

Dear Prof. Heinze

Thank you for handling our manuscript "*Burial-nutrient feedbacks amplify the sensitivity of carbon dioxide to changes in organic matter remineralisation*". In the following, you find a summary of the relevant changes made to the manuscript:

- change of title:

"Burial-nutrient feedbacks amplify the sensitivity of **atmospheric** carbon dioxide to changes in organic matter remineralisation "

(added word "atmospheric")

- extended (sediment)-model description

- extended discussion of model limitations and caveats

- the EOF analysis as been removed (including the corresponding figure 6)

For minor changes see point-to-point response to the reviewers comments on the next pages.

We hope that with these changes, our manuscript meets the quality standards for publication in ESD.

Sincerely
Raphael Roth

Original comments are given in black, our reply is given in red.

Anonymous Referee #1

Received and published: 9 May 2014

The manuscript by Roth et al. describes the impact on atmospheric CO₂, ¹³C and nutrients distribution of a deepening of the Particulate Organic Matter (POM) remineralization depth. 100 kyr long sensitivity experiments are performed with an Earth System Model of Intermediate complexity, which also includes a sediment model. It has been previously suggested that a deepening of POM remineralization during glacial times could have played a role in decreasing atmospheric CO₂ (Matsumoto 2007, Matsumoto et al. 2007, Kwon et al. 2009, Menviel et al. 2012 and Chikamoto et al. 2012). This study is thus relevant to understand glacial/interglacial changes in the carbon cycle and I recommend its publication in ESD with minor revisions.

We thank the reviewer for his valuable comments that helped to improve the manuscripts and for his time and effort to review this manuscript.

1) Introduction:

In general I find the Introduction a little messy with some inaccuracy and redundancies. Some specific examples include:

a) the latest references have been chosen to illustrate the hypotheses brought forward to discuss glacial changes in pCO₂ whereas I think that it might make more sense to cite the original reference. For example, even if Volker and Kolher 2013 paper is very interesting I think it makes more sense to cite Toggweiler et al. 2006.

Done. Siegenthaler and Wenk (1984) is cited as an early paper on the role of ocean circulation for atm. CO₂. In addition we refer the reader now explicitly to available reviews by modifying the text to read: (see reviews by Sigman and Boyle, 2000; Archer et al., 2000; Sigman et al., 2010; Fischer et al., 2010). It is beyond the scope of this MS to provide a comprehensive review and to cite all important papers on this subject of the last years.

b) Please do not only quote the abstract of Kwon et al. 2009, but instead it might more relevant to say that a 24m increase in the e-folding depth of the POM remineralization leads to a 10ppmv decrease. The 27 ppmv is obtained when export production is kept constant.

Done. Text modified to read: "Kwon et al. (2009) simulate a CO₂ drawdown by 10 and 27 ppm for an increase in the e-folding depth of POM remineralisation of 24 m in their nutrient-restoring and constant export-production model setups."

c) the impact of changes in viscosity on the speed of temperature (Taucher et al. 2014) is discussed both on lines 14 and 28 of p475.

Done. Text clarified. The mechanism is generally discussed on lines 10 to 21 in the original MS, while quantitative results obtained for the different studies are given on

line 21 ff, An new paragraph is started on line 21 and text on line 21ff of the original MS is modified to read: "The mechanism of a deepening of the POM remineralisation has been discussed and quantified in several studies..."

d) A little more information on Menviel et al. 2012 study in the introduction would be relevant. For example that a progressive increase of the POM and DOM remineralization depth over the glaciation led to a 31 ppmv pCO₂ decrease, while the increase back to initial condition during the deglaciation led to a 21 ppmv increase, thus suggesting that ocean-sediment interactions could play a role in amplifying the pCO₂ decrease and thus motivated the present study.

Done. Text modified to read: "Menviel et al. (2012) performed transient model simulations over the last glacial cycle with the Bern3D model applying a temperature-dependent remineralisation rate. They simulated a 31 ppm decrease in atmospheric CO₂ for a progressive increase of the POM and DOM remineralization depth over the glaciation, while CO₂ increased by 21 ppm due to altered remineralization over the deglaciation. This suggests that ocean-sediment interactions may contribute to the reconstructed atmospheric CO₂ variations, motivating the present study."

e) At the end of the introduction L14-18, I am not sure I agree with "previous estimates...underestimate the long term feedback". I would suggest to rephrase that sentence as either long previous studies could not study that feedback due to their model/experiment set up or they did point out that on timescales greater than 10kyr ocean sediment interactions could amplify the signal.

Done. Text modified to read: "We show that previous estimates of the sensitivity of POM remineralisation rate changes are not applicable for glacial-interglacial time scales as they do not include the long-term feedback, but point to a potential importance of this mechanism to explain low-frequency CO₂ and ¹³CO₂ variations."

2) Experiments:

This section needs some revision. The "closed system" set up might need some more explanation: usually in a closed system the riverine input balances the burial of organic matter and/or CaCO₃. Apparently here another method is used, indicating that the sediment model has been disabled, therefore there is no burial. Also I don't see how that is similar to "Kwon et al. 2009" as they use in their experiments "surface nutrient restoring" or "constant export".

Done. Sentence referring to Kwon et al. deleted and text defining "closed system" modified to read: "We also run the system in an atmosphere-ocean only setup without sediment and no river input and burial, referred-to as 'closed system'."

L 2, p483. Please rephrase as "Our discussion mainly focuses on ..."

Done. Text modified as suggested.

L.5-6, p483, please rephrase.

Done. Sentence reads now: "We restrict our analysis to idealized changes in in the remineralisation length scale (parameter α and l_{POM}) as the relationship between remineralisation and temperature changes is not well understood."

3) l_{calc} :

Experiments in which l_{calc} is changed are not described at all in the experiments section.

