Response to the review comments

We thank both reviewers for their time and effort to assess our manuscript and for their helpful comments and advice. In the reply, the original review comment is given in small fonts, the reply in normal fonts and changed manuscript text in blue. Page and line numbers are all given for the originally submitted manuscript.

A manuscript with changes highlighted is attached to this reply.

Review 1

Jeltsch-Thommes et al., provide a new estimate of changes in land carbon across the deglaciation. This new estimate is obtained by finding the best fit between paleoproxy constraints and results of factorial simulations with the Bern3D Earth system model coupled to Monte Carlo ensemble.

Their results suggest that there was less land carbon at the LGM than during the Holocene. Their new estimate of deglacial change in land carbon is higher than inferred by recent studies due to the inclusion of sedimentary processes. I recommend publication in Climate of the Past, granted the comments below are taken into account.

Thank you for your support and thoughtful comments.

1) The estimate of land carbon change across the deglaciation given here is higher than previous ones due to sedimentary processes. Since land carbon and organic matter have similar d13C signatures, a deglacial increase in land carbon can be partly compensated by a decrease in organic carbon burial. It is true that in a pure mass balance approach these sedimentary might not be taken into account (even if after ~10k, these effects are not expressed fully, cf. Fig. 4). However, the model used here (Bern3D) includes a sediment model. Therefore changes in temperature, circulation, remineralization profile, etc... lead to changes in organic and carbonate burial. Sedimentary processes are therefore already taken into account in this modelling framework. I am thus confused by the inclusion of an additional “organic weathering flux”. I can indeed imagine that an input of depleted d13C as would happen with a 100% increase in organic matter flux through river could significantly impact oceanic d13C. Maybe the rationale and numbers associated with the organic weathering flux should be explained in more details.

Done. While Bern3D includes an ocean sediment module to simulate dynamically the burial of organic material, the corresponding weathering flux is prescribed to be constant. This is explained in the appendix (page 31, line 7ff), but was not explained in the main manuscript text.

We added the following explanations at P10, L11: The weathering flux to the ocean (Tab. 1) is kept constant in the standard setup of the Bern3D (see also appendix A1), but might have varied with changes in climate and CO2. To account for this possibility, we varied the input of …

and added to the caption of Tab. 1, P7: The tracer input flux to the ocean resulting from weathering is set to compensate for the burial fluxes as diagnosed at the end of the spin up and kept constant in Bern3D standard simulations.

2) [2a] As a follow up on the previous comment, large parameters studies like this one are very useful to test the range of possibilities and derive statistics on possible parameter space. However, as a drawback, the solution can also include parameters that are not really realistic. This is hard to judge in the current state, since some parameters have been varied significantly and some additional description on all processes might be needed.

Figure C2 on P36 shows to which extent the parameters are varied a priori and in the constrained ensemble. We added further text to address this point.

On P9, L10 for the wind stress scaling: We do not consider such large changes in wind stress as realistic, but rather use prescribed Southern Ocean wind stress as a tuning knob to vary deep ocean ventilation in the Bern3D model. Consequently, we do not scale air-sea gas transfer rates with the changes in wind stress, but vary transfer rates independently.
and on P9, L27 for CaCO₃ deposition: [Here, we scale the reconstructed deposition history based on Vecsei and Berger (2004) (Fig. 3D) by applying a constant scaling] to vary CaCO₃ deposition within the published range.

In addition, we added a reference to Figure C2 panel D on P27, L18 and on P9, L31 for the rain ratio we change the last sentence to now read: There is a lack of estimates how the rain ratio has varied in the past. Here, we vary the global rain ratio for LGM conditions (see also Appendix A; Fig 3F; Tab. 2) between 0.045 and 0.098, while the rain ratio is 0.083 in the standard setup and for PI conditions.

and modified the sentence on P27, L6 by adding a reference to Figure C2 and providing additional literature references for the organic matter remineralization depth: A modest, and plausible (Bendtsen et al., 2002; Matsumoto et al., 2007), deepening of the remineralization of organic matter in the upper ocean (see Fig. C2 panel E) leads to …

[2b] For example, there might be some issues with which the changes in “Southern Ocean wind stress” are described. I think the authors start the LGM with weaker southern hemispheric westerlies, and these are linearly increased over the deglaciation. While Table 2 represents changes in parameter values across the deglaciation (e.g. a land carbon uptake of 445, 890 or 1335 GtC), the Southern Ocean windstress is marked as decreasing. This is also the case in Figure 7, with the notation (-10 to -60%), whereas, clearly the impact of increased southern hemispheric westerlies is shown. Same in the notation of Figure 9.

Done. Changed labels in Figs. 7 and 9 to be consistent with Table 2 and added further explanation to the captions of Tab. 2 and Fig. 7 and 9.

Table 2: [Overview of mechanisms and parameters considered in factorial sensitivity experiments. Bold entries mark the standard parameter values.] Values represent the export rain ratio at LGM, the change in remineralization profile (see text) at LGM, the scaling applied to the standard shallow water carbonate deposition history, and the land carbon uptake over the deglaciation in GtC. In the case of SO wind stress, SO gas transfer rate, and the organic weathering flux, values represent the ratio of LGM to PI forcing.

Fig. 7: [Sensitivities are shown relative to the standard forcings (white dots).] Values in panel (C) correspond to the entries in Tab. 2.

Fig. 9: [Simulated deglacial change in the isotopic signature of DIC, ∆δ¹³C_DIC (PI minus LGM). Values are displayed along sections across the Atlantic, Southern Ocean, and Pacific and in response to standard deglacial forcings and (B) to (H) due to changes in individual mechanisms as inferred from factorial simulations.] ∆δ¹³C_DIC is shown for a deglacial carbon uptake of 445 GtC by the land biosphere (B), Southern Ocean wind stress (C) and gas transfer rate (D) scaled by a factor of 0.4 during the glacial relative to PI, rain ratio reduced to 0.045 during the glacial(E), deglacial shallow water carbonate deposition scaled by a factor of 4 (F), a change in the organic matter remineralization between a linear profile during the glacial and the standard depth-scaling (G), and for a twofold increase in the organic weathering flux during the glacial compared to PI (H). [The absolute change in….]

[2c] P23, paragraph starting L.8: It might be easier to follow if everything goes in the sense of the deglaciation, i.e. describe an increase in the SO winds.

We modified the paragraph as requested and in addition clarified the description of the experimental setup at various places in the manuscript.

First, we note that the experimental setup used in this study differs from a classical step-change approach as, e.g., in Tschumi et al. (2011). We clarified the forcing history in the method section by replacing the sentence on P7, L9-10 with: The forcing history for changes in Southern Ocean wind stress, Southern Ocean gas transfer rate, the rain ratio, the remineralization profile, and the organic weathering rate are prescribed following an idealized glacial-interglacial evolution. PI values are set to LGM values at 40 yrBP, kept constant from 40 kyrBP to 18 kyrBP, scaled back to the PI value over the termination (18 kyrBP to 11 kyrBP) and kept constant thereafter (Fig. 3F and Tab. 2).
Changes in the other four mechanisms affect the spatial pattern of $\Delta^{13}C_{\text{DIC}}$ significantly (Fig. 9C, D, E, and G). We note that these patterns result from adjustment processes acting on different timescales. We recall the forcing history for changes in Southern Ocean wind stress, Southern Ocean gas transfer rate, the rain ratio, the remineralization profile, and the organic weathering rate; PI values are set to LGM values at 40 kyrBP, kept constant from 40 kyrBP to 18 kyrBP, scaled back to the PI value over the termination (18 kyrBP to 11 kyrBP) and kept constant thereafter (see also Fig. 3 and Table 2). Results are therefore not directly comparable to the step-change experiments described by Tschumi et al. (2011). As discussed in detail by Tschumi et al. (2011) and Menviel et al. (2015), a more poorly ventilated ocean due to reduced wind stress leads to an increase in $\delta^{13}C_{\text{atm}}$ and a subtle initial decrease in $\delta^{13}C_{\text{DIC}}$. Changes in temperature and circulation in response to decreased Southern Ocean wind stress at the beginning of our simulation (Fig. 3F) lead to lower marine oxygen concentrations and thus an increase in POC sedimentation. Subsequent removal of $^{13}C$ depleted carbon from the ocean increases $\delta^{13}C_{\text{DIC}}$. Restoring wind stress to its initial value over the deglacial reverses the changes. As the sediment feedbacks act on longer timescales, propagation of the signal to the whole ocean is not yet achieved. A complex interplay of processes such as changes in circulation and subsequent changes in export fluxes, oxygen concentrations, and remineralization of POC as well as changes in the lysocline and CaCO$_3$ cycling spatially overlay each other, yielding the $\Delta^{13}C_{\text{DIC}}$ distribution seen in Fig. 9C.

