress at year 2000 to Fig 1e (most other panels in fig. 1 are showing a year 2000 result). It would be helpful to see the recovery.

Response: We agree the reviewer's comment. We added the line for year 2000 in the revised manuscript (Fig. 2e).

35 In 6-15: PO4 is shown, but what about NO3? More generally, the paper discusses export production in general, but does not mention how diazotrophs and "other" phytoplankton react?

Response: Global NO₃ at the surface also decreases by about 20%. Regional NO₃ changes are similar to the PO₄ changes. Diazotrophs and "other" phytoplankton increase slightly, which is consistent with previous study (Schmittner et al., 2008). Increase in "other" phytoplankton is caused by faster nutrient recycling due to seawater warming. We added these description to page 8, line 30 and page 9, line 14 (page 21, line 30 and page 22, line 14 in this response).

pg 8:

45 ln 6: "...during constant atmospheric CO2..." - I would include the year 140, as in "...constant atmospheric CO2 after year 140..." for clarity.

Response: We corrected this sentence in page 10, line 7 (page 23, line 7 in this response).

In 27-33: I was interested in this section, and would like to see more spatial information. If possible, it would be really nice to see a Hovmoller, like Fig 1a, but for CO2 uptake/flux anomaly (of GW - CTL) (maybe in the SI).

Response: We agree the reviewer's comment. We added a Hovmoller figure to the supplementary figure 4.

pg 9:

10 ln 23-24: I suggest you reference these "uptake change" numbers back to table 2.

Response: We added the reference to table 2 to page 12 line 7 (page 25, line 7 in this response).

pg 10

15 In 19-23: Le Quere et al 2008 claim that the westerly wind increase is reducing Southern Ocean CO2 uptake (i.e. the opposite of what is being said here). Therefore, it is strange to cite as evidence without further explanation. I suggest it would be better to reference the Zickfeld et al. response to Quere et al. (who show that the CO2 uptake response to wind changes is time-scale dependent). The effect of circulation change on sDIC (Fig 5) is essentially a redistribution of carbon from the Atlantic to the Pacific. Interestingly, we saw a similar redistribution due to to wind stress induced circulation changes in Swart et al. (2012), which we linked back to changes in the Agulhas leakage and overturning circulation.

Response: Following the reviewer's comment, we will delete the reference to Le Quere et al 2008. We also added the description of carbon redistribution due to wind stress induced circulation changes to page 11 lines 25-27 (page 24, lines 25-27 in this response).

Figures

1. e : please add line for year 2000

- 30 Response: We added the line for year 2000 in the revised manuscript (Fig. 2e) as mentioned above.
 - 3. The colorbar is not perceptually uniform, which makes it hard to determine where large changes have actually occurred. Please consider using a perceptually uniform colorbar.
- Response: We changed the color bar in the revised manuscript (Fig. 4).
 - 6. Caption "Global upper-ocean" fix typo

Response: Thank you for pointing out. We fixed typo in page 28, line 2 (page 41, line 2 in this response).

Reference

Heuzé, C., Heywood, K. J., Stevens, D. P., and Ridley, J. K.: Changes in global ocean bottom properties and volume transports in CMIP5 models under climate change scenarios, J. Climate, 28, 2917–2944, doi:10.1175/JCLI-D-14-00381.1, 2015

Weaver, A. J. Sedlá cek, J., Eby, M., Alexander, K., Crespin, E., Fichefet, T., Philippon-Berthier, G., Joos, F., Kawamiya, M., Matsumoto, K., Steinacher, M., Tachiiri, K., Tokos, K., Yoshimori, M., and Zickfeld, K.:

Stability of the Atlantic meridional overturning circulation: a model intercomparison, Geophys. Res. Lett., 39, L20709, doi:10.1029/2012GL053763, 2012.

Response to Reviewer #2 (Our response highlighted gray.)

The manuscript by Yamamoto et al explores through a large suite of experiments under fixed atmospheric concentrations the role physical changes in climate play on ocean carbon uptake. Their conclusions suggest, in contrast to other papers, that the change of circulation dominate the response. It took me a little while to get into this paper, but once there I enjoyed the paper much and really appreciate the larger number of simulations that went into this work - thank you. Overall this is well conceived and executed piece of work, that will be of interested to a wide readership. I do have some minor comments that I feel once addressed would strengthen the paper, otherwise I am happy to recommend this paper for publication.

Response: We appreciate the positive evaluation and helpful comments from the reviewer. Referring to the comments, we will carefully revise the manuscript. Specific replies are as follows.

20 Minor Comments:

1. The authors predicate the study on global warming, and state that global warming will decrease ocean carbon uptake. However in the present day, as CO2 levels continue to rise - the ocean will take up carbon at a rate proportional to this i.e. gradient driven. I do understand in this study, if we assume fixed CO2 levels then this supposition is correct, but I do think this needs to clarified in the text.

Response: We can't understand this comment. Regardless of fixed CO₂ or continuous CO₂ rise, global warming increases surface pCO₂ due to seawater warming, leading to decrease in pCO₂ gradient and thus oceanic carbon uptake.

- 30 2. The study puts more heat and carbon into the ocean over a much shorter period than under CMIP3/5 change changes runs, even the business-as-usual scenario; this of course has implications for where the heat and carbon are stored. As the authors make a number comparison to these climate change runs could they comment on what the implications of this maybe perhaps on the timing of events e.g. sinks to sources etc, and whether its a fair comparison?
 - Response: As reviewer pointed out, input of heat and carbon into the ocean during the first 140 year of our experimental design are larger than SRES A2 and RCP8.5. In the first 140 year, the response of climate and oceanic carbon cycle would be somewhat different from the RCP8.5 simulations. After year 140, the influence of the initial differences of heat and carbon input on oceanic carbon cycle would weaken since CO₂ concentration is similar between 4xCO₂ and RCP8.5. Therefore, we think that the differences between 4xCO₂ and RCP8.5 have a limited impact on long-term response of climate and carbon cycle to global warming. These informations were added to page 5, lines 25-30 in the revised manuscript (page 18, lines 25-30 in this response).
- 3. The experimental methods section is super critical to this paper, however I needed to read this at least 5 times to be really clear. I recommend that the authors break up the 3rd paragraph to make it more accessible

Response: We break up the 3rd paragraph (page 7 in the revised manuscript, page 20 in this response).

4. The study uses offline simulations, which make sense, could the authors comments on whether on or offline makes much difference - given the challenges of capturing short-term processes in the fields needed to run the model. I am sure that they have tested this somewhere, and if not it should be acknowledged.

Response: The reviewer is right. We compared passive salinity tracer calculated in the offline model to online salinity in the AOGCM. There were no significant differences in salinity distribution between the two simulations. We added this information to page 5, lines 1-5 in the revised manuscript (page 18, lines 1-5 in this response).

- 5. The timescales calculated in the paper are based on a fixed atmospheric concentrations. In the real world i.e. driven by emissions, the ocean carbon uptake would significantly slow as the gradient between the ocean and atmosphere decreases. I think this probably needs to be mentioned in the discussion, as do the implications for timing of changes.
- Response: As reviewer pointed out, our simulation with prescribed CO₂ concentrations are idealized. On the other hand, there is an advantage of the simulations with prescribed CO₂ concentrations compared to the simulations with prescribed emissions. The simulations with prescribed CO₂ concentrations allow for a more rigorous separation of feedback processes since carbon sinks respond to the same atmospheric CO₂ concentration in all simulations (Zickfeld et al., 2011). We mentioned the difference between emission driven runs and concentration driven runs and usefulness of concentration driven runs in page 5 line 32 to page 6 line 2 (page 18, line 32 to page 19 line 2 in this response).
 - 6. Otherwise some minor typos etc need to be addressed, but I am sure they will be picked in the proofs.

Response: We carefully corrected typo and errors in the revised manuscript. Native speaker has performed proofreading of our manuscript and corrected errors and inappropriate expression in English sentences.

References:

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- Zickfeld, K., Eby, M., Matthews, H.D., Schmittner, A., and Weaver, A.J.: Nonlinearity of carbon cycle feedbacks, J. Climate, 24, 4255–4275, 2011.
- 35 Response to Reviewer #3 (Our response highlighted gray.)

The authors study long term ocean carbon cycle feedbacks over a time horizon of 2000 years by using an offline ocean biogeochemistry model driven by climate model output. By using different combinations of output fields from a control simulation (no global warming) and a global warming simulation, they separate the carbon cycle feedback into components originating from SST-changes, circulation changes, changes of the biological pump, and a few others. They find that changes in the biological pump contribute most to the carbon uptake reduction under climate change followed by solubility changes. The authors claim that this finding is "contrary to most previous studies".

The manuscript is clearly within the scope of Biogeosciences. The main conclusions, however, are partly inconsistent and not well enough supported by the results. Also, the manuscript as it stands now, it is not very novel. Many similar studies on ocean carbon-climate feedbacks have been published during the past 20 years, most of them with simpler models. However, the authors do not convincingly make the point as to why significantly different results could be expected because of enhanced model complexity. There are (or potentially are) interesting new aspects in the present study, but the authors do not elaborate these (see below).

Response: We are grateful for the careful review. The reviewer's comments helped us to improve our manuscript. Referring to the comments, we will carefully revise the manuscript. Specific replies are as follows.

1)The statement that the results are "contrary to most previous studies" is not convincingly supported by the results presented in this manuscript. Since the experimental set-up is different from (most of the) previous studies, it remains unclear what the effect of these differences might be. This is briefly discussed in section 5, following speculations (page 12, lines 20-30) about why models in previous studies possibly gave different results. These speculations are not convincingly supported by the results or the cited literature either. In my opinion it is most likely that differences in the experimental set-up explain much of the differences. The authors follow Zickfeld et al. (2008) in designing their experiments, and use the tendencies of DIC and ALK due to biological production/remineralisation from the CTL-experiment in the GW-experiment (and vice versa) to determine the effect of biology on CO2 uptake. This mimics pre-industrial organic matter and CaCO3 production/remineralisiation under a reduced circulation. I find this design questionable, since it weakens the upward transport of remineralised carbon and nutrients (leading to enhanced C-uptake), but at the same time keeps the export production at pre-industrial levels (leading also to enhanced uptake). The experiment design used in some of the previous studies (Joss et al. 1999, Plattner et al. 2001) is different: Here, archived preindustrial surface sPO4 and sALK fields are used in a global warming simulation to separate the "effect of biology". If I am not mistaken, the effect of reduced upwelling of DIC is cancelled out in this experiment. Other studies (Sarmiento et al. 1998, Matsumoto et al. 2010) use abiotic experiments.