This is not true. The first sentence in section 2.4 reads: "Sensitivity experiments are performed where l_{POM} (or equivalently α) and/or l_{calc} are changed in a step-wise manner." In response to the comment we added a reference to equation 6 to point explicitly to the link between l_{calc} and the flux of calcium carbonate particles. We also modified the text before eq. 6 to read: "The downward flux of calcite (including other forms of calcium carbonate such as aragonite or high-magnesium calcite particles) F_{calc} decreases exponentially with depth with a length scale l_{calc} "

It is as cryptic in most of section 3.2. as it is not mentioned in the text by how much l_{calc} is changed or any specificity. One has to wait until the end of paragraph 3.2. to get an example on what has been changed. Figure 10, where the l_{calc} results are shown is only discussed in the following paragraph.

Done. The first paragraphs of section 3.2 were modified and the text reads now:

"It has been suggested that changes in the rate of $CaCO_3$ dissolution in the upper ocean will be a significant feedback affecting atmospheric CO_2 concentrations and future climatic changes (e.g. Barrett et al., 2014). To this end, we prescribe in a further set of sensitivity simulations a step change in the e-folding dissolution length scale l_{calc} governing the dissolution profile of $CaCO_3$ particles within the water column (see eq. 6). l_{calc} is changed at the end of the spin up from its standard value of 2900 m to values ranging from 2100 to 3700 m; then the run is continued for another 50,000 years with the new value of l_{calc} . Export fluxes both of POC and calcite remain constant as changes in the dissolution of calcite do not affect productivity in our model.

The experimental setup with the assumption of an e-folding remineralisation profile for calcium carbonate particles (Eq. 6) is highly idealised. The mechanisms for the dissolution of calcite and other forms of $CaCO_3$ (e.g. aragonite or high-magnesium calcite) within the water column are quantitatively not well understood. Generally, dissolution of $CaCO_3$ particles within the water column is thought to be linked to low (undersaturated) concentration of carbonate ions in the surrounding water. However, considerable $CaCO_3$ dissolution may occur in the upper ocean (Berelson, 2007) in waters that are saturated with respect to $CaCO_3$ in the mineral form of calcite or even of aragonite, perhaps due ..

... The results for these additional sensitivity experiments are as follows. ... "

4) EOF:

EOF are a useful statistical method to highlight modes of variations and is widely used in oceanography, meteorology... Here the authors spend some time explaining

the method in section 2 and mention their EOF a little bit everywhere (abstract, introduction, section 3.1). While I have no doubt that EOF can be very useful I am a little bit more skeptical about their use here. Moreover because the 2nd EOF explains only 0.27% of the variance. Technically one would say that the 2nd EOF is not significant. A deepening of the POM remineralization leads to a DIC increase at depth, while the alkalinity might be little affected at first. This thus induces a decrease in deep [CO₃] and eventually dissolution of CaCO₃. Deep [CO₃] thus increases again. So basically it seems that the 2nd EOF is showing this initial deep [CO₃] decrease. If the only thing you want to show by using the EOF is that deep [CO₃] first decreases before increasing, there might be an easier and cleaner way to do that than using the EOF. I would thus suggest taking out all that is related to the EOF in the paper. For example I think that the “closed system” experiments provide all you need to make the point. I would first suggest to add the evolution of POM export, calcite export... for the closed system in Figure 3. Then you could add in the text that the experiments in the “open system” follow the ones of the “closed system” for a few thousand years after which ocean-sediment interactions start to play a significant role. Another way would be to make Hovmöller diagram as the one shown in Fig8b.

We agree with the reviewer's suggestion and removed the EOF-related text and figure from the MS.

5) C13:

L2, p 491, I would avoid “isotopically enriched calcite”.

Done. Sentence modified to read: “Similarly, burial of calcite, a flux which is isotopically enriched compared to the POM and the total burial flux (POM and calcite), is reduced ...”

And I doubt this has a significant effect on deep d¹³C.

The statement by the reviewer is not correct.

The isotopic signature of calcite particles is with 3 ‰ very similar to that of DIC (~0 ‰). Changes in the calcite cycle within the ocean (closed system) do hardly modify the ¹³C signature of DIC and atmospheric CO₂. On the other hand, the calcite burial flux is isotopically enriched by about 15 ‰ relative to the total carbon burial/weathering flux. The δ¹³C difference between calcite burial flux and weathering input flux is (on absolute terms) with 15 ‰ even twice as large than the difference of 7 ‰ between the POM burial flux and the weathering input. Thus, changes in calcite burial do affect ¹³C signatures.

The text has been modified to clarify this point:

“(i) The excess burial of isotopically light POC (δ¹³C ~ -20‰) during the first 50 kyr tends to increase the average δ¹³C signature in the ocean-atmosphere system; the δ¹³C signature of the POC burial flux is with -20‰ about 7‰ lower than the signature of the total carbon weathering/burial flux (-12.6‰). (ii) Similarly, burial of isotopically enriched calcite (δ¹³C ~ 3‰) is reduced (relative to initial conditions and the weathering flux) during the first 100 kyr; this tends to increase d¹³C during this period; the calcite burial flux is enriched by about 15‰ compared to the average signature of the weathering/burial flux.

Additionally the changes in $\delta^{13}\text{C}$ simulated are fairly small.

Simulated changes are order 0.1‰ in the ocean and atmosphere for an increase in the POM remineralisation length scale from 250 to 275 m. This may be compared with LGM-Holocene variations in whole ocean and atmospheric $\delta^{13}\text{C}$ of order 0.3 ‰ and an LGM-late Holocene difference of about 0.1‰ (Schmitt et al, 2012.)

I am quite surprised by the fact that the simulations do not reach an equilibrium after 200ky.

This fact is indeed interesting and mainly results from the CO_2 -dependent fractionation during photosynthesis. The reasons for the long timescales are discussed in the MS starting from L26, p491.

6) Conclusions: L 21, p497: Deep Atlantic Cd/Ca content was increased during the LGM indicating a greater deep $[\text{PO}_4]$ content.