Further, Fig. 3 was changed to show the whole setup, not only the deglacial and the following two sentences added to the figure caption: The freshwater forcing in the North Atlantic is varied over the last 40 kyr following Menviel et al. (2012). Dashed green lines indicate spin-up values.

[2d] Similarly, due to changes in winds and sea-ice the Southern Ocean gas exchange should increase across the deglaciation.

See comment to previous point. Text on P23, L19 was changed to read: [Decreasing the gas transfer rate] at the beginning of the 40 kyr simulation (Fig. 3F) [leads to a strong increase in...] A change in the remineralization profile across the deglaciation is fine, and remineralization would be expected to become shallower. However, it is not clear to me what is the rationale behind choosing “a linear profile”. In between which depths was the profile made linear?

For clarification we show the linear profile here (Figure 1) and added the following text on P10, L7: [...] and a linear profile (75 m to 2000 m).

![Figure 1: Fraction of remaining particulate organic matter (indicated by the amount of particulate organic phosphorus, POP) leaving the euphotic zone for the standard and linear remineralization profile. $z_0$ refers to the bottom of the euphotic zone in the model at 75 m.](image)
and moved and streamlined the text from P27, L11-17 to P10, L7: [The modification in the remineralization profile of particulate organic matter is different than those applied by Menviel et al. (2012) and Roth et al. (2014) in simulations with the Bern3D model. Here, only the remineralization profile in the upper 2,000 m is changed and, in contrast to these earlier studies, it is assumed that the fraction of exported particles that reach the sediments below 2,000 m is not altered. The implicit assumption is that cooler temperatures at the LGM only significantly affect the dissolution of relatively small or labile particles in the upper ocean, while large or refractory particles sink fast enough to reach ocean sediments both under LGM and PI conditions].

Based on the “best guess scenarios” of the parameters space, what is the most plausible change in land carbon?

We are not sure we understand the question of the reviewer. Change in land carbon storage is determined in the Bayesian framework and results in a median value of 850 GtC as given in the abstract. Change in land carbon is used as one of seven input parameters for the emulator and as such difficult to assess as a function of the other six forcing parameters.

3) I am a bit surprised by the sign of the oceanic d13C change when the SO wind increases. From figure 9, it seems like the stronger wind does not increase deep ocean ventilation: Tschumi et al., 2011 simulates positive d13C anomalies in the deep Pacific.

Several aspects lead to the differences in oceanic δ13C change as shown in Fig. 9C versus those given by Tschumi et al. in their Fig. 9. Most important is the different setup of the experiments as discussed above. When applying a step-increase in Southern Ocean wind stress to 180% as done in Tschumi et al. (2011), we obtain very similar results as illustrated in Figure 2 here below. Further differences arise from the different model resolution used by Tschumi et al. (2011) and in this study. See our answer to point [2c] above for changes to the MS.

4) In some of the experiments described in the manuscript it seems that global alkalinity changes. For example, p. 19, L.10: “a deglacial decrease in deep Pacific carbonate ions” is needed to explain the deglacial CO2 increase. Since [CO3]~[ALK]-[DIC], and since deep Pacific DIC is expected to decrease across the deglaciation, that means that the alkalinity must decrease more than DIC. Additional information on the magnitude and processes leading to the alkalinity decrease might be needed.

We added the following sentences on P19, L10 to provide an insight into DIC and Alk changes: Changes in alkalinity and DIC (PI-LGM) for the standard forcing amount to about 500 Gt eq and 365 GtC, respectively. In the case of rain ratio, coral reef growth, and changes in the remineralization profile, changes in alkalinity and DIC are largest and occur close to a 2:1 ratio. Changes in land carbon uptake
remove carbon from the system, leading to changes in alkalinity via CaCO$_3$ compensation whereas in the case of the organic weathering flux, not only carbon is added/removed from the system but also nutrients and alkalinity, following Redfield ratios (see appendix \ref{app:b3d}). Changes in alkalinity and DIC resulting from changes in Southern Ocean wind stress and gas transfer rate are small.

Minor points and typos:
- P3, L.32: please add “across the deglaciation”
  Done
- P4, section 2.1: A minimum information about the Bern3D is needed without having to go through the appendix: which components does the model include? What is the resolution of the model?
  The following text is added at P4, L25: It couples a dynamic geostrophic-frictional balance ocean, a thermodynamic sea ice component, and a single layer energy-moisture balance atmosphere. The horizontal resolution is 41x40 grid cells and the ocean has 32 layers. Marine productivity is simulated as a function of nutrient concentrations (P, Fe, Si), temperature and light and transferred to dissolved organic and particulate matter. Biogenic matter decomposes within the water column. Opal, calcite, and organic particles reaching the ocean floor are entering reactive sediments. A 10-layer sediment model is used to compute fluxes of carbon, nutrients, alkalinity and isotopes between the ocean, reactive sediments, and the lithosphere. Loss fluxes to the lithosphere are compensated at equilibrium by a corresponding input flux from weathering. A 4-box reservoir model is used to calculate the dilution of atmospheric isotopic perturbations by exchange with the land biosphere.
  Done
- P9, L.18: Please spell out “std”
  Done
- P10, para L. 14: This paragraph might need a bit of rephrasing. I think the argument of the authors here is that changes in Fe fertilization lead to changes in the oceanic organic matter content. These changes can somewhat be broadly included in the changes associated with “organic weathering” in their set of simulations.
  Done. We replaced the sentence on L15 (“The effect of iron fertilization on oceanic carbon and $\delta^{13}$C$_{DIC}$ is similar to the mechanisms outlined above and is thus not explicitly considered further.”) by:
  Iron fertilization leads to changes in the cycling of marine organic material and to related changes in the whole ocean inventory of $\delta^{13}$C$_{DIC}$ and other variables. These changes are broadly similar to the changes caused by altering the organic matter remineralization depth or the weathering input flux and implicitly included in the Monte Carlo simulations. Similar, other ..
- P27, L. 5: a parenthesis is missing
  Done
- P27, L. 8: “phosphorus”
  Done
- P29, L.5: “biogeochemical”
  Done
Review by Katsumi Matsumoto

This ms presents new modeling results of carbon redistribution in an earth system model, focusing on the change in land biosphere carbon storage between LGM and PI. It considers open system processes (i.e., weathering and burial) and uses observations of δ13C of atm CO2, δ13C of oceanic DIC, atm pcO2, and deep ocean [CO3=] as constraints. Since the seminal work by Shackleton (1977) the change in carbon storage on land over glacial-interglacial time scale has been an important topic in paleoclimatology and global carbon cycle. This ms is thus appropriate for CP readership.

The extensive simulations and analysis will add to the literature, and I am generally supportive of the study. However, I find the current form of ms requires some effort on the part of the reader to finish reading to the end, because it is quite long, a bit repetitive, and unclear in some places. Consequently this manuscript may not be as impactful as it can be. I make the following comments/questions/suggestions as a way to help improve this ms generally and increase its impact.

Thank you for your support and for your useful comments.

1) Overall the ms is long. There is some repetition of information in the Discussion that rehashes the Introduction without necessarily adding value. Effort should be made to shorten not just the Discussion but earlier sections too.

We deleted/shortened/streamlined text in several places in the manuscript. Large changes are listed below, small changes are highlighted in the attached diff file for better readability.

- P1, L1-L6. First sentence of abstract deleted and following text shortened to read: Past changes in the inventory of carbon stored in vegetation and soils remain uncertain. Earlier studies inferred the increase in the land carbon inventory (Δland) from the Last Glacial Maximum (LGM) to the recent preindustrial period (PI) from reconstructions of the stable carbon isotope ratio in the ocean and atmosphere with recent estimates yielding 300-400 GtC. Surprisingly...

- P1, L14-18: Text modified and shortened to read: Our study demonstrates the importance of ocean-sediment interactions and burial and weathering fluxes involving marine organic matter to explain deglacial change and suggests a major upward revision of earlier isotope-based Δland estimates.

- P1, L12: new sentence added. It is highly unlikely that the land carbon inventory was larger at LGM than PI.

- P2, L18-32 on earlier estimates of changes in land carbon since LGM in the introduction; part of its content is included in section 4.3 in the discussion and deleted in the introduction.

- P3, L7: Sentence deleted (‘The details of the approach...’)

- P3 L14 -21

- P3, L29-31

- P4, L10-L12

- P5, we simplified Figure 1 and shortened the caption by deleting the text from L2 to L5 (The targets ..)

- P14, L33: Deleted sentence 'We now turn...'

- P16, L6-7: replaced sentence with: We stratify the results by Δland and define four sets...

- P17, L8-9: Deleted sentence ‘Visual inspection...’