Response: As for the upward transport of remineralized carbon and nutrients, we think that the lack of our explanation of experimental design misled the reviewer. To quantify the effect of biology on CO₂ uptake, we compared the GW-experiment (GW bio) to the GW-experiment with biological production/remineralization from the CTL-experiment (GW_bio_om) ((2) – (9) in Table 2). In the GW_bio, the upward transport of remineralized carbon and nutrients are weakened by both circulation change and the reduction of remineralization. In the GW_bio_om, the upward transport of remineralized carbon and nutrients are weakened by only circulation change since biological production/remineralization are kept at pre-industrial levels. Comparing the GW_bio_om to the GW bio, the upward transport of remineralized carbon and nutrients are enhanced, leading to reduced carbon uptake. We added the experimental details to Page 7, lines 11-13 in the revised manuscript (page 20, lines

In order to demonstrate that the feedback-mechanisms are really substantially different from previous studies, the authors would need to quantify the differences arising due to the different experimental set-up (or different interpretations of the "biological effect"). This could be done by running additional sets of experiments following the design of previous studies. A discussion of which experimental set-up or definition of "biological pump contribution" is more useful or correct should also be provided. The authors state in the abstract that "quantifications of the contributions from different processes to the overall reduction in ocean uptake are still

unclear". Instead of adding to the confusion they could take the opportunity to assess what the experimental setup in different studies contributes to this. This would also be a novel and useful contribution to the field.

Response: We agree the reviewer's comments. In fact, before submitting the original manuscript, we conducted the abiotic experiments following the previous studies (Sarmiento et al. 1998 and Matsumoto et al 2010) (attached Table 1 and 2). In our abiotic experiments, the reduction in oceanic CO₂ uptake associated with global warming is caused by changes in the ocean circulation and SST (attached Figure 1). Biological effect slightly enhances CO₂ uptake. These contributions of individual mechanisms are consistent with previous estimation using abiotic experiments (Sarmiento et al. 1998 and Matsumoto et al 2010). Therefore, the different results are caused by the differences of experiments set-up, as pointed by reviewer.

We added the methods of abiotic experiment and it's results to Section 2.4.1 (page 6), Section 4.1 (page 11), Fig. 1, Fig. 4, Fig. 5, Table 1 and Table 2. We also discuss the reason for the different estimations between the abiotic and CTL-base/GW-base experiments in Section 4.3 (page 14-15).

Unfortunately, due to computer resources, it is difficult to conduct additional experiments based on other previous studies (Joss et al. 1999, Plattner et al. 2001) immediately. However, we believe the comparison of two different experimental set-up (CTL-base/GW-base and abiotic experiments) is useful for understanding of the feedback mechanisms.

In this context it would be also useful to discuss the limitations of the approaches to separate the feedback-mechanisms in a non-linear system. The authors have already performed two sets of experiments (GW-base and CTL-base), which could serve this purpose. Results from these sets of experiments are presented in Table 2, but are only mentioned in one brief sentence (page 9, lines 24-25) in the manuscript. Particularly, for the "Biology"-contribution, the authors find a considerable dependence on the base state (GW or CTL; 118 PgC difference while the total is 402 PgC). An explanation for this would be useful. Do the authors expect that the individual contributions would add up to the total, and is the residual given in Table 2 thus an indicator of non-linearity?

Response: According to the reviewer's comment, we added the explanation of non-linear response of biological effect in CTL-base/GW-base experiments to Page 14, lines 4-10 (page 27, lines 4-10 in this response).

2) The authors state towards the end of the introduction section that the "second purpose of this study is to investigate the usefulness of EMIC for long-term simulations of the ocean carbon cycle by comparing our results to previous studies." This sounds like "EMIC" would be a well defined class of models with homogeneous properties, which is not the case. Some of the cited EMICs (e.g. Zickfeld et al. 2008) employ a 3d state-of the art ocean model, which is not fundamentally different from the ocean model used in this study. The authors do not discuss sufficiently why specific feedbacks could be expected to be present in their model but not in a simpler model. They also do not provide an in depth comparison of their results with previous EMIC studies (which I would expect for an issue that is the "second purpose of this study"). I actually do not belive that the question as to the "usefulness of EMIC for long-term simulations of the ocean carbon cycle" could be answered in this study - this would require a dedicated model intercomparison study with a common experimental design. I would recommend to drop this "second purpose", and discuss results compared to previous EMIC studies as necessary to place the present study in the scientific context. Further, the conclusions regarding the "usefulness of EMIC" are inconsistent. On page 9, lines1-2, it is stated that "results support the usefulness of EMIC for long-term projections of the ocean carbon cycle". Later in the "Summary and Discussion" it is speculated about why the

simpler models used in previous studies would have significantly different feedback mechanisms. Should this be interpreted as "simpler models are right for the wrong reason, but this is still useful"?

Response: We agree the reviewer's comment. We removed this "second purpose", and speculation about different feedback mechanisms in the simpler models in the "Summary and Discussion". We simply compared our results with previous EMIC studies in the revised manuscript.

Minor points

10 page 1, line 14: at year 2000 -> after 2000 model years

Response: We modified this in page 1, lines 15 (page 14, line 15 in this response).

page 1, line 22: "...circulation change becomes a second order process." This is in contradiction to the statement that "changes in the biological pump via ocean circulation" is the dominant process.

Response: We rewrote this sentence to "circulation change plays a small direct role, but a large indirect role through nutrient transport and biological processes." in Page 1, lines 23-24 (page 14, lines 23-24 in this response).

20 page 2, line 4: "...over a 1000-year period" -> "on millennial time scales" or similar

Response: We modified this in page 2, line 4 (page 15, line 4 in this response).

page 2, lines 16-18: "In those previous studies...". This assertion is not correct. E.g. Maier-Reimer et al. 1996 state that both biological and physical carbon-climate feedbacks are small compared to the carbon concentration feedback. I do not think that the other cited studies make the point that biology is a second order process (but I have not checked in-depth).

Response: We removed the reference in page 2, lines 14-16 (page 15, lines 14-16 in this response).

page 4, line 11: setting -> settling (?)

Response: We fixed typo in page 4, line 25 (page 17, line 25 in this response).

35 page 4, line 22: "As for spin-up,..." -> "For the spin-up..."

Response: Following the reviewer's comment we modified this in page 5, line 9 (page 18, line 9 in this response).

page 5, line 9: an -> the

Response: We corrected this in page 6, line 4 (page 19, line 4 in this response).

page 5, lines 22-29: It should be made clearer here which effect is included in which experiment. E.g., the authors state that the experiments GW_om and GW_ca "evaluate the contributions of changes in the organic matter and
 CaCO3 cycles." This is not very precise, since these experiments evaluate changes in one part of the "cycles"

only (changes in production and remineralisation rates, but the rate of upward transport of reminerelised OM is not included).

Response: In the comparison between GW_bio and GW_bio_om, the changes in upward transport of remineralized OM due to reduced OM remineralization are included as mentioned in the response to the major points 1). The changes in upward transport of remineralized OM due to circulation change are calculated in the comparison between GW_bio and GW_bio_circ. According to the reviewer's comment, we added experimental details to page 7, lines 11-13 (page 20, lines 11-13 in this response).

10 page 5, line 30: "...are included in not..." check grammar

Response: Thank you for pointing out. We corrected this in page 7, line 20 (page 20, line 20 in this response).

page 5, line 31: I guess the pre-industrial sea ice fractions are used only in the gas exchange calculations? Please clarify.

Response: The reviewer is right. We clarified this sentence in page 7, lines 18-20 (page 20, lines 18-20 in this response).

20 page 6, line 8: "... are likely to reflect the non-linearity..." Please describe what the experiments reflect. There is no need to speculate ("likely").

Response: We removed "likely" (page 20, line 30, right column in this response).

5 page 7, line 11: "According to..." -> "Consistent with the..."

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Response: We corrected this in page 9, lines 1-2 (page 22, lines 1-2 in this response).

page 7, line 18: "...rain ratio increasing from 0.09 to 1.13..." Please check the numbers.

Response: We changed from 1.13 to 0.13 in page 9, line 20 (page 22, line 20 in this response).

page 7, lines 16-17: Please explain briefly why PP increases and export decreases. It is not obvious from the model description why this could happen (if necessary or helpful, please amend the model description accordingly)

Response: PP increase are also found in Schmittner et al (2008) and Taucher and Oschlies (2011). Our model is based on Schmittner et al (2008). Taucher and Oschlies (2011) show that PP increase is caused by temperature effects on biological processes such as remineralization and the microbial loop. We added these information to page 9, lines 13-16 (page 22, lines 13-16 in this response).

page 8, line 5: "of the same simulation using models..." -> "of the corresponding simulations"

Response: We corrected this in page 10, line 5 (page 23, lines 5 in this response).

page 12, line 33: Plattner et al. 2001 do have abiotic experiments, but they do not use this to quantify the contribution of biology

Response: We deleted Plattner et al. 2001 in the reference of abiotic experiments.

Figure 3: a separation into panels for surface and deep ocean would be useful (or a stretch of the depth scale in the upper 1000m)

Response: According to the reviewer's comment, we revised Figure 3 (Figure 4 in the revised manuscript).

Reference:

Taucher, J., and Oschlies, A.: Can we predict the direction of marine primary production change under global warming?, Geophys. Res. Lett., 38, L02603, doi:10.1029/2010GL045934, 2011.

Long-term response of oceanic carbon uptake to global warming via physical and biological pumps

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Abstract. Global warming is expected to significantly decrease oceanic carbon uptake and therefore increase atmospheric CO2 and global warming. The primary reasons given in previous studies for such changes in the oceanic carbon uptake are the solubility reduction due to seawater warming and changes in the ocean circulation and biological pump. However, the quantitative contributions of different processes to the overall reduction in ocean uptake are still unclear. In this study, we investigated multi-millennium responses of oceanic carbon uptake to global warming and quantified the contributions of the physical and biological pumps to these responses using an atmosphere ocean general circulation model and a biogeochemical model. We found that global warming reduced oceanic CO2 uptake by 13% (30%) in the first 140 years (after 2000 model years), consistent with previous studies. Our sensitivity experiments showed that this reduction is primarily driven by changes in the organic matter cycle via ocean-circulation change and solubility change due to seawater warming. These results differ from most previous studies, in which circulation changes and solubility change from seawater warming are the dominant processes. The reduction in the global efficiency of the oceanic biological pump, induced by circulation change and lower nutrient supply, diminishes the vertical DIC gradient and substantially reduces the CO2 uptake.