Done. We agree- thank you. Text clarified to read: “As deep glacial Cd/Ca ratio in the North Pacific, ..”

L25p 497: “ a 25m increase in the e folding...”

Done. Typo corrected.

There are some typos throughout the text. Some examples: L3, p479 : “calcifier” iof “calcifer”.

L 28, p 481 “so” is missing.

L16, p483 “induced”.

L 16, p 488 “ As a result”.

L8, p 495, “the experiments”.

L 14, p 495 “fixed”.

Thank you. Typos corrected.

References

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Sigman, D. M., Hain, M. P., and Haug, G. H.: The polar ocean and glacial cycles in atmospheric CO₂ concentration, *Nature*, 466, 47–55, doi:10.1038/nature09149, 2010.

Original comments are given in black, our reply is given in red.

D. Archer (Referee)

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This is a state-of-the-art calculation, the latest in a series of studies that are well described in the introduction section. The authors clearly describe the distinction between closed system behavior and open, which responds on a much longer time scale but with generally larger amplitudes.

We thank the reviewer for his valuable comments that helped to improve the manuscripts and for his time and effort to review this manuscript.

I guess the part of the simulation that is the weakest, a reflection of the state of the science rather than any deficiency in the paper, is in the calculation of organic carbon burial as a function of oxygen concentration and organic carbon deposition rate. Organic carbon burial depends also on the grain size of the sediment, and on the mineral deposition rate. River deltas can capture significant fractions of global carbon deposition. This uncertainty primarily affects the longer-term “open system” response, which is therefore much more uncertain than the shorter-term responses. The short term responses seem quite believable, and that we might actually be able to predict them. The longer term responses from model are probably better viewed as potential or hypothetical.

To make to reader aware of these shortcomings in our model, we extended the description of the model (see also answer to reviewer 4 (Wallmann)):

"Input of terrestrial organic matter into the ocean and burial of terrestrial organic matter is not explicitly considered (see e.g. Regnier et. al, 2013). Similarly, the cycling of P associated with iron and other oxides is neglected as estimates suggest that 97% of the P delivered to the sediment-water interface is in the form of organic matter (Delaney, 1998). The specific chemical composition of the organic matter, particle grain size of the sedimentary material and available area for absorption for organic matter (Hedges and Keil, 1995) as well as spatio-temporal variations in mineral deposition rates or sediment porosity, which likely influence organic matter preservation and burial (Burdige, 2007), are neglected."

In addition, we state now in the discussion section:

"There are also limitations regarding the sediment model. For example, the spatio-temporal variability in the deposition of mineral particles or the influence of particle grain size on organic matter preservation are neglected. The coarse resolution hampers the representation of coastal and continental boundaries, where most POM deposition, remineralization and burial occurs (e.g. Wallmann et al., 2012). The model does not resolve river deltas and estuaries and their carbon cycle (see e.g. Regnier et al, 2013). Another caveat is that denitrification within the sediment is not represented by our model, eventually leading to a bias in the

long-term response of POM degradation and thus burial-efficiencies. Therefore, our findings are to be confirmed and refined by a higher-resolved ocean models with a more complete representation of sediment processes."

Another frontier of science which this paper brings us to is the factors that determine the remineralization depth scale, and in particular the impact of CaCO₃ sinking, as ballast, on the organic carbon sinking depth scale.

We included the following sentence in the discussion section (p495, l12 of the original MS):" It is a task for future research to advance the mechanistic understanding of the processes governing particle fluxes and remineralisation rates."

I didn't follow the discussion of the prescribed-production model run in section 3.1.2. The rationale for doing it is understandable enough, as an attempt to deconvolve the impacts of changes in surface production vs. remineralization depth scale. The distinction is rather artificial, as demonstrated that the model blows up if you try to impose it for too long. But what I don't understand is the conclusion that there is no equilibrium CO₂ change for the open system. There is no equilibrium for the prescribed-production model, but that's not the normal open system model.

Text clarified. We replaced 'open system' with 'prescribed-production model' (p490, l1 of original MS) to read: "Therefore, no equilibrium-changes in CO₂ can be stated for the prescribed-production model .."

References:

Burdige, D. A., 2007. Preservation of organic matter in marine sediments: Controls, mechanisms, and an imbalance in sediment organic carbon budgets? *Chem. Rev.* 107, 467-485

Delaney, M. L.: Phosphorus accumulation in marine sediments and the oceanic phosphorus cycle, *Global Biogeochem. Cy.*, 12, 563–572, doi:10.1029/98gb02263, 1998

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Regnier, P., Friedlingstein, P., Ciais, P., Mackenzie, F. T., Gruber, N., Janssens, I., Laruelle, G. G., Lauerwald, R., Luyssaert, S., Andersson, A. J., Arndt, S., Arnosti, C., Borges, A. V., Dale, A. W., Gallego-Sala, A., Godd'ris, Y., Goossens, N., Hartmann, J., Heinze, C., Ilyina, T., Joos, F., LaRowe, D. E., Leifeld, J., Meysman, F. J. R., Munhoven, G., Raymond, P. A., Spahni, R., Suntharalingam, P., and Thullner, M.: Global Carbon budget and its anthropogenic perturbation in the land-ocean aquatic continuum, *Nature Geoscience*, 6, 597–607, doi:10.1038/ngeo1830, doi:10.1038/ngeo1830, 2013

Wallmann, K., Pinero, E., Burwicz, E., Haeckel, M., Hensen, C., Dale, A., and Ruepke, L., 2012. The global inventory of methane hydrate in marine sediments: A theoretical approach. *Energies* 5, 2449-2498.

Original comments are given in black, our reply is given in red.