- P21, L27-28: Deleted sentence ‘The small mean change...’

- P24, L9-10: Deleted sentence ‘These geological...’

- Deleted paragraph starting P24, L19
There are a number of uncertainties in our approach. We rely on characteristic forcing–response relationship as obtained with the Bern3D model and these may be different than in reality. The current crop of models...

The deglacial change in CO$_2$ stored in the unsaturated zone of aquifers is in the range of 12 to 86 GtC (Baldini et al., 2018) and small compared to the estimated change in land carbon of around 850 GtC. Perhaps more important, the modern inventory of refractory DOC is about 650 GtC (Hansell, 2013) and assumed constant in Bern3D, while changes in the small inventory of labile DOC are explicitly simulated. It is unclear how the DOC pool varied over the past. Further, [Cartapanis et al. (2016) suggest an approximately...]

Here, we varied organic-like carbon input by weathering in the unconstrained ensemble over a range corresponding to a cumulative PI-LGM anomaly of +780 to -1,560 GtC. [These anomalies in input...]

P26, L12-19: Deleted

Our approach is limited in that we only consider the change between LGM and PI for four relevant proxy variables. This restriction allowed us to build a cost-efficient and non-linear emulator to explore a very large range of parameter combinations. Future efforts to refine estimates of Δland may explicitly consider the spatial and temporal evolution for a large number of proxies.

P27, L18-21: Deleted (‘A decrease in...’ until ‘...by organic material.’

P27, L33-P28, L1 deleted ‘each associated with its own uncertainties and limitations.’

P27, L25-30: Deleted here and last two sentences moved to beginning of section 4.

P28, L3-5: Deleted sentence and shortened.

P28, L8-15: Shortened to read: The ice core CO$_2$ and δ$^{13}$C$_{atm}$ records constrain the preindustrial terrestrial carbon uptake over the Holocene to about 250 GtC (Elsig et al., 2009). Assuming the total estimate of 1,500 GtC by Adams and Faure (1998) was right...

P28, L30-31: Deleted sentence

2) [2a] Even while the title and Figure 3 try to impress that reader that the authors are investigating the entire deglacial, I do not get a full sense of this. Most of the study is actually focused on differences between two times, LGM and PI (Figs 5-9). There is not a single figure that shows model results of the entire transient deglacial that corresponds to the deglacial forcing shown in Fig 3.

[2b] Related to this title-content mismatch is that the authors present Delta (e.g., Dland) as PI minus LGM. I find this to be unnecessarily confusing, because what is changing is the simulation results of LGM. PI remains the same. A main focus is on explaining LGM land carbon storage. In their way of presenting, when LGM land C increases, Dland decreases. As a reader, I have to do that mental calculation and commit what little RAM I have in my brain to storing that information...making reading difficult. For example, as an explanation of Fig 9, I found it a lot easier to understand “a weaker and shallower Atlantic Meridional Overturning Circulation at the LGM compared to the PI” (line 6, p 25) than “the deepening and strengthening of the AMOC during the deglaciation lead to positive Dd$^{13}$C$_{DIC}$ changes in the deep Atlantic...” (line 24-26, p 21). They are saying the same but PI-LGM is harder, at least for me.

[2c] My preference/suggestion therefore would be that the authors focus on time slice comparisons and present Delta as LGM-PI.

[2a] We changed the title to read: Low terrestrial carbon storage at the Last Glacial Maximum: constraints from multi-proxy data

[2a] Our goal is to answer a scientific question. Namely, how large is the difference in global land carbon storage between the LGM and the PI. This is spelled out in the abstract and the introduction and
conclusions. To answer this question, we carried out more than 100 transient simulations over the deglacial period and figure 3 illustrates the experimental setup and how the model is forced. We keep figure 3 as it is an integral and important part of our method description. We have the impression from this comment and also from comment 7 below that we failed to explain the main purpose of our deglacial simulations – this is to quantify the influence of deglacial processes on the whole ocean δ\textsuperscript{13}C budget and therefore on estimates of Δland.

We modified the text in the introduction on P3, L29, P3, L32-34, on P4, L12, and reorganized the paragraph on P4, L13-L22 to read:

P3, L29: added text: These mechanisms should not be neglected when addressing the whole ocean budgets of carbon and δ\textsuperscript{13}C to infer changes in land carbon stocks.

P3, L32-34, reorganized and expanded sentence: The transfer of \textsuperscript{13}C and carbon between the ocean-atmosphere system and reactive sediments and the lithosphere is affected by the well-documented reorganization of the marine carbon cycle across the deglaciation (e.g. Sigman and Boyle, 2000; Sigman et al., 2010; Fischer et al., 2010; Elderfield et al., 2012; Ciais et al., 2013; Martínez-García et al., 2014; Jaccard et al., 2016; Cartapanis et al., 2016).

P4, L12-14 added text: As a consequence, these deglacial changes and their spatio-temporal evolution affect the mean δ\textsuperscript{13}C signature of the global ocean in addition to changes in land carbon.

P4, L13-22; reorganized/modified paragraph: The primary goal of this study is to provide a new, observationally constrained best estimate of Δland recognizing the role of marine deglacial processes, weathering and burial, and sediment interactions in an Earth system model. To this end, we establish the response sensitivities for mean ocean δ\textsuperscript{13}C and for other proxy targets (PI-LGM differences) to the changes in individual key deglacial carbon cycle mechanisms from transient simulations. An emulator of the Bern3D is built and the strengths of individual marine and terrestrial processes are varied in a large number of combinations to find all possible solutions for Δland that match a set of observational targets in a Bayesian Monte Carlo data assimilation framework (see Fig. 1). We further scrutinize the closed system assumption in the \textsuperscript{13}C mass balance approach with land carbon uptake as the only forcing. Specifically, we run idealized pulse-like carbon uptake simulations with the Bern3D Earth System Model of Intermediate Complexity (EMIC) both with and without simulating sediment interactions, and with and without interactive weathering.

We further discuss multi-proxy response relationships for different carbon cycle processes and briefly address the possible contribution of individual mechanisms to the deglacial change in atmospheric CO\textsubscript{2} and δ\textsuperscript{13}(CO\textsubscript{2}) and of deep ocean carbonate ion concentration. Our results demonstrate the importance of different deglacial mechanisms, of ocean-sediment interactions, and of the burial-weathering cycle for the whole ocean δ\textsuperscript{13}C signature. All previous δ\textsuperscript{13}C-based estimates of Δland neglected the influence of these processes and appear systematically biased low.

In agreement with the advice given by the reviewer in [2c], we continue to focus on the comparison between the LGM and PI time slice.

[2b] Please see also our answer to reviewer 1, comment [2c] and to your comment 1 and our revision of the abstract.

We appreciate the advice regarding the presentation of results and attempted to harmonize text formulations (as well as figures and tables) to the extent possible. Reviewer 1 and this reviewer, however, provide conflicting advice how to present the results. Reviewer 1 suggests (comment 2c) that “It might be easier to follow if everything goes in the sense of the deglaciation.” We prefer to follow the advice of reviewer 1 as it is likely easier for most readers to go forward in time in the sense of the deglaciation. It might also be confusing to talk about a negative glacial-interglacial change in atmospheric CO\textsubscript{2} with ΔCO\textsubscript{2} = -90 ppm. Therefore, we continue to present changes as differences between PI minus LGM.
We disagree somewhat with the argument of the reviewer that “what is changing is the simulation results of LGM. PI remains the same.” We are only able to constrain the change in land carbon storage, Δland, but do not attempt to estimate the PI carbon inventory. Further, we apply a transient protocol for our simulations which covers the past 40 kyrs as now clarified in the revised Fig. 3.

3) One of the main conclusions is that glacial land C was not larger than PI land C, thus refuting Zimov and Zech... This is a little bit of attacking the straw man, because the notion that glacial land C was larger is highly speculative to begin with, as the authors note (line 21-23, page 2). The more serious and “arguably most reliable” (line 33, page 2) hypothesis of glacial land C is that constrained by C isotopes by Shackleton. It seems to me that the main scientific contribution of this paper is that it credibly revised upwards Shackleton’s estimate by considering open system processes, not refuting a straw man argument. I would suggest that the ms be rewritten to make clear what the real scientific contribution is.

Please see our revision of the abstract in response to your comment 1. For example, we do not cite the literature range for Δland of -400 to 1500 GtC range anymore in the abstract and emphasize the upward revision of earlier δ13C based estimates in the newly added last sentence.

We shortened the text on the papers by Zimov et al. and Zech et al. in the intro on P2, L21ff (see response to comment 1 above).

We state now at the end of the introduction (see also response to [2a] above): All previous δ13C-based estimates of Δland neglected the influence of these processes and appear systematically biased towards lower estimates.