The weaker deep-ocean circulation decreases the downward transport of CO2 from the surface to the deep ocean, leading to a drop in CO2 uptake in high-latitude regions. Conversely, weaker equatorial upwelling reduces the upward transport of

drop in CO₂ uptake in high-latitude regions. Conversely, weaker equatorial upwelling reduces the upward transport of natural CO₂ and therefore enhances the CO₂ uptake in low-latitude regions. Because these effects cancel each other, circulation change plays only a small direct role in the reduction of CO₂ uptake due to global warming, but a large indirect role through nutrient transport and biological processes.

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1 Introduction

Since the beginning of the industrial era, global oceans have reduced the atmospheric CO₂ concentration and mitigated climate change by taking up approximately 25% of anthropogenic CO₂ emissions (Ciais et al., 2014). In contrast to terrestrial uptake, oceanic carbon uptake operates on millennial time scales (Plattner et al., 2008; Archer et al., 2009a; Zickfeld et al., 2013). Therefore, understanding changes in the ocean carbon cycle is crucial for predicting the long-term evolution of climate system components with a slow response time, such as ice sheets (Charbit et al., 2008), permafrost (Lawrence and Slater, 2005), and methane hydrate in continental margins (Archer et al., 2009b; Yamamoto et al., 2014) as well as atmospheric CO₂ concentration.

- Climate warming tends to reduce the oceanic uptake of CO₂ from the atmosphere, thereby increasing CO₂ accumulation in the atmosphere and causing further warming, a well-known positive climate-carbon cycle feedback (Cox et al., 2000; Dufresne et al., 2002; Friedlingstein et al., 2006; Randerson et al., 2015). Changes in CO₂ solubility due to seawater warming, ocean circulation, and the biological pump alter oceanic CO₂ uptake by affecting anthropogenic CO₂ uptake and altering the air sea balance of the natural CO₂ cycle. Most previous studies have shown that solubility change from seawater warming and circulation change are major contributors to the reduction in oceanic carbon uptake (Sarmiento et al., 1998; Joos et al., 1999; Matear and Hirst, 1999; Plattner et al., 2001; Matsumoto et al., 2010). In such previous studies, changes in the biological pump associated with ocean circulation change were regarded as a second-order process even though the biological pump plays a crucial role in the natural carbon cycle.
- 20 On the other hand, changes in the biological pump can potentially contribute significantly to a reduction in oceanic carbon uptake, considering that both previous and current generations of Earth system models (ESMs) show a consistent decrease in global mean export production between -6% and -20% by the year 2100 under strong climate warming (Steinacher et al., 2010; Bopp et al., 2013). In the natural carbon cycle, the biological pump maintains the surface-to-deep gradient in oceanic DIC, and therefore regulates the exchange of CO₂ between the atmosphere and the ocean. Model simulations suggested that
 a complete die-off of ocean life would lead to an atmospheric CO₂ increase of more than 150 ppm (Maier-Reimer et al., 1996; Sarmiento and Gruber, 2006). A significant reduction in CO₂ uptake associated with the weakening of the biological pump was found in Zickfeld et al. (2008) even though their investigation only focused on the uptake change due to the weakening of the Atlantic meridional overturning circulation (AMOC) by freshwater input.
- 30 To our knowledge, the contributions of both physical and biological effects to reductions in oceanic CO₂ uptake are not evaluated directly in the recent generation of coupled atmosphere ocean general circulation models (AOGCMs) and ocean biogeochemical models. Moreover, previous studies using Earth System Models of Intermediate Complexity (EMICs) show that global warming decreases oceanic CO₂ uptake continuously over a thousand years or more (Plattner et al., 2008;

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Schmittner et al., 2008; Archer et al., 2009a). These millennial-scale simulations are now feasible with an AOGCM due to increased computer power. Thus, we examined multi-millennium changes in oceanic carbon uptake due to global warming and the contribution of individual mechanisms to these uptake change using a series of global warming simulations conducted with an AOGCM and an offline ocean biogeochemical model. The methods applied here enable us to conduct multiple simulations of the ocean carbon cycle according to AOGCM climate simulations with low computational cost.

An important difference between most previous studies (e.g., Sarmiento et al., 1998; Joos et al., 1999; Matear and Hirst, 1999; Plattner et al., 2001; Matsumoto et al., 2010) and that of Zickfeld et al. (2008) is the experimental setup used to quantify the contribution of individual mechanisms to changes in oceanic carbon uptake. In the typical former method (Sarmient et al., 1998; Matsumoto et al., 2010), abiotic experiments (i.e., without a biological pump) were conducted to isolate physical processes from biological processes. Zickfeld et al. (2008) separated these two effects more directly. In their sensitivity studies, the archived source and sink terms for biological processes in pre-industrial climate experiments were replaced with those of global warming experiments. In our study, in order to assess the different feedback mechanisms arising from different experimental setups, we quantified the contributions of individual mechanisms to changes in oceanic carbon uptake based on both abiotic experiments and methods of Zickfeld et al. (2008). We then compared these results to discuss which experimental setup is more useful for quantification of feedback mechanisms.

The remainder of this paper is organized as follows. Section 2 introduces the models and experiment design. Section 3 describes the centennial- and millennial-scale changes in the climate and ocean carbon cycle. Section 4 presents the results of three sets of sensitivity studies that quantify the contributions of individual mechanisms to reductions in oceanic CO2 uptake. Here we also discuss causes for differing results arising from the different experimental setups before summarizing our research in Section 5.

2 Methods

2.1 Climate model

We used the global-warming experiments conducted by Yamamoto et al (2015) with the MIROC 4m AOGCM. This model is based on the MIROC 3.2 (K-1 model developers, 2004) that contributed to the Coupled Model Intercomparison Project phase 3 (Meehl et al., 2007), which was extensively cited in the Intergovernmental Panel on Climate Change Fourth Assessment Report. In the MIROC 4m, the coefficient of the isopycnal layer thickness diffusivity is set as 7.0×10^{-6} cm² s⁻¹ instead of the value of 3.0×10^{-6} cm² s⁻¹ used in the original MIROC 3.2. The atmospheric GCM has a horizontal resolution of T42 (~2.8°) and 20 vertical levels. The oceanic GCM has a zonal resolution of 1.4° and a spatially varying meridional resolution that is approximately 0.56° at latitudes lower than 8° and 1.4° at latitudes higher than 65° the resolution changes smoothly between these latitudes. The vertical coordinate is a hybrid of sigma-z, resolving 44 levels in total: 8 sigma-layers

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Previous studies using Earth System Models of Intermediate Complexity (EMICs) show that global warming decreases oceanic CO₂ uptake continuously over a thousand years or more (Plattner et al., 2008; Schmittner et al., 2008; Archer et al., 2009a). These millennial-scale simulations are now feasible with atmosphere-ocean general circulation models (AOGCMs) due to increased computer power. Therefore, we conducted multi-millennium global warming simulations and sensitivity experiments using AOGCM and an offline ocean biogeochemical model. The second purpose of this study is to investigate the usefulness of EMIC for long-term simulations of the ocean carbon cycle by comparing our results to previous studies. *

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near the surface, 35 z-layers beneath, and one bottom boundary layer (Nakano and Suginohara, 2002). The sea-ice component is based on a two-category thickness representation, zero-layer thermodynamics (Semtner, 1976), and dynamics using elastic_viscous_plastic rheology (Hunke and Dukowicz, 1997). A model spin-up has been performed under pre-industrial boundary conditions such as insolation and an atmospheric CO₂ concentration of 285 ppm.

5 2.2 Offline ocean biogeochemical model

The ocean carbon cycle was calculated by the MIROC-based offline biogeochemical model (Yamamoto et al., 2015) that enables us to conduct numerous sensitivity experiments of the ocean carbon cycle forced by outputs from AOGCM climate simulations with low computational cost. The horizontal and vertical resolutions are the same as in the MIROC 3.2. For the tracer calculation, the monthly output data of velocity, temperature, salinity, sea surface height, sea-surface wind speed, seaice fraction, and sea-surface solar radiation from MIROC are applied to an offline tracer scheme (Oka et al., 2011).

This biogeochemical model is a modified version of the nutrient–phytoplankton–zooplankton–detritus (NPZD) ecosystem model of Keller et al. (2012). The model includes two phytoplankton classes (nitrogen fixers and other phytoplankton), zooplankton, particulate detritus, nitrate (NO₃), phosphate (PO₄), dissolved oxygen (O₂), dissolved inorganic carbon (DIC), alkalinity (ALK), two carbon isotopes (¹³C and ¹⁴C), and an ideal age tracer. Constant stoichiometry relates the C, N, and P content of the biological variables and their exchanges to the inorganic variables (NO₃, PO₄, O₂, ALK, and DIC). Optimal uptake kinetics, which assume, a physiological trade-off between the efficiency of nutrient encounters at the cell surface and the maximum assimilation rate (Smith et al., 2009), are adopted for the nutrient limitation of the growth rate of phytoplankton. The maximum phytoplankton growth and microbial remineralization rates are assumed to increase with seawater temperature according to Eppley (1972).

The formulations of air₃sea gas exchange and carbon chemistry follow the protocols of the Ocean Carbon Cycle Model Intercomparison Project–Phase 2 (OCMIP2; http://www.ipsl.jussieu.fr/OCMIP/phase2/) (Najjar and Orr, 1999). The production rate of calcium carbonate (CaCO₃) is assumed to be proportional to that of particulate organic carbon (POC, CaCO₃:POC = 0.03) and the settling flux of particulate CaCO₃ in the water column decreases with an e-folding depth of 3500 m (Schmittner et al., 2008). Although iron cycling is not explicitly calculated in our model, we adopted the iron limitation for the phytoplankton growth rate using monthly dissolved iron concentration outputs from pre-industrial experiments conducted using the biogeochemical elemental cycling model (AOU_06 case in Misumi et al., (2013)). Thus, we cannot consider changes in iron cycling and their impact on ecosystems in the global-warming experiment. Although this model does not include a marine-sediment module, the reaction of the dissolved CO₂ with CaCO₃ in deep-ocean sediments only plays an important role at timescales of thousands of years or longer (Archer et al., 1998).