J. Segschneider (Referee)

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General comments:

The paper describes experiments with the Bern3D model in which the remineralization depth is shifted upwards and downwards by 25 m intervals. This is meant to represent temperature dependent remineralization in a warming/cooling ocean on glacial/interglacial time scales. The model is integrated for 100 to 200 kyr either in 'open system' mode (with an active sediment) or in 'closed system' mode (without an active sediment). Atmospheric pCO₂ and δ¹³C are analyzed from these experiment and the main finding of the study is that pCO₂ changes are, on longer time scales than 1000 yr, significantly higher when nutrient deposition in the sediment is taken into account. This assumes that weathering fluxes are constant in time. The authors provide also a value of change in atmospheric pCO₂ per metre shift of the remineralization profile.

As such it provides a valuable contribution to the impact of changes in remineralization of organic matter on the carbon cycle, in particular regarding glacial/interglacial time scales. The paper is generally well written and figures and tables are well thought out. At some points it could be a bit more precise, and the guidance of the reader could be improved. Below are more specific comments that hopefully will improve the paper.

We thank the reviewer for his valuable comments that helped to improve the manuscripts and for his time and effort to review this manuscript.

Specific comments

General: POC/POM/POP are used, I suggest to stick to one name (POM seems most frequent)

Done. We checked the use of POC, POM and POP. We use now the term POM when referring to organic matter in a general sense, but still use POP and POC when fluxes of phosphate or carbon are discussed.

you do not discuss changes in surface alkalinity due to reduced productivity in the open system experiments. Is this because you found that it is irrelevant?

It is not the aim of this paper to discuss surface ocean biogeochemistry changes due to changes in marine productivity. Especially in the open system, the separation of production, remineralization and sedimentation effects are difficult to separate.

In our model, CaCO₃ export decreases in parallel to POM export except in the Southern Ocean where the rain ratio decreases (as silicic acid is not as depleted as phosphate, favouring diatoms w.r.t. calcifier). But this effect is confined to the SO. Thus, reduced productivity increases surface Alk (dominated by the weakened carbonate counterpump) and increases DIC (dominated by the weakened soft-tissue pump). This is in agreement with Segschneider et al. (2013) who find a decrease in TA for an increased rate of remineralisation.

To summarize this, we included the following sentence in the results section (p485, l14):

"The reduced export triggers some minor changes in the ecosystem structure in the Southern Ocean, where phosphate depletion favours the growth of diatoms, leading to a local decrease in the export rain ratio. The net effect of the production changes is thus a surface-ocean increase in DIC and Alk, except for the Southern Ocean where surface Alk remains constant."

Title: I suggest to state that atmospheric carbon dioxide is meant

Done. Word atmospheric added

Abstract p474 l3: I suggest to change 'provide a positive feedback under climate change' to sth like 'provide a positive feedback mechanism with atmospheric CO₂ and hence climate change'

Done. Text changed

l5 CO₂ -> atmospheric CO₂

Done.

l5/6 it is stated that the response of tracer fields for which observations and paleo proxies exist is analyzed, but there is never a comparison with proxies in the ms. nor are the proxies simulated that are mentioned in the 'Discussion and conclusion' section. so I suggest to either be more specific and come back to this in the text or remove the statement. (also p477 ln 5)

We removed the term paleo proxy from p477 ln 5.

A direct comparison with proxy data is not meaningful as the experiments here are of illustrative nature. However, we do discuss results for tracers for which direct observations and paleo proxy data exist. These include ¹³C, CaCO₃ and POM burial, oxygen and carbonate ions. Thus, we left the text in the abstract unchanged as we view the statement as correct

l8 it is not the 'initial' response, but the 'long term' response in atm. CO₂ which is amplified by the sediment burial-nutrient feedback (see Fig. 7a) (also p477 l18)

Done. Word 'initial' deleted

l8/9 why is this called a 'temporary' imbalance? (It is called 'transient' on p476 l17, which I find more appropriate, and 'sustained imbalance in Sec 3.1.2, p489 l6)

Done. 'temporary' replaced by 'transient' and 'sustained' by 'long-lasting

l13/14 I suggest to insert 'atmospheric' before δ¹³C signatures and CO₂ sensitivity

Done. 'Atmospheric' added for CO₂ sensitivity, but not for the δ¹³C signature as not just the atmospheric signature is changing.

Introduction

l26 'calls for' too strong? what about pCO₂ as driver of temperature changes?
suggest to replace 'tight coupling' by 'larger temperature changes than one would expect from delta pCO₂ based on climate sensitivity of current climate models' if that is meant

Done. Sentence deleted to avoid confusion.

l27 I suggest to change CO₂ drawdown 'during' the Last Glacial Maximum to 'leading to' the last LGM

Done. Text modified as suggested.

p475 l14 to 'increase viscosity and thus the speed of sinking particles'?? sign correct
I would expect a smaller sinking velocity for increased viscosity. It should be ...'to decrease viscosity and thus to increase the speed of sinking particles'
Taucher et al. 2014: 'As rising temperatures reduce seawater viscosity, the sinking velocity of particles will accelerate'

Thank you – mistake corrected

l17/18 I suggest to delete the 'increase' before 'respired carbon storage'.... 'such changes' refers also to Bendtsen et al, Taucher et al., and there the changes will result in decreased respired carbon storage in the deep ocean

Done. 'increase' deleted.

l19 Also the 'As a result' assumes that changes in remin-depth lead to 'increased' storage in the deep. Deleting 'As a result' is an easy way out here, then the two sentences are correct.

Done. Text modified as suggested.

p476 l3 'all these studies neglect ocean-sediment interactions' this is not strictly true, e.g., the model of Segschneider & Bendtsen includes a sediment (see their Fig. 6) - for the time-scales of 100 yr they discuss, the sediment is of minor importance, as also seen in Fig. 7a of this ms. Also Tschumi et al. 2011 have an active sediment included and discuss briefly the amplification of the closed system by taking into account ocean-sediment interactions (their Section 2.3.3). I suggest to be a bit more specific about how this study differs from the one of Tschumi et al. 2011 (e.g., different focus, constant physics...)