A recent publication by Kemppinen et al., 2018 suggests based on a large ensemble of EMIC simulations that land carbon storage was larger at the LGM than PI. Thus, there are at least three studies that suggest a higher carbon storage at LGM than at PI and it seems not justified to entirely ignore these. We state in the revised abstract that a larger land carbon inventory at LGM than PI is highly unlikely.

4) Why is the control PI run biased high in d13C of DIC? (line 11, page 6)

Several reasons potentially lead to this constant offset. During the spin-up, the model was forced with a prescribed δ13C = -6.305‰, while δ13C = -6.4‰ might have been preferable for AD 1765 conditions. The implementation of fractionation during air-sea gas exchange and primary production in the ocean follows the literature (Mook, 1986; Freeman and Hayes, 1992). Already small uncertainties in these formulations will lead to offsets in the equilibrium values.

5) I realize that details of ocean biogeochemistry is not central to this ms, but some more description would have been nice... e.g., how many phytoplankton functional types are there that are limited by P, Fe, and Si? (line 13, page 30)? How much did new production change in the “standard” glacial run compared to PI in terms of Dd13C (Fig 9a) or contribute to the DCO2=27.8 ppm? The authors adjusted the remin depth scale later to modify the impact of new production, but was it that the new production was not sensitive to nutrient supply changes in the standard run?

P30, L15: Text added: Bacteria and plankton are not explicitly represented.

P17, L16: Text added: New production of organic matter decreases from 11.98 to a mean value over the glacial of 11.25 GtC/yr.

We did not quantify the influence of the new production change on atmospheric CO2. This is beyond the scope of this study. This would require additional factorial simulations and an adjusted model setup. The quantification is also not straightforward as ocean circulation changes affect the three classical carbon pumps all simultaneously.
6) To me, the pulse experiments (section 3.1) are novel and the most interesting part of this study. It shows that neglecting the open system processes, as earlier studies have done, leads to underestimation of Dland. I think though, this section needs more clarification and discussion.

[6a] I was confused about the 100 Gt negative pulse emission in a closed system (line 26-29, page 12). . .what this is supposed to represent, because in reality 100 Gt does not disappear. It gets redistributed. I think it’s supposed to represent the terrestrial uptake, no? If so, it should be stated plainly. Otherwise, you’d have to put it into the ocean (after all, it’s a closed system), so that d13C of atm CO2 should increase but d13C of DIC should decrease (not increase as in Fig 4B unless 100 Gt is magically removed or taken up by land).

The starting sentence of section 3.1, P12, L26 is modified to: We start by investigating how atmospheric CO2 and δ13C signatures evolve in response to carbon uptake by the land biosphere when all other forcings remain unchanged. To this end, 100 GtC of light carbon (δ13C=-24 ‰) are instantaneously (in the first time step of the year) removed from the model atmosphere to test the Bern3D response for different model setups (see Appendix A3). This yields the Green’s functions (or impulse response functions) for carbon and 13C shown in Fig. 4.

We added references for the CO2 closed system pulse response on P13, L1 to point the reader to publications discussing the value of and the theory behind impulse response functions: ..by a typical impulse-response recovery (e.g. Maier-Reimer and Hasselmann, 1987; Siegenthaler and Joos, 1992; Joos et al., 2013)

and the δ13C response in the closed system P13, L5 ..(Fig 4A; Joos et al., 1996)

and the CO2 open system response on P13, L10: The evolution of atmospheric CO2 (see Archer et al., 1998) … , while we are not aware of a peer-reviewed publication showing the isotopic response for the open system.

[6b] By the way, I found the inverted y-axis of d13Catm (Fig 4A) to be confusing. Why invert? The reader has to commit that to memory as well.

Axis adjusted as requested.

[6c] Why does d13C_atm recover faster than pCO2 (Fig 4A, line 4, page 13)? In standard chemical oceanography, we learn that C isotope equilibration is a lot longer than CO2 chemical equilibration in the surface ocean (roughly 10 yrs. vs. 1 yr).

Indeed, this is standard chemical oceanography knowledge, though maybe it is not understood. We added the following sentence on P13, L6: The difference in the response of CO2 and δ13C is related to their different properties. CO2 represents a concentration, whereas δ13C stands for the 13C to 12C isotopic ratio. The capacity of the ocean to remove an atmospheric perturbation in CO2, and equally in 13CO2 and 12CO2, is limited by the acid-base carbonate chemistry described by (Revelle and Suess, 1957). In contrast, the removal of a perturbation in the isotopic ratio 13CO2/12CO2 is hardly affected by this chemical buffering; the buffering affects 13CO2 in the nominator and 12CO2 in the denominator about equally leading to a near cancellation of its effect on the ratio. The resulting change...

Regarding the specific point about surface ocean equilibration by air-sea exchange, it may be useful to consider the atmosphere and surface ocean while not considering exchange with the deep. At equilibrium, the perturbation in the isotopic ratio for atmospheric CO2 and surface ocean DIC become approximately equal. The atmosphere, in the form of CO2, and the surface ocean, in the form of DIC, contain about equal mass of carbon. Accordingly, half of the initial perturbation in the isotopic ratio is removed from the atmosphere to the surface ocean.

In contrast, the acid-base carbonate chemistry leads to a much smaller relative perturbation in DIC than in CO2 (acid-base buffering). With a Revelle factor of 10, roughly 90% of the initial perturbation remains in the atmosphere and only about 10% enters the surface ocean.

The surface ocean DIC concentration is equilibrated quickly as the surface layer only absorbs 10% of the initial perturbation. In contrast, the isotopic equilibration is slower as a larger fraction of the perturbation has to be taken up. This leads to the faster decline in the impulse response function (IRF) of δ13C_atm
compared to the IRF of CO$_2$. This reasoning can be extended considering surface-to-deep mixing and the whole ocean DIC content.

[6d] What is the authors’ definition of achieving equilibrium (line 11, page 13)? 60 kyr seems quite long as a time scale of carbonate compensation.

Text on P13, L11 modified to read: ... and equilibrium is approached with an e-folding time scale of about 14,000 years.

[6e] The most interesting part is the open system response of d$\delta^{13}$C of DIC (line 3-16, page 14). I feel that the main message is a little bit lost in the details. Is another way of putting it that open system damps out the penetration signal of light d$\delta^{13}$C from the atmosphere into the ocean? So when benthic foraminifera records seawater d$\delta^{13}$C, its perturbation signal would appear to be much smaller than closed system/actual, and therefore the reconstructed D$\text{land}$ would be underestimated?

Text from P13, L13 ff reformulated to read: [... thereafter.] The novel and most important finding from the pulse experiment is that the $\delta^{13}$C perturbation in the ocean-atmosphere-land biosphere system is increasingly and eventually completely removed by open system processes. Fig. 4B shows the $\delta^{13}$C$_{\text{DIC}}$ perturbation to decrease on a multi-millennial timescale in contrast to the closed system assumption (dashed lines 4B). In other words, benthic foraminifera record a much smaller perturbation in seawater $\delta^{13}$C$_{\text{than}}$ expected from closed system modelling. The resulting difference in the $\delta^{13}$C$_{\text{DIC}}$ perturbation between the open and closed system causes an erroneous mass balance inference for the terrestrial biosphere, when applying conventional closed system equations (Eq. A2 and A3). On a timescale of 10 kyr, underestimation of the terrestrial carbon inventory change amounts to 30 %. The error increases as times evolves.

In detail, the difference between the open and closed system pulse responses is explained as follows. The $^{13}$C budget in the open system changes due to imbalances between input and burial fluxes of POC and CaCO$_3$ and due to changes in the $\delta^{13}$C signatures of the respective fluxes.

The uptake of isotopically light land carbon from the atmosphere leads to an increase in the $\delta^{13}$C signature of surface waters and by that in the $\delta^{13}$C signatures of exported and eventually buried POC and CaCO$_3$. The mean $\delta^{13}$C signature of the burial flux amounts to -8.7‰ ($\delta^{13}$C$_{\text{CaCO}_3}$: 2.9‰) and $C_{\text{org}}$ ($\delta^{13}$C$_{\text{POC}}$: -20.2‰) for the first 10 kyr compared to -9.1 ‰ for the weathering input. This difference tends to mitigate the $\delta^{13}$C$_{\text{DIC}}$ perturbation. Further, carbon–climate feedbacks cause changes in temperature and ocean circulation, which affect the export of POC and CaCO$_3$. Burial fluxes of POC further depend on O$_2$ concentrations and burial fluxes of CaCO$_3$ on CO$_3^{2-}$ concentrations which evolve over the course of the simulation. As a result, the cumulative sedimentation-weathering imbalance amounts to a removal of 48.5 GtC during the first 10 kyr. The contribution to differences in the carbon isotopic budget relative to the closed system is dominated by changes in POC cycling and its associated $\delta^{13}$C signature, while changes in the CaCO$_3$ cycle play a smaller, but non-negligible, role. Overall, $^{13}$C is lost from the atmosphere–ocean system and $\delta^{13}$C$_{\text{DIC}}$ is decreasing.