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As we forced the offline ocean biogeochemical model with monthly mean data from the AOGCM, a certain amount of climate variability was averaged over. To confirm the validity of the offline calculations and their sensitivity to the forcing frequency, we compared the passive salinity distribution in the offline model to online salinity distribution in the AOGCM. There were no significant differences in basin-scale distributions between the two simulations, suggesting that short-term processes had limited impact on our results.

The biogeochemical model <u>was</u> initialized from the annual mean climatology <u>data</u> based on the World Ocean Atlas 2005 (WOA2005: Garcia et al., 2006) for dissolved NO₃ and PO₄ and Global Ocean Data Analysis Project (GLODAP: Key et al., 2004) for DIC and ALK. For the spin-up, the last 50 years of data in the MIROC pre-industrial experiment <u>were</u> cyclically applied to the offline ocean biogeochemical model. A model spin-up of more than 8000 years has been performed in order to eliminate the model drift in the global inventory of all tracers. In the last century of the spin-up run, the global net atmosphere ocean CO₂ exchange is 3.0 × 10⁻⁴ PgC year⁻¹, which is smaller than the OCMIP2 threshold of 0.01 PgC year⁻¹ (Orr, 2002). The resultant initial storage of ocean carbon (35,400 PgC), ocean net primary production (49.6 PgC year⁻¹) and export production (8.1 PgC year⁻¹) are comparable to observations and other estimates (Antoine et al., 1996; Schlitzer, 2000; Ciais et al., 2014). The simulated basin-scale distributions of nutrients (PO₄ and NO₃), DIC, and ALK <u>under pre-industrial conditions</u> are generally in agreement with the <u>modern distributions in the</u> WOA2005 and GLODAP datasets (Fig. S1). All physical and biogeochemical tracers, except salinity, have correlation coefficients with <u>observational data</u> of more than 0.85 and normalized standard deviation values between 0.7 and 1.1 (Fig. S2).

2.3 Global warming experiment

- 20 After the spin-up, the prescribed atmospheric CO₂ concentration for the climate model was increased by 1% per year from the pre-industrial value (285 ppm) until it reached 4 × CO₂ levels (1140 ppm) at year 140. This value was then held constant until reaching 2000 model years. Using the output of the global warming experiment conducted by the climate model, the offline biogeochemical model was integrated for 2000 years (GW_bio in Table 1).
- 25 The prescribed CO₂ concentration during the first 140 years of our experimental design are higher than the business-as-usual scenarios such as SRES A2 and RCP8.5, so the response of climate and oceanic carbon cycle would be somewhat different. After year 140, the influence of the initial difference would weaken because of the similar CO₂ concentrations between the 4 × CO₂ and business-as-usual scenarios. Therefore, the CO₂ differences between these scenarios would have a limited impact on the long-term response of climate and ocean carbon cycle to global warming. We compare our results with previous long-term simulations under the business-as-usual scenario in Section 3.

In the real world, the climate and carbon cycle change in response to CO₂ emissions; our simulation with prescribed CO₂ concentrations is thus highly idealized. However, there is an advantage to simulations with prescribed CO₂ concentrations as

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opposed to those with prescribed emissions: the former allow for a more rigorous separation of feedback processes since carbon sinks respond to the same atmospheric CO₂ concentration in all simulations (Zickfeld et al., 2011).

2.4 Sensitivity experiments

We conducted additional experiments for the first 500 years using the offline biogeochemical model (Table 1). In the constant-climate run (CTL_bio), the same prescribed CO₂ concentration as used in GW_bio but with the climate conditions of the MIROC pre-industrial experiment were applied to the offline biogeochemical model. This experiment relied on the same concept as the uncopled simulation in Friedlingstein et al. (2006), in which changing atmospheric CO₂ concentrations did not affect the radiation balance and therefore the climate. The total change in oceanic CO₂ uptake due to global warming is obtained from the difference between GW_bio and CTL_bio ("Total" in Table 2). We conducted three sets of experiments to total change in sea surface temperature (SST), ocean circulation, biology, freshwater flux, and sea ice, (Table 1) assuming that the individual contributions to total change are linear.

2.4.1 Abiotic experiments

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The first set (3–5 in Table 1 and "abiotic" in Table 2) are abiotic experiments, a typical method used in previous studies (Sarmient et al., 1998; Matsumoto et al., 2010). Following established methodology, the offline ocean biogeochemical model, which does not include a biological pump (no production and remineralization of POC and CaCO₃), is used in this first set. The abiotic offline biogeochemical model was spun up under the climate conditions of the MIROC pre-industrial experiment, as was the original offline biogeochemical model. After the spin-up, the surface-to-deep gradient of DIC decreased significantly (Fig. 1), as also shown in Sarmiento and Gruber (2006). The influence of the reduced DIC concentration in the ocean interior is discussed in Section 4.3.

CTL abio and GW abio are identical to CTL bio and GW bio, respectively, except that they are calculated using the abiotic model. Experiment GW abio SST is the same as GW abio, except that the solubility is calculated using the pre-industrial SST. The SST effect is calculated from GW abio minus GW abio SST, and the circulation effect is calculated from GW abio SST minus CTL abio ("SST" and "Circulation", respectively, in Table 2). The biological effect is calculated as the residual ("Total" – "SST" – "Circulation", as described in Table 2). In this set, the effects of changes in freshwater flux and sea ice coverage on changes in the CO₂ uptake are neglected, as in previous studies.

2.4.2 GW-base and CTL-base experiments

The second set of sensitivity experiments (GW-base) are based on Zickfeld et al. (2008). The experiments are based on GW_bio; however, each mechanism is replaced from GW_bio to CTL_bio (6-12 in Table 1). The contributions of individual mechanisms to the CO₂ uptake change are calculated from the difference between GW_bio and each experiment ("GW-base"

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Experiment GW_bio_circ is based on GW_bio; however, it uses the preindustrial ocean's vertical and horizontal velocities,

5 sea surface height, and vertical diffusivity. Note that the archived monthly mean data of the source and sink of DIC and alkalinity due to the biological pump (the organic matter cycle and the CaCO₃ cycle) in GW_bio are applied to the offline biogeochemical model. The CO₂ uptake sensitivity to ocean circulation change is isolated in this experiment.

Experiments GW_bio_om and GW_bio_ca are similar to GW_bio, except that the archived monthly mean data of the source

10 and sink of DIC and alkalinity due to the organic matter and CaCO3 cycles in CTL_bio, respectively, are applied to the

offline biogeochemical model. These two sensitivity studies have the same ocean circulation as GW_bio. Comparing

GW_bio to the GW_bio_om ("Organic matter cycle" of GW-base in Table 2), reduced export production decreases carbon

uptake but reduced remineralization decreases upward transport of remineralized carbon, leading to enhanced carbon uptake.

These experiments evaluate the contributions of changes in the organic matter and CaCO₃ cycles. The biological effect is obtained from the sum of these two effects. Note that changes in productivity due to stratification and circulation changes are included in these biological effects rather than in circulation effects.

GW_bio_si is similar to GW_bio, except that the preindustrial sea ice fractions in both hemispheres are used. Note that the pre-industrial sea ice fractions are used only in the gas exchange calculations. The effects of biological and circulation changes due to the sea ice fractions are not included in this experiment.

GW_bio_fw is identical to GW_bio, except that the freshwater flux of the pre-industrial experiment is applied to the offline biogeochemical model. This experiment evaluates the influence of changes in salinity, DIC, and alkalinity due to the hydrological cycle change on the CO₂ uptake.

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The contributions of individual mechanisms to the CO₂ uptake change are also estimated from sensitivity experiments under pre-industrial climate conditions. In the third set (CTL-base), following GW-base, sensitivity experiments are based on CTL_bio; however, each mechanism is replaced from CTL_bio to GW_bio (13-19 in Table 1). The contributions of individual mechanisms are obtained from the differences between each experiment and CTL_bio ("CTL-base" in Table 2). The differences in the estimated contributions between GW-base and CTL-base reflect the non-linearity of each mechanism to the CO₂ uptake change.

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3 Multi-millennium responses to global warming

3.1 Climate and ocean biogeochemical cycles

In this section, we briefly describe the regional changes in climate and ocean biogeochemical variables; a full summary of global mean changes was reported in Yamamoto et al. (2015). At the end of the global warming simulation, the global mean surface air and ocean temperature increase by 8.4 °C and 6.5 °C, respectively. The AMOC decreases from 16.5 Sv to 3.8 Sv in the first 500 years and does not recover by the end of the simulation. The strength of the Antarctic Bottom Water (AABW) formation decreases from 6.2 Sv to 2.4 Sv in the first 500 years. Thereafter, the strength of the AABW formation recovers and overshoots the pre-industrial condition after year 1000. These global mean changes are comparable to previous long-term simulations with EMIC and AOGCM (see Yamamoto et al. (2015) for a more detailed discussion).

At the time of CO₂ quadrupling (year 140), polar amplification occurs only in the Northern Hemisphere (Fig. 2a). The surface temperature over the Southern Ocean and Antarctica gradually increases, and polar amplification in both hemispheres is observed at approximately year 1000. This slow southern polar warming is related to deep-ocean heat uptake in the Southern Ocean. According to the slow deep-ocean heat uptake, the ocean interior is still warming up at the end of the simulations (Fig. 2b). Slow southern polar warming and ocean heat uptake have also been reported in previous multimillennial simulations conducted by ECHAM5/MPIOM (Li et al., 2013). In the ocean, greater warming of the upper ocean (above 2000 m) occurs in the Southern Hemisphere (Fig. 2c). This is because the drastically weakened AMOC and enhanced AABW formations cause the heat transport to be larger in the Southern Hemisphere than in the Northern Hemisphere (Fig. 2d). Weakening in AMOC reduces the heat transport from 1.8 PW to 0.8 PW across 30° N. By contrast, the enhanced AABW increases heat transport by 0.7 PW in the Southern Hemisphere.

Strengthening and poleward shifting of the subpolar westerlies occurs in the Southern Hemisphere (Fig. 2e), which is consistent with the observations and a robust feature of climate projections in global-warming experiments (Fyfe and Saenko, 2006; Meijers, 2014). A 12% strengthening and a 2.8° poleward shift of the maximum annual mean zonal wind stress occur at the time of CO2 quadrupling. Subsequently, the subpolar westerlies in the Southern Hemisphere are gradually weakened but are still slightly stronger than the pre-industrial conditions. This weakening occurs because the slow southern polar warming weakens the thermal gradient between the tropical and polar areas. The contributions of ocean-circulation change caused by strengthened westerlies to the ocean carbon cycle are discussed in Section 4.2.