Done. Sentence changed to read: 'As a caveat, these studies either focus on the decadal-to-century scale response (Segschneider, 2013) or neglect ocean--sediment interactions and the weathering-burial cycle.'

Sentence added to clarify difference to study by Tschumi et al; 'However, these authors did not investigate changes in the remineralization depth of POM. '

l19 Change 'These consequences' to 'The consequences of this imbalance' have not been discussed...?

Done. Text modified as suggested.

p480 l12 an e-folding depth (l_POM): e-folding depth 'as length scale' (l_POM) would make it more easy to understand why it is called 'l_POM') and could avoid confusion throughout the text (where 'depth', 'mean remineralisation depth', 'profile', 'POM deepening' and 'length scale' are used for 'l_POM')

Done. Text modified as suggested.

l21 Is there really 'advection' in the sediment?

Done. 'advection' replaced by 'transport' to avoid confusion.

p483 l20 I suggest to change 'during the experiments' to 'during our experiments' to make clear that not the experiments in the referenced studies are meant

Done. Text modified as suggested.

Results

p485 l2 3.1 l_POM changes: I suggest a slightly more informative heading

Done. Heading changed to read: 'Changes in the remineralisation length scale of POM'

l4 I would not start the Results section by 'We start discussion by'....

Done. Text modified to read: 'We first analyze ..'

l11 'subsurface water' - you could be a bit more precise here. Is the water directly below the euphotic layer or within the euphotic layer meant?

Done. 'subsurface water' replaced with 'upper thermocline'.

l12 how robust is the decrease in global export with respect to the neglect of remineralisation within the euphotic layer? If you would consider remineralisation in the euphotic layer, a downward shift of the remineralization profile would cause increased export.

Done. On average, export production is balanced by the input of nutrients into the euphotic zone. We thus assume that potential changes in the ratio between recycled and new production do not affect export out of the euphotic zone. We added the following caveat: "Note that we only consider changes in remineralisation below the euphotic zone."

l14 'leads to an initial spike in POM deposition' does not give the sign, so I suggest to change it to 'spike-like increase' and to add a ref to Fig.3d

Done. Text modified as suggested and reference to figure added.

l22ff would it be useful to add at the end of the sentence 'Second (the) whole ocean...'

...after the step change 'due to reduced productivity in the euphotic zone'?

Done. Text clarified by adding: 'due to excess burial of POP.'

p486 l6 suggest to replace 'Finally' by 'Eventually' (I guess this is meant)

Done. Text modified as suggested.

l22 I suggest to insert 'decreased' between 'by' and 'calcite burial' to make the sentence easier to follow or perhaps reverse it: The loss of carbon due to enhanced burial of POM is counteracted by a small gain due to reduced calcite burial, while....

Done. Text modified by adding decreased.

p487 l7 suggest to add (Fig. 4a) after ocean

Done. Text modified as suggested.

l17 suggest to add (Fig. 4c) after deposition

Done. Text modified as suggested.

p488 l1 suggest to add (Fig. 5a) after PO₄ inventory

Done. Reference to figure added at end of sentence.

l6 'As expected, this pattern is mirrored ...by DIC, oxygen, d13C
In the closed system, the patterns of oxygen and CO₃ are inverse to the pattern of PO₄ (see Fig. 5a, 5c, 5g) whereas for DIC it is similar (Fig.5e) so I doubt that 'mirrored' is meant. I also assume that CO₃ is meant, not d13C, which is not shown in Fig. 5? Also I suggest to point to the figure panels and to add a 'not shown' for variables not in Fig. 5 (d13C, ALK, CaCO₃) in the text. (Alk is discussed relatively heavy in this section so you might want to show it in Fig.5)

Done. Figure references added and text revised to read: "As expected, this pattern is similar in subsurface waters for DIC (Fig. 5e) and inverse for d13C (Fig. 8d) and oxygen (Fig. 5c) as these tracers are linked to PO₄ by constant Redfield elemental ratios in biological fluxes."

The pattern for Alk change is quite similar to the pattern of the carbonate ion change. An additional panel for ALK is added to Figure 5.

l25 I suggest to start a new subsection here and discuss the EOFs in more detail, or to skip this para and to skip Fig. 6 Why are the EOFs computed for (only) CO₃?

We decided to remove the material linked to the EOFs analysis, see also the comment to referee #1.

l27 I would change 'by 2' to 'the first two'

Done. Text modified as suggested.

l28 and insert 'of the principal components' after 'corresponding time series'

Done. Text modified as suggested.

l28/29 'The resulting patterns strongly resemble those shown with (in) Fig.5g and h, i.e. the open(-) and closed(-)system response'..... I recommend to be more precise: 1st EOF resembles open system (Fig. 5h), 2nd EOF resembles closed system (Fig. 5g)

Done. Text modified as suggested.

p489 l14 'The CO₂ decrease is more than 100ppm for a change in I_POM to 375m' Can you say if the implied temperature change is in agreement with LGM temperature?

It is beyond the scope of this MS to link CO₂ changes to temperature changes. Ice core data show that atmospheric CO₂ was about 100 ppm lower at the LGM than in the late Holocene. The associated radiative forcing contributed to the cold conditions at the LGM, but other forcings such as a higher albedo due to larger ice sheets and enhanced snow cover, changes in vegetation cover and dust loading contributed. For further information see Jansen et al (2007)

l21 check use of 'standard experiment' here. It has been referred to as I_POM=250m, here it seems to be used for I_POM=275m (also line 25)

Done. Text clarified to read: "Global export production decreases when increasing the remineralisation length scale in the standard model setup."

l22 'to disentangle the influence of the reduced export flux vs. the change in remineralisation profile' is this possible using this setup? Also, by disentangling, I would expect some more elaborate results, e.g., change in remin causes this and export causes that, not just the factor of 2-4 for the (unrealistic) constant export exp.