[6f] I think you mean, compared to the “closed system,” not the “open system” in line 14, page 13.

Done, text deleted.

[6g]I don’t understand “the removal of light terrestrial carbon” in line 5-6, page 14. I think this whole section should be more carefully worded, perhaps even expanded, so that its important message is up front and clearer.

Done, see response to 6e

7) [7a] Pages 16-17 make clear that d$\delta^{13}$C of DIC provides the single most important constraint on D$\text{land}$, which I believe is what most people in the community thought anyway: “arguably most reliable” (line 33, page 2). So it seems to me that, in the bigger scheme of things, the place of this work in the literature is that it builds on the C isotope estimated D$\text{land}$ by making the important point that open system processes lead to underestimation. I think that is how the ms should be framed, not refuting straw man argument of Zimov or trying to explain glacial pCO$_2$. . .
The statement of the reviewer on the importance of $\delta^{13}C_{DIC}$ is incorrect and not supported by our results – see Figure 5. We started a new paragraph on P15, L5 to make the point more readily accessible by the reader and modified the text from L5-8 to read:

Next, we show that it is not sufficient to use $\Delta\delta^{13}C_{DIC}$, or any other target, in isolation to constrain $\Delta$land. The probability distribution of $\Delta$land covers the whole range of sampled values almost uniformly (Fig. 5) when applying only one out of the four targets. Considering either $\Delta\delta^{13}C_{DIC}$ or $\Delta\delta^{13}C_{atm}$ together with $\Delta$CO$_2$ as constraints moves the $\Delta$land distribution towards the multi proxy-based distribution (blue line in Fig. 5; not shown). We conclude that the incorporation of multiple constraints is essential to narrow uncertainties in $\Delta$land.

Please see our response to your comments 1 and 3 with respect to the manuscript framing and Zimov et al.

[7b] We do not agree with the reviewer. Please see our response to your comment 2b above. In addition, we modified the last paragraph of section 3.1 to read:

The pulse responses make it clear that open system processes such as sediment interactions, burial, and weathering affect the carbon and $\delta^{13}$C budgets on deglacial time scales. Taken at face values, the results suggest that the classical $\delta^{13}$C-based estimates of $\Delta$land are substantially biased low, because of the neglect of open system processes. However, uptake by land carbon is the only forcing in these idealized pulse simulations, whereas climate and biogeochemical cycles underwent a massive reorganization over the glacial-interglacial transition. This reorganization affected open system processes and the $^{13}$C budget in addition to carbon uptake by the land biosphere. In the next section, we estimate $\Delta$land by taking into account deglacial processes.

[7c] Reference added.

[7d] The statement by the reviewer is incorrect. It is stated at several places in the MS that we use all four target variables as constraints and that all four variables must fall within the proxy range given in Tab. 2. Fig. 6 shows the kernel density estimates for the four targets in the constrained ensemble. In addition, Fig. C2 in the appendix gives the posteriori parameter distribution. We believe that this point is made sufficiently clear in the revised MS.

8) The ms has awkward phrasings/words here and there, which need to be rephrased. The following is not an exhaustive list:
- is “paleo soils” (line 22, p.2) correct? is it “paleosols”
  Done
- replace “by” with “to” (line 24, p. 3)
  Done
Meant is the buried oceanic organic material.

- insert “on land” after “organic material” (line 10, p.10)

Done

- insert “the pulse to” after “apply” (line 27, p. 12)

Done

- insert “marine” before “sediments” (line 10, p. 13)

Done

- last line, p.13 is confusing; rephrase

Sentence changed

- replace “recognize” with “represent” and rephrase rest of sentence (line 33, p. 14)

Done

- line starting “Also, it is visible. . .” (line 7, p. 17) is confusing; rephrase

Done

- line starting “In summary. . .” (line 28, p. 17) makes little sense; rephrase

Shortened and changed

- insert “of” at the end of line 16, p. 19 after “mechanism”

Done

- line starting “Though, the overall. . .” (line 18, p. 19) is awkward; rephrase

Sentence deleted, information was not essential

- replace “The following” with “Subsequent” on line 23, p. 10

Done

- replace “build” with “built”, line 19, p. 24

Done

- line 30, p. 25: Is “Information in” necessary? Or are the authors inferring from Baldini’s study? Awkward in any case. . .rephrase.

Done

- misspelling of phosphorus, line 8, p. 27

Done

- a brief description of the 4 box veg model might be helpful

Provided in Appendix A


Revelle, R., and Suess, H. E.: Carbon dioxide exchange between atmosphere and ocean and the question of an increase of atmospheric CO$_2$ during the past decades, Tellus, 9, 18-27, 1957.


A large increase in the Low terrestrial carbon inventory of the land biosphere since storage at the Last Glacial Maximum: constraints from multi-proxy data

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Abstract. Atmospheric CO₂ increased by about 90 across the transition from the Last Glacial Maximum (LGM) to the end of the preindustrial (PI) period. The contribution of changes in land carbon stocks to this increase remains uncertain. Estimates of the PI-LGM difference in land biosphere Past changes in the inventory of carbon stored in vegetation and soils remain uncertain. Earlier studies inferred the increase in the land carbon inventory (Δland) range from −400 to +1,500 GtC, based on upscaling of scarce paleo soil carbon or pollen data. A perhaps more reliable approach infers Δland from reconstructions of the between the Last Glacial Maximum (LGM) and the preindustrial period (PI) based on marine and atmospheric stable carbon isotope ratio in the ocean and atmosphere assuming isotopic mass balance with recent studies yielding Δland values of about 300–400 GtC. Surprisingly, however, earlier studies considered a mass balance for the ocean–atmosphere–land biosphere system only. Thereby Notably, these studies neglect carbon exchange with marine sediments, weathering-burial flux imbalances, and the influence of the transient deglacial reorganization on the isotopic budgets. We show this neglect to significantly bias low deglacial simplification to significantly reduce Δland in simulations using the Bern3D Earth System Model of Intermediate Complexity v2.0s. We constrain Δland to ~850 GtC (median estimate; 450 to 1250 GtC) by using reconstructed changes in atmospheric δ13C, marine δ13C, deep Pacific carbonate ion concentration, and atmospheric CO₂ as observational targets in a Monte Carlo ensemble with half a million members. It is highly unlikely that the land carbon inventory was larger at LGM than PI. Sensitivities of the target variables to changes in individual deglacial carbon cycle processes are established from factorial simulations over the past 21,000 years transient factorial simulations with the Bern3D model. These are used in the Monte Carlo ensemble and provide forcing–response relationships for future model–model and model–data comparisons. Uncertainties in the estimate of Δland remain considerable due to model and proxy data uncertainties. Yet, it is likely that Our study demonstrates the importance of ocean–sediment interactions and burial as well as weathering fluxes involving marine organic matter to explain deglacial change and suggests a major upward revision of earlier isotope-based estimates of Δland is larger than 450 and highly unlikely that the carbon inventory in the land biosphere was larger for the LGM than during the recent preindustrial period.
1 Introduction

Atmospheric CO\(_2\) varied between about 180 and 300 ppm over the past 800,000 years (Neftel et al., 1982; Siegenthaler et al., 2005; Lüthi et al., 2008; Marcott et al., 2014; Bereiter et al., 2015). These CO\(_2\) variations were tightly coupled to glacial–interglacial climate change (Petit et al., 1999; Siegenthaler et al., 2005) and amplified orbitally driven climate variations (Jansen et al., 2007). Despite their climatic importance, the exact mechanisms behind these past CO\(_2\) variations remain enigmatic. A wide range of explanatory processes that are related to the marine and terrestrial carbon cycle as well as to exchange processes with reactive ocean sediments, coral reefs, and the lithosphere has been proposed (e.g. Archer et al., 2000; Sigman and Boyle, 2000; Sigman et al., 2010; Fischer et al., 2010; Menviel et al., 2012; Ushie and Matsumoto, 2012; Jaccard et al., 2013; Galbraith and Jaccard, 2015; Heinze et al., 2016; Wallmann et al., 2015). While the first simulations covering glacial–interglacial cycles, including dynamic ocean and land models, that represent reconstructed atmospheric CO\(_2\) variability emerge, these models are not yet able to reproduce variations in important proxy data and to represent the timing of CO\(_2\) changes over the last glacial termination adequately (Brovkin et al., 2012; Menviel et al., 2012; Ganopolski and Brovkin, 2017). To make progress in this research area requires the combination of multiple lines of evidence where proxy reconstructions are compared to quantitative model analyses. This will help to constrain underlying processes and to quantify their contribution.