30 Global PO₄ and NO₅ concentration at the surface decreases by about 25% and 20%, respectively. These decreases are attributed to reduced nutrient supply into the euphotic zone due to enhanced stratification and slower deep-ocean circulation.

As a result, the global export production decreases by 22% (see Fig. 1d in Yamamoto et al., 2015), which is comparable with CMIP5 models (7%=18% reduction in export production from the 1990s to 2090s for RCP8.5) (Bopp et al., 2013).

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Remarkable reductions in surface PO₄ concentration occurs in the tropical ocean and the North Atlantic (Fig. 2f). Consistent with this reduction, the largest declines in export production are also located around the equator and in the North Atlantic (Fig. 2g). The increases in export production are primarily observed in the Southern and Arctic Oceans. These increases are related to reductions in light limitation and/or increased growth rates due to rising temperatures as shown by Steinacher et al. (2010)

The efficiency of the oceanic biological pump is calculated following Ito and Follows (2005) as the biologically sequestered fraction of the total phosphate inventory, $P_{\text{remi}}/P_{\text{tot}}$. Here, P_{tot} is the total phosphate inventory and P_{remi} is remineralized phosphate concentration ($P_{\text{remi}}=AOU \times R_{\text{PO}}$), where AOU is the apparent oxygen utilization (AOU = $O_2^{\text{sat}} - O_2$), O_2 is the dissolved oxygen concentration, and R_{PO} is a constant phosphorous to oxygen ratio. $P_{\text{remi}}/P_{\text{tot}}$ decreases from its preindustrial value of 0.47 to 0.34 by the end of the simulation.

Global primary production increases by 2.5 PgC year¹ from the preindustrial value, which is consistent with previous studies (Schmittner et al., 2008; Taucher and Oschlies, 2011). Both diazotrophs and other phytoplankton increase slightly. Taucher and Oschlies (2011) showed that increases in primary production are caused by temperature effects on biological processes such as remineralization and the microbial loop.

Due to the constant production ratio of POC and CaCO₃ in the model formulation and the increase in the global primary production, CaCO₃ production is increased by 6%. The export of CaCO₃ also increases in spite of a reduction in the POC export, resulting in the rain ratio increasing from 0.09 to 0.13. Two distinct export responses are caused by faster and shallower remineralization of POC in a warmer ocean; however, the remineralization rate of CaCO₃ is independent of the seawater temperature in our model. The enhanced CaCO₃ production decreases (increases) alkalinity in the surface (deep) oceans as shown by changes in the salinity-normalized alkalinity (dashed lines in Fig. 2h). Reduced surface alkalinity, is also caused by the longer residence time of surface waters, allowing a more efficient biological utilization of the carbonate ions.

The enhanced surface-to-deep gradient of the salinity-normalized alkalinity and the alkalinity associated with changes in CaCO₃ cycling and ocean circulation has been reported in previous long-term simulations (Plattner et al., 2001; Schmittner et

Changes in the freshwater flux also alter the alkalinity. At low latitudes, enhanced evaporation concentrates the surface alkalinity. Dilution of the surface alkalinity by enhanced precipitation at high latitudes reduces the alkalinity in the deep ocean. As a result, changes in the freshwater flux causes weaker surface-to-deep alkalinity gradients. The effects of changes in the CaCO₃ cycling and freshwater flux cancel each other out, resulting in apparently slight changes in the surface-to-deep alkalinity gradient, in our model (solid lines in Fig. 2h). The effects of changes in the CaCO₃ cycle and freshwater flux to the CO₂ uptake change are quantitatively discussed in Section 4.1.

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3.2 Ocean carbon cycle

We first show the cumulative oceanic uptake of CO₂ and the reduction of oceanic CO₂ uptake due to global warming. At the time of CO₂ quadrupling (year 140), the cumulative CO₂ uptake is 567 PgC in the global warming run (GW_bio)_and 654 PgC in the constant climate run (CTL_bio), a projected reduction due to global warming of 13%. These values are close to those of corresponding simulations in the Coupled Model Intercomparison Project 5 (CMIP5, (Taylor et al., 2012)) in which the modeled mean CO₂ uptake is 613 PgC and the uptake reduction is 11% (Arora et al., 2013). Even though oceanic CO₂ uptake is decreased during constant atmospheric CO₂ after year 140, a new equilibrium is not yet reached by the end of the simulation. In the last decade of the experiments, the ocean still takes up approximately 0.31 PgC year⁻¹ due to the millennial timescale of the deep ocean ventilation (Key et al., 2004). At the end of the simulation, the cumulative CO₂ uptake is 2028 PgC and the uptake reduction due to global warming increases to 30% (Fig. 3a and Table 1). These values and the increasing trend of the uptake reduction are also comparable to the results of previous long-term EMIC simulations (Plattner et al., 2001; Schmittner et al., 2008).

The time evolutions of oceanic CO₂ uptake in each basin are shown in Figure 3b. During the first few decades, the Pacific Ocean is the dominant sink of atmospheric CO₂ because it possesses the largest area. After atmospheric CO₂ stops increasing, the surface Pacific waters are close to saturation and CO₂ uptake rapidly decreases because the deep Pacific waters are slowly ventilated. Conversely, the decrease in CO₂ uptake in the Southern Oceans is slower than that in the Pacific Ocean. Carbon transport from the surface to deep ocean via these well-ventilated waters causes continuous CO₂ uptake there. In the pre-industrial condition, the pCO₂ in the Southern Ocean is kept higher than the atmospheric pCO₂ owing to the upwelling of carbon-rich deep waters, resulting in a source of atmospheric CO₂. The Southern Ocean alters from a source to a sink of atmospheric CO₂ immediately after the atmospheric CO₂ increase. This phenomenon is consistent with observation-based reconstructions of present-day and pre-industrial air sea CO₂ fluxes (Gruber et al., 2009). After excess CO₂ is mixed throughout the deep ocean, the pCO₂ in the Southern Ocean again exceeds the atmospheric pCO₂. The Southern Ocean returns to being a source of atmospheric CO₂ at approximately year 1400. At the end of the global warming experiment, the Atlantic and Southern Oceans are the major sink and source basins of atmospheric CO₂, respectively, as in the pre-industrial condition.

We also investigate the regional differences in uptake reduction due to global warming. In the first 500 years, approximately half of the total uptake reduction occurs in tropical and subtropical regions between 32.5°S and 32.5°N (Fig. 3c. and s4).

The contributions of the high latitudes in the Northern and Southern Hemispheres have similar magnitudes. At a basin scale, the contributions of the Atlantic and Pacific Ocean have similar magnitudes and that of the Indian Ocean is approximately half those contributions. These regional differences have also been found in similar long-term simulations by Plattner et al. (2001).

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削餘: The global and regional changes of the oceanic CO2 uptake projected in this study using AOGCM and the offline ocean biogeochemical model are comparable to previous long-term simulations with EMIC. Therefore, our results support the usefulness of EMIC for long-term projections of the ocean carbon cycle.

Figure 4 shows the zonal mean distribution of the salinity-normalized DIC (sDIC) in the global warming run. In the first 500 years, changes in sDIC from the pre-industrial condition decrease from the surface to the deep oceans. Significant sDIC changes in the deep water are observed in the North Atlantic. The shallower CO2 invasion into the North Atlantic compared to the constant climate run (Fig. S3) is caused by the weaker and shallower AMOC. After year 1000, due to the enhanced AABW formation, AABW with a relatively high sDIC concentration intrudes into the deep North Atlantic below 2000 m. As a result, the largest sDIC change in the deep water occurs in the North Atlantic at the end of the simulations. The smallest sDIC change is found in the deep North Pacific in accordance with the millennial ventilation timescale.

4 Effects of individual mechanisms on the reduction in oceanic CO2 uptake associated with global warming

The decrease in cumulative CO₂ uptake due to global warming projected by our model is consistent with the previous centennial and millennial simulations using ESMs and EMICs. In this section, we disentangle the contributions of individual mechanisms to CO₂ uptake reduction using three sets of sensitivity experiments (Abiotic, GW-base, and CTL-base, Tables 1 and 2). To compare our results with previous studies, we focus on changes in the CO₂ uptake in the first 500 years. The total reduction in the cumulative CO₂ uptake until year 500 is 402 PgC ("Total" in Table 2).

15 4.1 Abiotic experiments

In the abiotic experiments, the reduction in oceanic CO₂ uptake associated with global warming is caused by changes in the ocean circulation and increases in the SST that reduce the global CO₂ uptake by 308 PgC and 140 PgC, respectively (Table 2). Weaker AMOC and AABW formations decrease the downward transport of CO₂ from the surface to the deep ocean, leading to a significant drop in the CO₂ uptake at high latitudes (Fig. 5a) and thus a reduction in sDIC in the deep ocean (Fig. 20 6a).

An increase in sDIC also occurs in the Antarctic intermediate water. As previously mentioned, the intensification of the Southern Hemisphere westerly winds enhances the mixing and overturning circulation of the Southern Ocean (Fig. 6a), increasing the CO₂ uptake and sDIC (Fig. 5a). An enhanced CO₂ uptake due to intensified westerly winds in the Southern Ocean has also been observed in previous studies using simple box models and coupled general-circulation models (Zickfeld et al., 2007; Matear and Lenton, 2008). A similar redistribution of carbon due to wind-stress-induced circulation changes are found in Swart et al (2012), in which the Agulhas leakage and overturning circulation are linked.

The contribution of the SST increase to CO₂ uptake diminishes continuously from south to north. Biological changes 30 enhance CO₂ uptake at high latitudes and reduce it at low latitudes; the resulting global CO₂ uptake increase by 46 PgC due to biological changes. These global and regional contributions of individual mechanisms are consistent with previous estimations using abiotic experiments and other methods (Sarmient et al., 1998; Matsumoto et al., 2010).