Done. "disentangle" replaced by "explore".

p490 l3 It might be useful to explain why these experiments have been made. Is there a reason to assume that changes in remin-depth have occurred in confined regions during glacial periods?

Done. Sentence added: "The hypothesized change in the remineralisation depth may be different in different regions."

p491 l10 what is meant by 'checked by' here? balanced?

Done. 'checked by' replaced by 'balanced by'.

Discussion and conclusion

p494 l22 It is shown that 'on long time scales' ocean sediment interaction...

Done. Text modified as suggested.

p497 l5 Gangstoe et al. 2011 ...did not apply a sediment model. Even though Gangstoe et al do not mention a sediment module, PISCES usually runs with active sediment, so it may be worthwhile to check if this statement is correct.

Done. Statement is correct.

Fig. 8a any idea why the red lines (d13C open system) in Fig.8a cross all at 50 kyr and at the level of the control experiment?

As all the perturbation experiments underly the same timescales of re-adjustment as they lay within the quasi-linear regime. Due to reasons of symmetry, this crossing must take place at the level of the control experiment.

Fig. 10 does not show a 'map'.
also it would be helpful to state the times for which the plots are produced

Done. Word 'Map' deleted and the following text is added in the caption: ' and for equilibrium (see main text).' It is explained in Section 3.3 how equilibrium values are estimated.

is it easy to understand why the open system amplification is highest around $I_{POM} = 250m$? I guess this is due to the division by small numbers (closed system response is close to zero for $I_{POM}=250m$), so this may be a bit misleading as it implies that the open system amplification is particularly strong for small perturbations of I_{POM} .

It is not straightforward to understand why the amplification is larger for small perturbations in I_{POM} as changes in ratios are difficult to interpret in such a non-linear and complicated system. However, we note that the amplification shows a decreasing trend when increasing I_{POM} over the range from 250 to 360 m (color gradient in figure 10c). Thus, we consider the results shown in Fig. 10c as numerically robust .

Technical errors

p474 l 10 ...lead to sustained changes 'in' the ('in' missing)

p475 l 21 This mechanisms (sgl/pl)

p478 l3 correct 'arithmically'

p479 l3 correct calcifer

p480 l17 change 'lengthscale' to length scale

p483 l16 change 'as induces' to 'as induced'

p481 l23 change 'prescribed to' to 'set to' or 'prescribed as'

p482 l2 change 'Global integrated' to 'Globally integrated'

p484 l7 change 'describe' to 'describes'

p487 l2 change to ...after 32kyr (Fig.3g) (not 3f)

l18 change 'region' to 'regions'

l20 change Indian ocean to Indian Ocean

p488 l16 change 'As a results' to 'As a result'

l21/22 change This ... 'anomalies are' to 'anomaly is' (or 'These...')

p490 l3 change 'pumps' to 'pump'

p492 l16 dot is missing after 'in sensitivity simulations'

Done. (logarithmically is correct)

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Segschneider, J. and Bendtsen, J.: Temperature-dependent remineralization in a warming ocean increases surface pCO₂ through changes in marine ecosystem composition, *Global Biogeochem. Cy.*,27, GB004684, doi:10.1002/2013GB004684, 2013

Original comments are given in black, our reply is given in red.

K. Wallmann (Referee)

Received and published: 19 May 2014

The paper by Roth et al. is well written, clearly structured, and a pleasure to read. It illustrates the importance of benthic processes for the long-term evolution of ocean chemistry and atmospheric pCO₂. More specifically, it shows that a shift of POM degradation to larger water depths has a strong impact on seawater composition and atmospheric pCO₂ when sediments are included in the model set-up. The paper is very nice and innovative.

We thank the reviewer for his valuable comments that helped to improve the manuscripts and for his time and effort to review this manuscript.

However, I find it very difficult to evaluate the model results since the benthic model is not fully explained. Even though I read the accompanying paper by (TSCHUMI et al., 2011) which provides more detail on the benthic model, I have a number of questions that should be addressed in the preparation of the final version:

1. The model excludes the burial of neritic carbonates at continental shelves and uses a low estimate for pelagic carbonate burial (only 0.096 Gt C/yr, Tab. 1). On the other hand, the global POC burial rate is quite high and apparently includes POC burial at continental margins (0.181 Gt C/yr, Tab. 1). The authors should explain how their model distributes POC burial between the deep-sea (>1000 m water depth) and the continental margins (<1000 m water depth). Sedimentary data show that about 80 – 90 % of global POC burial occurs at continental margins while the deep-sea contributes only about 0.01 – 0.05 Gt C/yr to the total POC burial rate (BERNER, 1982; BURDIGE, 2007; BURWICZ et al., 2011; HEDGES and KEIL, 1995; MIDDELBURG et al., 1993; WALLMANN et al., 2012). Does the model reproduce and consider these important observations?

Our coupled model is tuned to match observational estimates of a total POC burial flux of 0.19±0.07 GtC/yr (Sarmiento & Gruber, 2006, page 261, table 6.5.1). The POC burial flux is distributed in our model as 70% (<1000 m water depth) and 30% (>1000m water depth). Although these numbers do not fall within the stated range (80-90% on continental margins), we think that our model does a reasonable job, given the fact our coarse-resolution ocean model uses a strongly lowpass-filtered bathymetry by design.

The following lines were added in the section where the preindustrial model state is discussed (p481):

"The partitioning of POM burial between the deep ocean (>1000 m water depth) and the continental margin (<1000 m water depth) is 70% and 30%, respectively, while observations indicate that 80-90% of the POM burial is on continental margins. This model bias is likely linked to the coarse horizontal resolution and the simple

continental runoff-scheme, compromising the representation of near-coast processes."