One of the processes is the storage of carbon in the land biosphere but its change over the last deglaciation is debated. Most studies addressing past land biosphere carbon focus on the change in land biosphere carbon inventory between the Last Glacial Maximum around 21,000 years before present (LGM; 21 kyrBP) and the recent preindustrial period (PI). This inventory difference (PI minus LGM) is here termed \(\Delta\)land and includes changes in the various land biosphere reservoirs, such as plants, mineral soils, permafrost, peatland, yedoma, and wetlands as well as changes on shelves exposed during the LGM. Available proxy-based \(\Delta\)land estimates (e.g. Shackleton, 1977; Adams et al., 1990; Bird et al., 1994; Adams and Faure, 1998; Zimov et al., 2006, 2009; Zech et al., 2011; Ciais et al., 2012; Peterson et al., 2014; Menviel et al., 2017) encompass values ranging from -400 GtC to 1,500 GtC. This is large compared to the current land biosphere stock amounting to around 4,000 GtC (Ciais et al., 2013) as well as to the deglacial atmospheric change of around 200 GtC (1 ppm is equivalent to 2.12 GtC). The high end of the \(\Delta\)land range, implying much smaller land carbon storage during the LGM than today, is based on estimates relying on pollen records (Adams et al., 1990; Van Campo et al., 1993; Crowley, 1995; Adams and Faure, 1998). Pollen and macrofossils recorded in various archives hold information on past vegetation distribution, but do not allow constraining carbon inventories neither in soils nor in vegetation. The low end is based on reconstructions of the carbon content in paleo soils (Zimov et al., 2006, 2009; Zech et al., 2011). These reconstructions are scarce, of local nature, and restricted to cold high-altitude or high-latitude regions, rendering extrapolations to the global scale speculative. In a recent data synthesis study, Lindgren et al. (2018) suggest that the loss of carbon from northern permafrost areas from the LGM to PI was lower than initially suggested by Zimov et al. (2009) and that total land carbon storage in northern mid–and high-latitudes increased by about 400 over the deglaciation. Simulations with spatially explicit terrestrial biosphere models generally yield a positive \(\Delta\)land (see references in Joos et al., 2004; Prentice et al., 2011; O’Ishi and Abe-Ouchi, 2013; Ganopolski and Brovkin, 2017; Davies-Barnard et al., 2017). These simulations are based on models, which typically do not simulate changes in neither wetland, peatland, and permafrost.
nor carbon stocks in deep deposits of loess and yedoma, in lakes, and in subglacial sediments (Tarnocai et al., 2009; Zimov et al., 2009; Yu et al., 2010). Furthermore, the results are impaired by our limited understanding of how low atmospheric CO$_2$, generally colder and drier climate, altered nutrient fluxes, and largely exposed shelves in the tropics affected carbon inventories.

The classical (Shackleton, 1977) and arguably most reliable approach to reconstruct $\Delta$land considers the deglacial change in the marine stable carbon isotope signature as recorded in sediments and the mass balance of carbon and $\text{^{13}C}$. The argument is that the uptake of isotopically light carbon by the land biosphere causes a corresponding, measurable perturbation in the average $\delta^{13}C$ value in the ocean–atmosphere system. The $\text{^{13}C}$ mass balance approach, using marine sediment and ice core records, provided consistent and converging estimates of $\Delta$land with recent estimates ranging from 300 to 400 GtC (Curry et al., 1988; Duplessy et al., 1984; Crowley, 1991; Bird et al., 1994; Crowley, 1995; Bird et al., 1996; Joos et al., 2004; Ciais et al., 2012; Peterson et al., 2014; Menviel et al., 2017). Early estimates relied on spatially limited $\delta^{13}C$ sediment records (Duplessy et al., 1984; Curry et al., 1988; Bird et al., 1994, 1996) but recently, more comprehensive $\delta^{13}C$ data compilations (Oliver et al., 2010; Peterson et al., 2014) became available and _These_ were also used for to determine mean ocean $\delta^{13}C$ value and $\Delta$land determinations applying a dynamic ocean model (Menviel et al., 2017). The details of the approach are outlined in the literature (e.g. Ciais et al., 2012).

All previous $\text{^{13}C}$ mass balance studies assumed a closed system. They thus neglected the carbon and isotopic exchange between the ocean and reactive marine sediments, the input flux from rock weathering, and carbon burial in consolidated sediments by focussing on the atmosphere–ocean–land system only. It has been argued (e.g. Ciais et al., 2012) that changes in weathering contributed little to the deglacial CO$_2$ rise and that large changes in the calcium carbonate (CaCO$_3$) burial in sediments (and coral reefs) are inconsistent with the reconstructed changes in the depth of the lysocline.

There are, however, several important reasons to question these arguments as well as the closed system assumption. The potential role of changes in CaCO$_3$ dissolution and burial for regulating past CO$_2$ has been widely discussed (e.g. Broecker and Peng, 1987; Archer and Maier-Reimer, 1994; Archer et al., 2000; Cartapanis et al., 2018). Concerning the role of particulate organic carbon (POC), Tschumi et al. (2011) and Roth et al. (2014) identified a “nutrient–burial feedback” and demonstrated its importance also for $\delta^{13}C$ for a range of mechanisms with the potential to explain low glacial CO$_2$. They conclude that the long-term balance between burial of organic material and tracer input into the ocean through weathering—typically neglected in earlier studies, should be considered when investigating the glacial–interglacial evolution of atmospheric CO$_2$, $\delta^{13}C$, and related tracers.

The first specific reason to challenge the closed system assumption is the continuous weathering–burial cycle. The first, a continuous flux of biogenic particles made out of calcium carbonate calcareous biogenic particles and organic matter carries any perturbation in $\delta^{13}C$ in the coupled atmosphere–surface ocean system to ocean sediments (Tschumi et al., 2011; Menviel et al., 2012; Roth et al., 2014; Cartapanis et al., 2016). A second reason is related to the chemical buffering of land induced CO$_2$ perturbations by to marine CaCO$_3$ sediments, a process known as carbonate compensation (e.g. Archer et al., 2000; Broecker and Clark, 2001). Carbonate compensation causes a net transfer of carbon and $\text{^{13}C}$ between the ocean and reactive ocean–marine sediments. Third, and of even greater importance with regard to $\delta^{13}C$, are changes in the cycling and weathering–sedimentation imbalances of POC–organic carbon that have the potential to exert a large impact on the $\text{^{13}C}$ budget as shown in
earlier studies (Tschumi et al., 2011; Roth et al., 2014). These mechanisms should not be neglected, and a range of processes related to the deglacial reorganization of the marine carbon cycle likely influenced their signal in the available proxy data (Schmitt et al., 2012; Yu et al., 2013; Peterson et al., 2014; Kerr et al., 2017; Qin et al., 2017; Luo et al., 2018), when addressing the whole ocean budgets of carbon and $\delta^{13}C$ to infer changes in land carbon stocks.

The documented transfer of $^{13}$C and carbon between the ocean-atmosphere system and reactive sediments and the lithosphere is affected by the well-documented reorganization of the marine carbon cycle across the deglaciation (e.g. Sigman and Boyle, 2000; Sigman et al., 2010; Fischer et al., 2010; Elderfield et al., 2012; Ciais et al., 2013; Martinez-Garcia et al., 2014; Jaccard et al., 2016; Cartapanis et al., 2016) affected the transfer of $^{13}$C and carbon between the ocean–atmosphere system and reactive sediments and the lithosphere. The reorganization includes changes in CO$_2$ solubility, ocean ventilation (e.g. Burke and Robinson, 2012; Sarnthein et al., 2013; Skinner et al., 2017) and water mass distribution (e.g. Duplessy et al., 1988; Peterson et al., 2014; Lippold et al., 2016; Menviel et al., 2017; Gottschalk et al., 2018), air–sea gas transfer rates, for example via changes in sea-ice extent (e.g. Stephens and Keeling, 2000; Gersonde et al., 2005; Waelbroeck et al., 2009; Sun and Matsumoto, 2010), export (e.g. Kohfeld et al., 2005; Jaccard et al., 2013) and remineralization (e.g. Bendtsen et al., 2002; Matsumoto, 2007; Taucher et al., 2014) of biogenic material associated with the cycling of organic carbon, CaCO$_3$, and biogenic opal (Tschumi et al., 2011; Roth et al., 2014; Matsumoto et al., 2014), changes in coral reef growth (e.g. Berger, 1982; Milliman and Droxl er, 1996; Kley pas, 1997; Ridgwell et al., 2003; Vecsei and Berger, 2004; Menviel and Joos, 2012), and in the input of dust, nutrients and lithogenic material by atmospheric deposition (e.g. Lambert et al., 2015) and rivers, and coastal and shelf exchange processes (Ushie and Matsumoto, 2012; Wallmann et al., 2015). These changes influence the particle flux of organic and inorganic carbon, opal, and mineral particles adding carbon and mass to the sediments. They further influence the partitioning between sediment dissolution and consolidation (burial) by altering the concentration of dissolved inorganic carbon (DIC) and as a consequence, these deglacial changes and their spatio-temporal evolution affect the mean $\delta^{13}$C of DIC and POC, alkalinity, carbonate ion, oxygen, and other tracers in near-sediment bottom water and in sediment pore water, signature of the global ocean and estimates of $\Delta$land. For this reason, it is essential to explicitly consider the causes of the glacial–interglacial variations in CO$_2$ to quantify $\Delta$land.