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4.2 GW-base and CTL-base experiments

4.2.1 Global change

The contributions of individual mechanisms to the uptake change estimated from the combination of the GW-base and CTL-base experiments are summarized in Table 2 and Figure 5b; averages of these experiments are used below. Changes in the organic matter cycle and in the solubility due to the SST increase are the dominant mechanisms for the global reduction in CO2 uptake due to global warming (Fig. 5b). By year 500, these changes decrease the global carbon uptake by 276 PgC and 151 PgC, respectively (Table 2). A significant nonlinearity in the uptake change (from -216 PgC to -336 PgC) is caused by the organic matter cycle. The ocean circulation change decreases the global CO2 uptake by 6.5 PgC.

Enhanced CaCO₃ production reduces the surface alkalinity and therefore leads to an increase in the surface pCO₂. Changes in the CaCO₃ cycle reduce the CO₂ uptake by 14 PgC. The reduced sea ice cover allows air_sea gas exchange over a larger area and leads to a 15 PgC increase in the CO₂ uptake, Dilution of the surface salinity by 1 psu leads to a pCO₂ reduction of approximately 4% (Takahashi et al., 1993) and an increase in the CO₂ uptake. Enhanced evaporation at low latitudes increases pCO₂ resulting in an uptake reduction, whereas enhanced precipitation at high latitudes increases uptake. The
 global contribution of freshwater-flux changes is an uptake reduction of 2.5 PgC.

These significant contributions of changes in the organic matter cycle to the uptake reduction due to global warming and the secondary importance of circulation changes are consistent with Zickfield et al. (2008), who investigated the reduction in the oceanic CO₂ uptake due to freshwater hosing and AMOC weakening. However, these results are different from most previous studies (Sarmiento et al., 1998; Joos et al., 1999; Matear and Hirst, 1999; Plattner et al., 2001; Matsumoto et al., 2010), in which ocean circulation was the dominant mechanisms for reductions in oceanic CO₂ uptake. Below, we investigate what causes these greater biological effects and smaller circulation effects in our global warming simulations.

4.2.2 Effects of circulation change

The effects of circulation change display large regional variations (Fig. 5b). Similar to the abiotic experiments, reductions in CO₂ uptake and sDIC in the deep ocean (induced by weaker deep ocean circulation) occur at high latitudes, and sDIC is enhanced by increased westerly winds in the Southern Ocean. On the other hand, CO₂ uptake at low latitudes is enhanced by ocean circulation. We attribute this increase to weaker upper-ocean overturning and tropical upwelling (Fig. 7a). The tropical upwelling across 200m decreases from 38 Sv to 22.5 Sv in the first 200 years and continues to the end of the simulation (Fig. 7b). Weakening the equatorial upwelling reduces the upward transport of natural CO₂ from the ocean interior to the surface.

The resulting enhanced CO₂ uptake at low latitudes increases sDIC in the subsurface water, especially in the Pacific Ocean (Fig. 5b). The importance of enhanced CO₂ uptake associated with weaker upper-ocean overturning is also suggested in decadal variability of oceanic CO₂ uptake (DeVries et al., 2017).

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Enhanced CO₂ uptake induced by weaker equatorial upwelling and increased westerly winds cancel out the reduced CO₂ uptake associated with stratification and the weaker deep ocean circulation. Therefore, the effects of circulation changes are responsible for only a small fraction of the total reduction in the CO₂ uptake in our simulations. In the first 200 years, increases in CO₂ uptake caused by weaker equatorial upwelling and strengthened westerly winds are the dominant processes in the effect of circulation change (Fig. 8). Subsequently, the effects of weaker AMOC and AABW formations are enhanced and overcome those of the strengthened westerly winds and weaker equatorial upwelling. As a result, the CO₂ uptake is reduced by the circulation changes after year 450.

4.2.3 Effects of changes in the biological pump

The reduced efficiency of the biological pump cannot maintain the vertical gradient of DIC between the surface and deep oceans. Excess DIC stored in the deep ocean by the biological pump is thus transported to the surface, increasing surface pCO₂ and reducing CO₂ uptake. Reduced CO₂ uptake occurs at both low, and the high latitudes in the Northern Hemisphere (Fig. 5b), according to changes in export production (Fig. 2g). Similarly, significant sDIC decreases occur in the North Atlantic, equatorial Indo-Pacific Ocean, and North Pacific (Fig. 9). These reductions in CO₂ uptake induced by the biological pump are consistent with previous model simulations quantitatively linking increases in atmospheric CO₂ concentration to reductions in the efficiency of biological pump (Ito and Follows, 2005; Marinov et al., 2008).

Subsequently, the effect of the reorganization of the biological pump on the reduction in CO₂ uptake is decomposed into the effects of the temperature dependence of production and remineralization and the change in nutrient supply via the circulation change. In our model, the production and remineralization rate doubles for every 10°C increase in the seawater temperature (Eppley, 1972). The warming-induced increase in production reduces the partial pressure of CO₂, therefore enhancing the oceanic CO₂ uptake. Enhanced remineralization of organic matter reduces the carbon transport into the deep ocean, leading to a lower CO₂ uptake. The lower nutrient supply due to stratification and slower thermohaline circulation decreases export production and therefore CO₂ uptake. The effect of the temperature dependence of the biological pump is calculated in GW-base and CTL-base ("Temperature for biology" in Table 2). The effect of the change in the nutrient supply is calculated as the residual ("Biology" – "Temperature for biology" as summarized in Table 2).

In the first 100 years, a warming-induced increase in production occurs more rapidly than an increase in remineralization because the surface waters are warming more rapidly than the subsurface. Therefore, the temperature dependence slightly increases the CO₂ uptake (Fig. §). Subsequently, the effect of the enhanced remineralization overcomes that of the enhanced production, therefore the temperature dependence decreases the CO₂ uptake, consistent with the findings of Matsumoto (2007). By, year 500, lower nutrient supply and temperature dependence decreases the CO₂ uptake by 241.5 PgC and 48.5 PgC, respectively (Table 2). Our results indicate that the reduction in the oceanic CO₂ uptake from biological change is

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削餘: Reduced nutrient supply into the euphotic zone due to circulation change and faster remineralization from seawater warming decrease the POC flux by 22% at a depth of 100m and 70% at a depth of 100 m (Fig. 1e and 8a). The weaker carbon transport by the biological pump

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mainly caused by reductions in nutrient supply via the circulation change, and that temperature effects on production and remineralization are not negligible.

The contribution of changes in the biological pump depends considerably on the base state (GW-base or CTL-base; 118 PgC

5 difference while the total is 402 PgC) and mainly causes the non-linearity of feedback-mechanisms. This nonlinearity would be caused by difference in ocean circulation between GW-base and CTL-base. The enhanced CO₂ uptake associated with reduced upward transport of remineralized carbon partly cancel out the reduced CO₂ uptake due to lower export production. This process is larger in CTL-base than in GW-base because of slower deep ocean circulation in the latter. As a result, the contribution of changes in the biological pump is smaller in CTL-base than in GW-base. These nonlinearities need to be properly accounted for when separating the contribution of individual feedback-mechanisms to CO₂ uptake change.

4.3 Assessment of differences between the abiotic and CTL-base/GW-base experiments

The contributions of changes in the ocean circulation and the biological pump to the reduced CO₂ uptake differ between the abiotic and CTL-base/GW-base experiments even when using the same model and scenario, as suggested in previous studies

(Sarmiento et al., 1998; Zickfeld et al., 2008; Matsumoto et al., 2010). Our results show that these different results are caused by the estimation methods used.

We attribute these different results to the vertical gradient of DIC under the pre-industrial condition of the abiotic and biotic experiments (i.e., CTL-base and GW-base) (Fig. 1). With regards to the circulation effect of the abiotic experiments, the enhanced CO₂ uptake associated with reduced upward transport of natural CO₂ from the deep ocean to the upper ocean is underestimated because the vertical DIC gradient in the abiotic experiments is much smaller than in the biotic experiments. The effect of circulation change in the abiotic experiments primarily represents reduced CO₂ uptake due to reduced downward transport of anthropogenic CO₂ from the surface to deep ocean. In the CTL-base/GW-base experiments with a realistic vertical DIC gradient, the effect of the circulation change represents both a reduced upward transport of natural CO₂ and a reduced downward transport of anthropogenic CO₂. Therefore, the reduction in CO₂ uptake due to weaker deep-ocean circulation is larger in the abiotic experiments than in the CTL-base/GW-base experiments. An increase in CO₂ uptake associated with weaker equatorial upwelling is found only in the CTL-base/GW-base experiments.

In the abiotic experiments, the biological contribution is calculated as the residual. The enhanced CO₂ uptake owing to the

reduced upward transport of natural CO₂ is included in the biological effect. This effect overcomes the reduced CO₂ uptake
due to the reduced efficiency of the biological pump, leading to an enhanced CO₂ uptake due to the biological change.

Therefore, the CTL-base/GW-base experiments are more useful for quantifying the contributions of circulation and
biological change. We conclude that changes in the organic matter cycle and increases in the SST are the primary

contributors to the reduction in CO₂ uptake due to global warming in our model. Although our experimental design differs from that used in some previous studies (e.g., Joos et al., 1999; Plattner et al., 2001), the identification of differences in feedback mechanism between all experimental designs is beyond the scope of this work and should be the subject of future research.

5 Summary

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In this study, we investigated multi-millennium changes in the ocean carbon cycle under a quadrupling of the atmospheric carbon dioxide using an AOGCM and an offline biogeochemical model. At the end of the simulation, the cumulative CO₂ uptake is 2028 PgC and global warming reduces the uptake by 30%, this primarily occurs in the tropical and subtropical regions. These projected global and regional changes in oceanic CO₂ uptake are similar to those in previous long-term simulations using EMIC (Platterr et al., 2001; Schmittner et al., 2008).

To isolate and quantify the individual mechanisms responsible for the reduction in the oceanic CO₂ uptake due to global warming, we conducted three sets of sensitivity experiments based on previous studies. Our results are consistent with the previous studies: in abiotic experiments, the primary contributors are changes in the ocean circulation and reduced solubility due to sea-surface warming (Sarmient et al., 1998; Matsumoto et al., 2010), while in the CTL-base/GW-base experiments, the primary contributors are changes in the organic matter cycle via ocean circulation change and sea-surface warming (Zickfeld et al., 2008). It is conceivable that the differences in results between these sensitivity studies are caused by the vertical gradient of DIC under the pre-industrial condition, i.e., smaller DIC gradients in abiotic experiments and realistic DIC gradients in CTL-base/GW-base experiments (Fig. 1). In the abiotic experiments, the circulation effects are underestimated with respect to enhanced CO₂ uptake associated with reduced upward transport of natural CO₂ from the deep ocean to the upper ocean. On the other hand, in the CTL-base/GW-base experiments, the effects of the circulation changes include both reduced upward transport of natural CO₂ and a reduced downward transport of anthropogenic CO₂. Therefore, the CTL-base/GW-base experiments are more useful for quantifying the individual feedback mechanisms.