2. In the real ocean, the benthic turnover of phosphorus and organic carbon are partly decoupled and do not follow Redfield stoichiometry. The mean molar ratio between POC and total P in deep-sea sediments is not 106 but rather 20-30 since phosphate released from organic matter forms authigenic minerals in sediments and adsorbs to iron oxides and other sediment surfaces (BATURIN, 2007; WALLMANN, 2010). Moreover, a decrease in bottom water oxygen tends to enhance POC burial (BURDIGE, 2007) while less P is buried under low oxygen conditions (VAN CAPPELLEN and INGALL, 1994). I do not fully understand how these opposing trends are considered in the benthic model. Does the benthic model assume Redfield stoichiometry? How does bottom water oxygen affect the burial efficiency of POC, P, and POM?

A description of the sediment diagnosis model and the governing equation is given in Tschumi et al., 2011. The spatial model domain is restricted to the diagenetical zone of the sediments which is assumed here to be the top 10 cm of the surface sediments. Any solid material leaving this domain disappears into the subjacent diagenetically consolidated zone.

The model strictly relies on fixed Redfield ratios in organic matter (C:P = 117:1). Adsorption and desorption of P on oxides is not taken into account. For example, the oxidation rate of organic carbon, R_{oxy} , is governed by the concentration of oxygen in the pore water ($[O_2]$) and the concentration of POC in the solid phase, c_{POC} :

$$R_{oxy} = r_{oxy}^* c_{POC} [O_2] \quad \text{for } [O_2] > 0$$

r_{oxy}^* is a rate constant. Thus, the higher the oxygen concentration and the higher the POC concentration the larger the flux of carbon from the solid phase to the pore water. Fluxes of other elements are coupled to fluxes of carbon by fixed Redfield ratios (P:N:C:O₂=1:16:117:-170).

The ratio of C:P in organic material found in various sediments has found to deviate by more than an order of magnitude (between 1:20 up to ~4500) from the classical Redfield ratio (1:106). This was interpreted as evidence for preferential burial of organic P compared to organic C under low oxygen conditions and as preferential burial of organic C under high oxygen conditions (e.g. van Cappellen and Ingall, 1994).

This view is challenged by (Anderson et al., 2001) who suggest that early analyses suffered from limited analytical abilities and that detrital P must be distinguished from other phases of P for a correct interpretation of P:C ratios in sediments. These authors define reactive P as the sum of organic P, oxide-associated P and authigenic P and call for the conversion of organic P to oxide-associated P and authigenic P. Anderson et al. state: "*Because most P is delivered to the sediment as P_{organic} and is transformed to P_{authigenic}, sometimes with P_{oxide-associated} as an intermediate phase, organic C/P_{reactive} ratios give a better indication of the original C /P ratios of the organic matter buried in the sediments.*" The mean over all their data yields a C_{organic}/P_{reactive} ratio of 124 +/- 63, indistinguishable from the Redfield ratio used in our model. Thus, they explain high C:P ratios in organic matter within consolidated sediments by the transfer of organic P to authigenic P.

These authors explain the lower than Redfield C:P ratio of organic matter in sediments with low organic carbon (< 2% weight-fraction) with the degradation of POC with age within buried sediments.

Here, we follow the view of Anderson et al. and apply fixed Redfield ratios. Thus the burial flux of POP and POC scale with a factor of 117 and no preferential burial of C or P occurs. We complicity state this fact in the paragraph discussing caveats (p495,114).

The description of the sediment module at the bottom of p480 is modified to read:

A 10-layer sediment diagenesis model (Heinze et al., 1999; Gehlen et al., 2006) is coupled at the ocean floor. It features the same horizontal resolution as the ocean model. It dynamically calculates the transport, remineralisation/redissolution and bioturbation of solid material within the top 10 cm of the seafloor as well as pore-water chemistry and diffusion as described in detail in Tschumi et al. (2011). Modeled tracers are the four solid components (CaCO_3 , opal, POM and clay) and the eight pore water substances (DIC, DIC-13, DIC-14, total alkalinity, phosphate, nitrate, oxygen and silicic acid). The pore water CO_3^{2-} concentration determines whether, and at which rate, CaCO_3 dissolves. The inclusion of the dissolution and burial process of CaCO_3 is crucial for simulating the so-called carbonate compensation. The oxidation rate of POM within the diagenetic zone depends linearly on the pore water concentration of O_2 and the weight fraction of POM within the solid phase. Denitrification is not taken into account in this version of the model. The corresponding reaction rate parameters are global constants and a decrease in the reactivity of organic material by aging within the diagenetic zone is not considered (Middleburg et al., 1993). Fluxes of carbon and related elements due to POC degradation are coupled by fixed Redfield ratios ($\text{P:N:C:O}_2 = 1:16:117:-170$ for oxidation). The model assumes conservation of volume, i.e. the entire column of the sediments is pushed downwards if deposition exceeds redissolution into pore waters. In this manuscript, the term “burial” refers to the net tracer-flux at the ocean–sediment interface, i.e. deposition–redissolution of the particulate material. The burial-efficiency, i.e. the ratio burial/deposition of a solid species, is controlled by i) the rate of redissolution within the sediments and ii) by the rain-rate of solid species, which controls how fast the sediment-column is pushed downwards. Any solid material that is pushed out of the diagenetic zone (top 10 cm) disappears into the subjacent diagenetically consolidated zone. The fate of the material pushed below 10 cm depth is of no further interest for this study (it is known that preferential degradation of POC versus that of POP and the conversion of POP to oxide-associated P and authigenic P within the consolidated zone cause C:P ratios of organic material to deviates substantially from the classical Redfield ratio (Anderson et al., 2001)). Input of terrestrial organic matter into the ocean and burial of terrestrial organic matter is not explicitly considered (see e.g. Regnier et. al, 2013). Similarly, the cycling of P associated with iron and other oxides is neglected as estimates suggest that 97% of the P delivered to the sediment-water interface is in the form of organic matter (Delaney, 1998). The specific chemical composition of the organic matter, particle grain size of the sedimentary material and available area for absorption for organic matter (Hedges and Keil, 1995) as well as spatio-temporal variations in mineral deposition rates or sediment porosity, which likely influence organic matter preservation and burial (Burdige, 2007), are neglected.