The primary goal of this study is to provide a new, observationally constrained best estimate of $\Delta$land recognizing the role of marine deglacial processes, weathering and burial, and sediment interactions in an Earth system model. A second goal is to establish how this response sensitivities for a range of proxy targets to mean ocean $\delta^{13}$C$_{DIC}$ and for other proxy targets (PI-LGM differences) to the changes in individual key deglacial carbon cycle mechanisms from transient simulations. An emulator of the Bern3D has been developed and the strengths of individual marine and terrestrial processes are varied in a large number of combinations to find all possible solutions for $\Delta$land that match a set of observational targets in a Bayesian Monte Carlo data assimilation framework (see Fig. 1). We further scrutinize the closed system assumption in the $^{13}$C mass balance approach. This is tackled by probing the role of the weathering–burial cycle and sediment compensation. We with land carbon uptake as the only forcing. Specifically, we run idealized pulse-like carbon uptake simulations with the Bern3D Earth System Model of Intermediate Complexity (EMIC) both with and without simulating sediment interactions, and with and without interactive weathering. The further discuss multi-proxy response relationships for different carbon cycle
processes and briefly address the possible contribution of individual mechanisms to the deglacial change in atmospheric CO₂ and δ¹³C atm, and of deep ocean carbonate ion concentration. Our results demonstrate the importance of these mechanisms and suggest that previous δ¹³C-based estimates of Δland have been systematically underestimated. Sensitivities of individual marine and terrestrial processes contributing to deglacial change are determined and varied in a large number of combinations to find all possible solutions that match a set of observational targets in a Bayesian Monte Carlo data assimilation framework (see Fig. 1). Neglected the influence of these processes and appear systematically biased towards lower estimates.

2 Methods

2.1 The Bern3D model

The Bern3D EMIC is briefly described in the Appendix A, where also couples a dynamic geostrophic-frictional balance ocean, a thermodynamic sea ice component, and a single layer energy-moisture balance atmosphere. The horizontal resolution is 41x40 grid cells and the ocean has 32 layers. Marine productivity is simulated as a function of nutrient concentrations (P, Fe, Si), temperature and light and transferred to dissolved organic and particulate matter. Biogenic matter decomposes within the water column. Opal, calcite, and organic particles reaching the ocean floor are entering reactive sediments. A 10-layer sediment model is used to compute fluxes of carbon, nutrients, alkalinity and isotopes between the ocean, reactive sediments, and the lithosphere. Loss fluxes to the lithosphere are compensated at equilibrium by a corresponding input flux from weathering. A 4-box reservoir model is used to calculate the dilution of atmospheric isotopic perturbations by exchange with the land biosphere.

Further information on the model and the model spin-up and experimental settings for the impulse response experiments are detailed given in the Appendix A. The sediment module has been updated compared to previous work to include most recent observational information on sediment composition and fluxes (Cartapanis et al., 2016, 2018; Tréguer and De La Rocha, 2013) as detailed in Appendix B. Key model parameters were adjusted within their uncertainties to best reproduce the observation-based spatial distributions and global stocks of CaCO₃, POC, and opal within reactive sediments, observational estimates of global burial and redissolution fluxes, and of euphotic-zone export fluxes of biogenic particles (Table 1). This updated version of the Bern3D model is introduced as Bern3D v2.0s. The simulated oceanic DIC inventory for PI is 37,175 GtC, similar to the 37,310 GtC estimated based on the GLODAP v.2 data set (Lauvset et al., 2016).

¹³C stocks and exchanges are modeled in the atmosphere–ocean–sediment–landbiosphere system. ¹³C fluxes to the lithosphere associated with the burial of POC and CaCO₃ and weathering input to the ocean are explicitly simulated. The burial of POC and CaCO₃ results in an isotopic signature of the burial flux of around -9‰, intermediate between the isotopically light POC (~-20‰) and the isotopically heavy CaCO₃ signature (~2.9‰; see Appendix A for implementation of isotopic discrimination). An isotopic perturbation in the atmosphere, e.g., through uptake of isotopically light carbon by the land biosphere, is transferred to the ocean through air–sea gas exchange and to the land by photosynthesis. Within the ocean,
Standard model forcing for period 40 kyrBP to pre-industrial:
- orbital parameters
- coral reef regrowth
- greenhouse gas radiative forcing
- North Atlantic freshwater input
- land ice albedo

& changes in seven mechanisms (parameters $p_i$):
- Southern Ocean wind stress
- export rain ratio
- coral reef regrowth
- Southern Ocean air-sea gas transfer rate
- organic matter remineralization
- organic weathering flux
- land carbon stock

Sensitivity to each mechanism $i$ for each target $T$:
$$S_{Ti}^T = \frac{\partial T}{\partial p_i}$$

Emulator of Bern3D results for target $T$:
$$\Delta T = a^T + b^{T} \cdot (\sum_{i=1}^{7} \Delta p_i \cdot S_{Ti}^T)$$

50 Bern3D factorial runs
60 Bern3D multi-parameter runs
500,000 multi-parameter runs with emulator and uniform prior distribution

Constrain with proxy targets $T$
$\delta^{13}C_{DIC}$ $\delta^{13}C_{atm}$ $CO_2$ $CO_3$

Observation-constrained change in land carbon ($\Delta$land)

Figure 1. Flow chart outlining the steps applied to achieve an estimate of possible $\Delta$land values. To this end, seven generic deglacial carbon cycle mechanisms (Table 2 and Section 2.2) and four observational targets (Section 2.2.3) were included. The targets are the PI-LGM change in atmospheric $\delta^{13}C$ ($\Delta \delta^{13}C_{atm}$) recorded in Antarctic ice (Schmitt et al., 2012); the PI-LGM change in the global ocean average $\delta^{13}C$ of dissolved inorganic carbon ($\Delta \delta^{13}C_{DIC}$) as reconstructed from 480 benthic foraminiferal records (Peterson et al., 2014); the PI-LGM change in carbonate ion concentration in the deep Pacific inferred from boron (B) to calcium (Ca) ratios in foraminifera (Yu et al., 2013); and the PI-LGM change in atmospheric $CO_2$ (Petit et al., 1999; Monnin et al., 2001; Siegenthaler et al., 2005; Lüthi et al., 2008). The seven processes are varied individually by systematic parameter variations in addition to the well-established forcings (top of figure) in 50 factorial simulations with the Bern3D. This yields a first set of sensitivities or forcing–response curves for our four targets and seven processes. Next, we apply a Latin Hypercube parameter sampling to vary the processes in combination and probe for non-linear interactions in 60 multi-parameter simulations with the Bern3D model. The results are used to adjust the forcing response relationship; adjustments are small, except for $CO_2$ response curves (see Fig. C1 in the Appendix). Analytical representations of the forcing–response curves are used to build a simple emulator of the Bern3D model (Section 2.2.4). The emulator is applied in a Monte Carlo ensemble with half a million members to probe a wide range for each individual process. Finally, the ensemble results are constrained by the four proxy targets to yield a best estimate and an uncertainty distribution for $\Delta$land.
Figure 2. Observed (A) versus simulated (B) pre-industrial $\delta^{13}$C$_{DIC}$ distributions along a cross section through the Atlantic ($25^\circ$W), across the Southern Ocean ($58^\circ$S) and into the Pacific ($175^\circ$W). (A) shows data from Eide et al. (2017) based on ocean measurements and corrected for the Suess-effect. (B) shows $\delta^{13}$C as modeled in the Bern3D model under pre-industrial boundary conditions.

any perturbation signal is communicated between the surface ocean and the deep ocean by advection, convection, and mixing. In addition, POC and CaCO$_3$ export fluxes from the surface ocean communicate to ocean sediments and therefore contribute to the burial flux.