The reduced CO₂ uptake associated with weaker deep ocean circulation and stratification is counteracted by the enhanced uptake due to weaker equatorial upwelling and strengthened subpolar westerly winds in the Southern Hemisphere. The reduction in the efficiency of the biological pump resulting from circulation change and the enhanced remineralization owing to seawater warming diminishes the DIC gradient between the surface and deep ocean, leading to a significant reduction in oceanic CO₂ uptake. Our results suggest that changes in the biological pump and solubility are the primary drivers of reductions in oceanic CO₂ uptake associated with global warming.

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Circulation plays a small direct role on changes in oceanic carbon uptake due to global warming, but a large indirect role through nutrient transport and the biological pump. On a centennial timescale, the weakening of AMOC and AABW formation in our simulation is consistent with the results of CMIP5 models (Weaver et al., 2012; Heuzé et al., 2015). Changes in oceanic carbon uptake and reductions in export production due to global warming are in close agreement with current simulations using ESMs, as mentioned in Section 3. Therefore, we speculate that similar contributions to the reduction in CO2 uptake from changes in ocean circulation and biological processes will be found in other models. On the other hand, the longer-term responses of AMOC and AABW are very uncertain. The recovery and overshoot of the AABW formation that occurred after 1000 years in our simulation have not been reported in the previous multi-millennium simulations (Schmittner et al., 2008, Li et al., 2013). To fully assess the robustness of millennial-scale reductions in oceanic

Although our results show, that, changes in the biological pump are, crucial for, reductions in, oceanic CO2 uptake due to global warming, several biological processes were, not considered in our model. A, reduction in the saturation levels of calcium carbonate in seawater associated with CO2 invasion into the ocean can have, adverse effects on marine calcifying organisms.

The subsequent reduction in CaCO3 production would increase the surface ocean alkalinity and therefore enhance, oceanic carbon uptake (Riebesell et al., 2000), but, its modelled impact has been very different in, previous, studies (Heinze, 2004; Gehlen et al., 2007; Ridgwell et al., 2007; Matsumoto et al., 2010). In addition, an, increase in biotic carbon-to-nitrogen drawdown in response to pCO2 changes can enhance, oceanic CO2 uptake (Oschlies et al., 2008). Reduced seawater viscosity in a warmer ocean accelerates particle sinking speed, therefore increasing the efficiency of the biological pump and enhancing oceanic CO2 uptake (Bach et al., 2012; Taucher et al., 2014). These effects remain poorly understood but could potentially have a large impact on oceanic carbon uptake and atmospheric CO2 concentration on a multi-centennial to millennial timescale. The inclusion of these processes in models may be required for a more comprehensive understanding of the response of the ocean carbon cycle to global warming.

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climate model and complexity of biogeochemical model. Our model has an increased atmospheric and oceanic resolutions, so we hypothesise it captures better weaker equatorial upwelling and enhanced mixing due to strengthened subpolar westerly wind in the Southern Hemisphere. These physical changes are not reported in the previous studies. Therefore, the effect of circulation changes i our simulation would be smaller than in the previous studies. These physical changes also affect the nutrient supply and production. Moreover, the temperature dependence of production and remineralization was not considered in most of previous studies using simple ocean biogeochemical models (such as nutrient-削除: the 削除: consistent 削除: to the reduction in CO2 uptake 削除: our results for the 削除: analyses 削除: Our study 削除: s **削除:** the **削除:** is 削除: the 削除: · however 削除: there are 削除: that are 削除: The 削除: has 削除: the 削除:: 削除: however

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Zickfield et al. (2008) in which the reduction in the oceanic CO₂

uptake due to freshwater hosing and AMOC weakening was investigated. However, these results are different from most of the previous global warming studies (Sarmiento et al., 1998; Joos et al., 1999; Matear and Hirst, 1999; Plattner et al., 2001; Matsumoto et al.,

2010), in which ocean circulation was the dominant mechanisms for the reduction in oceanic CO₂ uptake. The important differences

between most previous studies and this study are the resolution of

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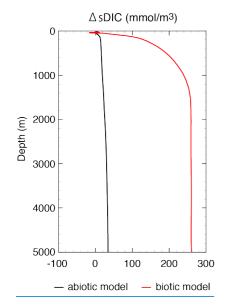


Figure 1. Vertical profile of the salinity-normalized DIC under the pre-industrial condition. The black and red lines show the abiotic and biotic models, respectively.

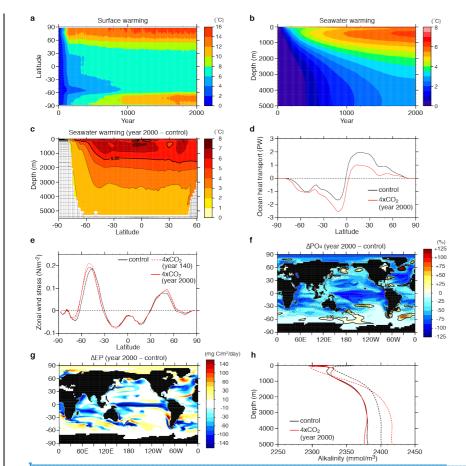
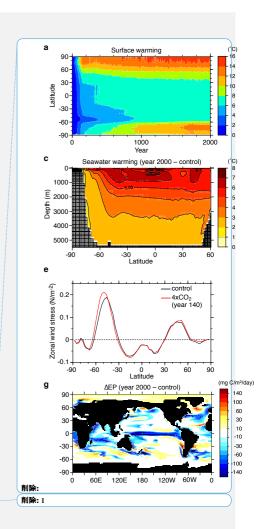
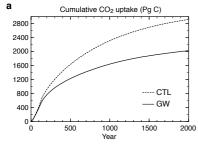
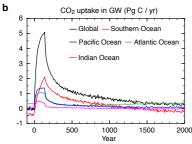


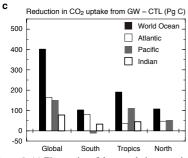
Figure 2. Changes in the physical and biological variables in the global warming experiment. Hovmöller diagrams of (a) the zonally averaged anomaly of the surface temperature and (b) the horizontally averaged anomaly of the seawater temperature. (c) Zonal mean changes in the seawater temperature between the control run and the end of the global warming experiment (year 2000). (d) Northward ocean heat transport for the control (black) and the end of the global warming runs (red). (e) Zonal mean wind stress for the control run (black) and at the time of CO₂ quadrupling (years 140 and 2000; red dashed and solid, respectively). Changes in (f) the surface PO₄ (averaged over the top 50 m) and (g) the export production at the end of the global warming run. (h)Vertical profile of the alkalinity (solid lines) and salinity-normalized alkalinity (dashed lines) for the control (black) and global warming (red) runs.



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Figure 3. (a) Time series of the cumulative oceanic CO_2 uptake for CTL (dashed line) and GW (solid line). (b) Time series of the annual mean values of the oceanic CO_2 uptake for the global ocean (black) and different ocean basins (colors) in GW. (c) Reduction in the oceanic CO_2 uptake due to global warming for the global ocean and three ocean basins at 500 years (GW-CTL). The contributions of the global ocean and individual ocean basins are separated into three regions (South: south of 32.5° S; Tropics: 32.5° S to 32.5° N; North: north of 32.5° N). The positive and negative values represent uptake reductions and uptake increases, respectively.

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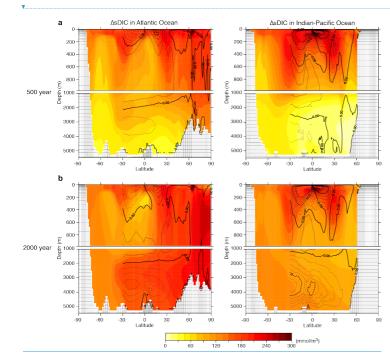
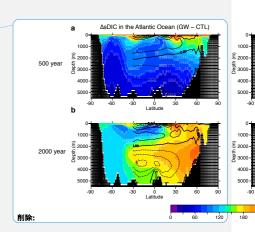
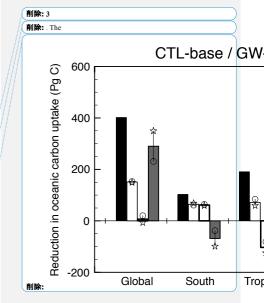


Figure 4. Zonal mean distribution of changes in the salinity-normalized DIC (colors) and meridional overturning stream function (contours) for (a) 500 years and (b) 2000 years in the global warming run (GW). The left and right panels show the Atlantic and Indo-Pacific Oceans, respectively, contour interval is 2 Sv.





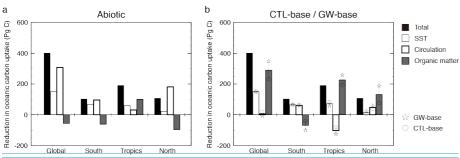
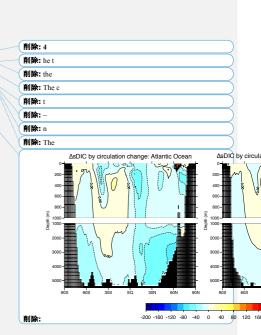


Figure 5. Contributions of the primary mechanisms to the reduction in the oceanic CO₂ uptake due to global warming at 500 years for (a) the abiotic experiments and (b) the CTL-base/GW-base experiments. Total change and effects of SST, circulation, and organic matter are calculated as summarized in Table 2. Contributions of the individual mechanisms are separated into three regions (South: south of 32.5° S; Tropics: 32.5° S to 32.5° N; North: north of 32.5° N). In panel (b), bars refer to the averages of CTL-base and GW-base.



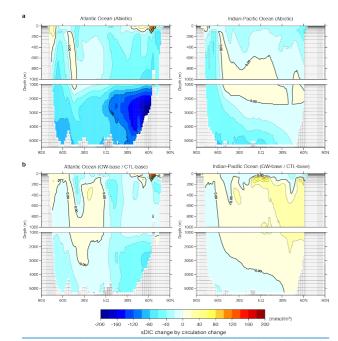


Figure 6. Zonal mean changes in the salinity-normalized DIC induced by circulation changes for (a) the abiotic experiments and (b) the CTL-base/GW-base experiments at 500 years. The left and right panels show the Atlantic and Indo-Pacific Oceans, respectively.