3. Towards the end of the transient model runs, the global POM burial rate relaxes to the steady state value determined by the constant riverine phosphorus flux (s. Fig. 3f) while the depositional rate of POM (= POM rain rate to the seafloor) is maintained at an elevated level by the deepening of the remineralisation depth (s. Fig. 3d). The burial efficiency of POM =burial rate/rain rate is thus reduced at this stage (50 – 100kyr) compared to the control run. The authors should explain how their benthic model facilitates this change in burial efficiency. Is this change related to the changing oxygen contents of ambient bottom waters? Where does this change happen: in the deepsea or at continental margins? What is affected: POC, P or both (POM)? The POM deepening experiments result in a dissolved oxygen depletion at the deep-sea floor and an oxygen increase in shallow waters (<1000 m water depth, s. Fig. 5d). Are these changes responsible for the overall decrease in POM burial efficiency and -if so, what model assumptions are made?

The burial efficiency is mainly driven by the availability of oxygen in sediment pore waters and by the total particle rain. The burial efficiency does only slightly change in our standard POM deepening experiment, namely by ~5% from 0.29 to 0.276. The reason for this small change is that the burial-efficiency increase in the deep sea roughly cancels the burial-efficiency decrease at shallow depths. This change is indeed driven by the change in ambient oxygen concentrations (Fig. 5d). The following sentence has been added in the sediment-model description:

"The burial-efficiency, i.e. the ratio burial/deposition of a solid species, is therefore controlled by i) the rate of redissolution within the sediments and ii) by the rain-rate of solid species, which controls how fast the sediment-column is pushed downwards"

The model's response is now discussed on page 486, l2ff. This text now reads:

"Increased POM deposition (rain) tends to increase POM burial and to alter POM oxidation. The change in the amount of POM oxidised in the sediments varies in space and time: in the deep ocean, reduced oxygen availability in the porewater decreases local remineralisation. On the other hand, increased oxygen levels at coastal margins promote the remineralisation in the sediments in these regions. At the equilibrium POM burial rate has to balance input of phosphorus by weathering. The result is an initial spike in POM burial by almost 80%, about four times larger than the relative initial increase in deposition, in response to an initial oxygen reduction in the pore water. Afterwards, POM deposition and POM burial decreases quickly within a few centuries and then more slowly to approach steady state over the next few millennia. As POM deposition stabilizes on a higher level while POM burial relaxes to the initial value, the so-called burial-efficiency=burial/deposition slightly decreases in the long run by ~5%.

4. As far as I understand, the burial efficiency is a key model parameter since it exerts a strong bottom up control on the final steady state results attained in the model runs (including atmospheric pCO₂). The authors should thus carefully explain how their model controls the POM burial efficiency and how the burial efficiencies generated by their benthic model compare to benthic observations.

For a given POM deposition field, the burial efficiency in the model is governed by the availability of oxygen in the pore water affecting POM oxidation and by the deposition of solid material affecting how fast the sediment is pushed towards the consolidated zone. In the long run, POM burial has to match the input flux by weathering in the model. If burial exceeds weathering input, ocean phosphate inventory decline and in

turn export production and POM deposition decline to achieve a balance between burial and weathering flux.

In the global average, our benthic model simulates a steady-state burial-efficiency of ~0.29 which is too high compared with benthic observations of ~0.1 (Sarmiento & Gruber, 2006, page 261, table 6.5.1). This discrepancy is again caused by the poorly resolved bathymetry at continental margins, leading to too low deposition and remineralization and thus the model overestimates the burial-efficiency there with efficiencies close to 1 in certain gridboxes. In the area-weighted global average — which is dominated by the deep ocean — the modelled steady-state burial-efficiency is 0.085 and thus agrees well with observations.

5. The authors should try to discuss to what degree a mismatch between the predictions of the benthic model and benthic observations would affect the major conclusions of their study.

We argue that changes in the imbalance between globally integrated weathering and burial fluxes are the dominant drivers on millennial time scales for changes in seawater chemistry and atmospheric CO₂ in response to changes in the remineralisation depth. The absolute value of the burial-efficiency seems not to be a crucial factor. We therefore believe that our findings are robust, at least qualitatively. A situation where an increased POM rain (due to a slowed watercolumn remineralization) would result in a *decreased rate* of POM burial seems rather unlikely.

Still, we agree that it is important to discuss these points. We added the following lines in the discussion (caveats) section on p495:

"There are also limitations regarding the sediment model. For example, the spatio-temporal variability in the deposition of mineral particles or the influence of particle grain size on organic matter preservation are neglected. The coarse resolution hampers the representation of coastal and continental boundaries, where most POM deposition, remineralization and burial occurs (e.g. Wallmann et al., 2012). The model does not resolve river deltas and estuaries and their carbon cycle (see e.g. Regnier et al, 2013). Another caveat is that denitrification within the sediment is not represented by our model, eventually leading to a bias in the long-term response of POM degradation and thus burial-efficiencies. Therefore, our findings are to be confirmed and refined by a higher-resolved ocean models with a more complete representation of sediment processes."

The paper should definitely be published but it needs a more detailed presentation and critical discussion of the benthic model module.

Done, benthic model description extended (see above).

Further minor comments are given below:

Page 486, bottom: “The adjustment to a new equilibrium takes longer for the phosphorus inventory, co-governing POM burial than for the alkalinity inventory, co-governing calcite burial.” should be replaced by: “The adjustment to a new equilibrium takes longer for the alkalinity inventory, co-governing calcite burial than for the phosphorus inventory, co-governing POM burial.”

done

Page 488 line 20: “The results” should be replaced by “These results”

Text changed to "The result is ..."

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