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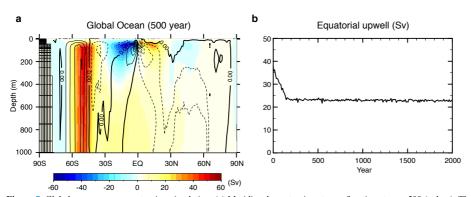


Figure 2. Global upper-ocean overturning circulation. (a) Meridional overturning stream function at year 500 (colors). The positive numbers represent clockwise circulation, and the negative numbers represent counterclockwise circulation. The contours show the differences between meridional overturning stream function in year 500 and the pre-industrial condition.

5 The contour interval is 4 Sv. (b) Time series of global equatorial upwelling (through 200 m, between 10° S and 10° N)

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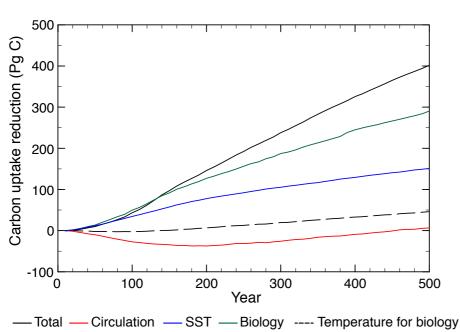


Figure §. Time series of the contributions of primary mechanisms to the reduction in the oceanic CO2 uptake in the CTL-base/GW-base experiments. The positive and negative values represent uptake increases and decreases, respectively. The contributions of circulation (red), biology (green), and SST (blue) and the temperature dependences of production and remineralization (black dashed line) are illustrated. The quantification methods for each mechanism are summarized in Table 2.

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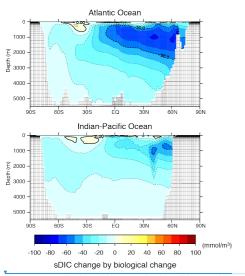
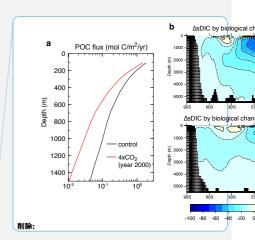


Figure 2. Contribution of the reorganization of the organic matter cycle. Zonal mean changes in the salinity-normalized DIC induced by changes in the organic matter cycle in the CTL-base/GW-base experiments at 500 years. The top and bottom panels show the Atlantic and Indo-Pacific Oceans, respectively.



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削除: (a) Vertical distribution of the POC flux for the control (black) and global warming runs (red). (b)

	Experiments	Changing mechanisms	Cumulative 500year	uptake (Pg C) 2000year	SST
1	CTL	=	1629	2888	CTL
2	GW	all	1227	2028	GW
3	GW_SST	SST	1376		CTL
4	GW_fw	freshwater flux	1230		GW
5	GW_circ	circulation	1220		GW
6	GW_om	organic matter cycle	1563		GW
7	GW_ca	CaCO3cycle	1240		GW
8	GW_si	sea ice	1211		GW
9	GW_biotemp	temperature for biology	1278		GW
10	CTL_SST	SST	1476		GW
11	CTL_fw	freshwater flux	1627		CTL
12	CTL_circ	circulation	1609		CTL
13	CTL_om	organic matter cycle	1413		CTL
14	CTL_ca	CaCO3cycle	1614		CTL
15	CTL_si	sea ice	1643		CTL
16	CTL_biotemp	temperature for biology	1583		CTL

*Pre-industrial seawater temperature is used for calculating biologi

**Seawater temperature of GW_bio is applied for calculating biological contents.

	Experiments	Changing mechanisms	Cumulative 500year	uptake (Pg C) 2000year	SST	Dilution	Circulation	Organic matter cycle	CaCO3 cycle	Sea ice
1	CTL_bio	-	1629	2888	CTL	CTL.	CTL	CTI.	CTL.	CTI.
2	GW_bio	all	1227	2028	GW	GW	GW	GW	GW	GW
3	CTL_abio	-	1819		CTL	CTL	CTL	-	-	CTL
4	GW_abio	SST circulation	1371		GW	GW	GW	-	-	GW
5	GW_abio_SST	SST	1511		CIL	GW	GW			GW
	GW_bio_SST	SST	1376		CIL	GW	GW	GW	GW	GW
7	GW_bio_fw	freshwater flux	1230		GW	CTL	GW	GW	GW	GW
8	GW bio circ	circulation	1220		GW	GW	CLT	GW	GW	GW
9	GW_bio_om	organic matter cycle	1563		GW	GW	GW	CTL	GW	GW
10	GW_bio_ca	CaCO3 cycle	1240		GW	GW	GW	GW	CTL	GW
11	GW_bio_si	sea ice	1211		GW	GW	GW	GW	GW	CTL.
12	GW_biotemp	temperature for biology	1278		GW	GW	GW	GW*	GW*	GW
13	CTL_bio_SST	SST	1476		GW	CTL	CTL	CTL	CTL	CTL
14	CTL_bio_fw	freshwater flux	1627		CTL	GW	CTL	CTL	CTL	CTL
15	CTL_bio_circ	circulation	1609		CTL	CTL	GW	CTL	CTL	CTL
16	CTL_bio_om	organic matter cycle	1413		CTL	CTL	CTL	GW	CTL	CTL
17	CTL_bio_ca	CaCO3 cycle	1614		CTL	CTL	CTL	CTL	GW	CTL
18	CTL_bio_si	sea ice	1643		CTL	CTL	CTL	CTL	CTL	GW
19	CTL_biotemp	temperature for biology	1583		CTL	CTL	CTL	CTL**	CTL**	CTL

Table 1: Description of the model experiments and results for oceanic CO2 uptake.

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•	Mechanisms	GW-base	uptake change (Pg C)
	Total	(2) – (1)	-402
	SST	(2) - (3)	-149
	Freshwater	(2) – (4)	-3
	Circulation	(2) – (5)	7
	Sea ice	(2) - (8)	16
	Biology	(Organic matter cycle) + (CaCO3 cycle)	-349
	Organic matter cycle	(2) – (6)	-336
	CaCO3 cycle	(2) – (7)	-13
	Temperature for biology	(2) - (9)	-51
	Nutrient supply	(Biology) – (Temperature for biology)	-298
۵.	residual		76

^{*}Pre-industrial seawater temperature is used for calculating biological term in GW_biotemp.

**Seawater temperature of GW_bio is applied for calculating biological term in CTL_biotemp.

Mechanisms	Abiotic	uptake change (Pg C)	GW-base	uptake change (Pg C)	CIL- base	uptake change (Pg C)	average of GW-base and CIL-base (Pg C)
Total	(2) - (1)	-402	(2) -(1)	-402	(2) -(1)	-402	-402
SST	(4) - (5)	-140	(2) - (6)	-149	(13) - (1)	-153	-151
Freshwater			(2) - (7)	-3	(14) - (1)	-2	-2.5
Circulation	(5) - (3)	-308	(2) - (8)	7	(15) - (1)	-20	-6.5
Sea ice	-	-	(2) = (11)	16	(18) - (1)	14	15
Biology	(Total) = (SST) = (Biology)	46	(Organic matter cycle) + (CaCO3 cycle)	-349	(Organic matter cycle) + (CaCO3 cycle)	-231	-290
Organic matter cycle			(2) - (9)	-336	(16) - (1)	-216	-276
CaCO3 cycle	_	-	(2) = (10)	-13	(17) - (1)	-15	-14
Temperature for biology	_	-	(2) - (12)	-51	(19) - (1)	-46	-48.5
Nutrient supply			(Biology) – (Temperature for biology)	-298	(Biology) – (Temperature for biology)	-185	-241.5
residual				76		-10	33

Table 2: The contributions of individual mechanisms to the reduction in the CO₂ uptake due to global warming in the first 500 years.

The significant contribution of changes in the organic matter cycle on the uptake reduction due to global warming and the secondary importance of the circulation change are consistent with Zickfield et al. (2008) in which the reduction in the oceanic CO₂ uptake due to freshwater hosing and AMOC weakening was investigated. However, these results are different from most of the previous global warming studies (Sarmiento et al., 1998; Joos et al., 1999; Matear and Hirst, 1999; Plattner et al., 2001; Matsumoto et al., 2010), in which ocean circulation was the dominant mechanisms for the reduction in oceanic CO₂ uptake. The important differences between most previous studies and this study are the resolution of climate model and complexity of biogeochemical model. Our model has an increased atmospheric and oceanic resolutions, so we hypothesise it captures better weaker equatorial upwelling and enhanced mixing due to strengthened subpolar westerly wind in the Southern Hemisphere. These physical changes are not reported in the previous studies. Therefore, the effect of circulation changes in our simulation would be smaller than in the previous studies. These physical changes also affect the nutrient supply and production. Moreover, the temperature dependence of production and remineralization was not considered in most of previous studies using simple ocean biogeochemical models (such as nutrient-restoring model). These mechanisms would lead to larger biological effects relative to previous studies.

The methods used to quantify the effect of ocean circulation on the changes in oceanic carbon uptake are also different between most previous studies and this study. In the former typical method (Sarmiento et al., 1998; Plattner et al., 2001; Matsumoto et al., 2010), abiotic experiments (i.e., without a biological pump) were conducted to isolate the physical process from the biological process. In the abiotic experiments, the vertical gradient of DIC under the pre-industrial condition is much smaller than realistic vertical gradient of DIC because biological process primarily maintain most of the surface-to-deep gradient of DIC as shown in Sarmiento and Gruber (2006). The circulation effect estimated by the abiotic experiments would

underestimate the enhanced CO₂ uptake associated with the reduced upward transport of natural CO₂ from the deep ocean to the upper ocean. The reduced CO₂ uptake due to the reduced downward transport of anthropogenic CO₂ from the surface to deep ocean are primarily represented. This study based on Zickfeld et al. (2008) separated these two effects more directly. In our methods (CTL-base/GW-base experiments) with a realistic vertical gradient of DIC, the effect of the circulation change represents both a reduced upward transport of natural CO₂ and a reduced downward transport of anthropogenic CO₂. Therefore, the contribution of circulation changes to the reduction in CO₂ uptake in the previous studies would be larger than that in this study.