Figure 2 shows modeled $\delta^{13}$C in a section through the Atlantic, Southern Ocean, and Pacific under pre-industrial (1765 CE) boundary conditions with a prescribed atmospheric $\delta^{13}$C signature of -6.305 ‰. Compared to measured data corrected for the Suess-effect (Eide et al., 2017) it is visible that modeled $\delta^{13}$C values are biased high towards higher values by a seemingly constant offset of about 0.4 ‰, while the $\delta^{13}$C patterns are faithfully captured by the model.

2.2 Transient LGM to PI sensitivity simulations

2.2.1 Standard transient forcings

Starting from PI steady state conditions, the model is first run for 20 kyr under constant LGM forcing and then for an additional 20 kyr under transient forcing from the LGM to PI. Forcings (Fig. 3) include radiative forcing imposed by CO$_2$, N$_2$O, and CH$_4$ (Joos and Spahni, 2008) and variations in orbital parameters (Berger, 1978). Radiative forcing of CO$_2$ is prescribed and biogeochemistry is calculated determined with interactive CO$_2$. Ice sheet extent and related changes in albedo are prescribed based on benthic $\delta^{18}$O data (Lisiecki and Raymo, 2005) and ice sheet reconstructions (Peltier, 1994). Additionally, freshwater (FW) pulses are prescribed in the North Atlantic as detailed in Menviel et al. (2012), also accounting for the LGM to PI change in salinity of about 1 PSU. Coral reef regrowth is implemented from 14 kyrBP onward (Vecsei and Berger, 2004) by uniformly removing the respective amount of alkalinity, carbon, and $^{13}$C from the uppermost ocean grid cells. These forcings (Fig. 3A-D) are termed standard (std.) forcings.
Figure 3. Forcings applied for the LGM to PI simulations. (A) Radiative forcing imposed by CO$_2$, CH$_4$, and N$_2$O, (B) benthic $\delta^{18}$O, used to scale ice sheet extent, (C) freshwater forcing into the North Atlantic, and (D) coral reef regrowth as applied as standard transient forcing. (E) Prescribed evolution of land biosphere carbon uptake. Cumulative land biosphere uptake is varied over the termination across factorial simulations, but kept invariant (254 GtC) over the Holocene (11–0 kyrBP) following Elsig et al. (2009); here a scenario with a total uptake of 445 GtC is shown for illustration. And (F) idealized evolution of scaling factors as prescribed in factorial experiments to vary the remineralization profile, rain ratio, Southern Ocean wind stress and gas transfer rate, or organic weathering flux on land between LGM and PI values. The freshwater forcing in the North Atlantic is varied over 40 kyr. Dashed green lines indicate spin-up values.
Table 1. Export, burial, and deposition fluxes as well as sediment inventories of CaCO$_3$, opal, and particulate organic carbon (POC) as determined in the model after a pre-industrial spin-up and literature estimates. The tracer input flux to the ocean resulting from weathering is set to compensate for the burial fluxes diagnosed at the end of the spin up and kept constant in Bern3D standard simulations.


<table>
<thead>
<tr>
<th>variable</th>
<th>units</th>
<th>Bern3D</th>
<th>observational estimates</th>
</tr>
</thead>
<tbody>
<tr>
<td>CaCO$_3$ export</td>
<td>GtC yr$^{-1}$</td>
<td>0.98</td>
<td>0.72-1.05$^a$</td>
</tr>
<tr>
<td>opal export</td>
<td>Tmol Si yr$^{-1}$</td>
<td>109.69</td>
<td>88-122$^b$</td>
</tr>
<tr>
<td>POC export</td>
<td>GtC yr$^{-1}$</td>
<td>11.98</td>
<td>6.5-13.1$^c$</td>
</tr>
<tr>
<td>CaCO$_3$ deposition</td>
<td>GtC yr$^{-1}$</td>
<td>0.49</td>
<td>0.5$^d$</td>
</tr>
<tr>
<td>opal deposition</td>
<td>Tmol Si yr$^{-1}$</td>
<td>77.39</td>
<td>78.8$^b$</td>
</tr>
<tr>
<td>POC deposition</td>
<td>GtC yr$^{-1}$</td>
<td>0.66</td>
<td>1.7-3.3$^c$</td>
</tr>
<tr>
<td>CaCO$_3$ burial</td>
<td>GtC yr$^{-1}$</td>
<td>0.23</td>
<td>0.1-0.14$^c$</td>
</tr>
<tr>
<td>opal burial</td>
<td>Tmol Si yr$^{-1}$</td>
<td>6.64</td>
<td>2.7-9.9$^b$</td>
</tr>
<tr>
<td>POC burial</td>
<td>GtC yr$^{-1}$</td>
<td>0.24</td>
<td>0.12-0.26$^c$</td>
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<tr>
<td>CaCO$_3$ stock</td>
<td>GtC</td>
<td>948</td>
<td></td>
</tr>
<tr>
<td>opal stock</td>
<td>Tmol Si</td>
<td>20,484</td>
<td></td>
</tr>
<tr>
<td>POC stock</td>
<td>GtC</td>
<td>518</td>
<td></td>
</tr>
</tbody>
</table>

2.2.2 Factorial sensitivities

Standard transient deglacial forcings in the Bern3D model are complemented by changes in seven additional mechanisms, grouped into generic classes and introduced below (see top of Fig. 1). A set of 50 factorial experiments (Table 2), covering the same time-interval as the model run with standard transient forcings, is conducted to quantify the sensitivities of the carbon cycle to changes in these seven mechanisms. The parameters forcing history for changes in Southern Ocean wind stress, Southern Ocean gas transfer rate, the rain ratio, the remineralization profile, and the organic weathering rate are prescribed following an idealized glacial-interglacial evolution. PI values are set to their glacial values instantaneously at the beginning of the glacial simulation and scaled back towards their modern values over the deglacial as illustrated in LGM values at 40 kyrBP, kept constant from 40 kyrBP to 18 kyrBP, scaled back to the PI value over the termination (18 kyrBP to 11 kyrBP) and kept constant thereafter (Fig. 3D,E, and F, F and Table 2). The sensitivities of carbon cycle properties are obtained by subtracting the standard forcing run from the runs with complementary changes in the seven mechanisms. This also yields characteristic response relationships between different carbon cycle properties, e.g., changes in atmospheric CO$_2$ versus changes in whole ocean $\delta^{13}$C, for each process. The objective is to schematically represent the space of plausible property–property relationships for key proxies. In other words, the selected mechanisms are assumed to be representative on the global scale for the myriad of processes influencing the carbon cycle and will be motivated below.
**Physical Mechanisms:** Changes in ocean circulation, for example in response to altered wind stress, stratification, eddy mixing, or buoyancy forcing, affect the carbon inventory of the ocean (Adkins, 2013) by altering the cycling of organic and inorganic carbon. The Southern Ocean has been identified as an important region modulating CO$_2$ changes and a reduced Antarctic overturning circulation has been invoked to, at least partly, explain low glacial atmospheric CO$_2$ (Sigman and Boyle, 2000; Sigman et al., 2010; Fischer et al., 2010; Skinner et al., 2010; Tschumi et al., 2011; Ferrari et al., 2014; Watson et al., 2015; Roberts et al., 2016; Gottschalk et al., 2016; Jaccard et al., 2016). Under standard forcing, Southern Ocean circulation changes are weak in the Bern3D model. In this study, the circulation is modified by uniformly scaling the wind stress field over the Southern Ocean (>48°S, south of South America) (see also Tschumi et al., 2008) to invoke a reduced reduction deep ocean ventilation. The scaling factor is varied linearly over the termination between LGM values (given in Table 2) and the PI value of unity (Fig. 3F). We do not consider such large changes in wind stress as realistic, but rather use prescribed Southern Ocean wind stress as a way to vary deep ocean ventilation in the Bern3D model. Consequently, we do not scale air-sea gas transfer rates with the changes in wind stress, but vary transfer rates independently.

Proxy reconstructions suggest a larger sea-ice extent in the Southern Ocean during glacial times (Gersonde et al., 2005; Waelbroeck et al., 2009). Sea-ice hinders air–sea gas exchange and thus exerts a large impact on atmospheric δ$^{13}$CO$_2$ (Lynch-Stieglitz et al., 1995). Additionally, changes in Southern Ocean winds (Kohfeld et al., 2013) may have affected glacial Southern Ocean air–sea gas exchange rates. Here, the standard air–sea gas transfer rate is scaled (Fig. 3F; Table 2) in the Southern Ocean to estimate related carbon cycle sensitivities. We note that changes in sea-ice and a corresponding change in air–sea gas transfer rates are reasonably well simulated by the Bern3D model. The effect of changes in ocean temperature (Waelbroeck et al., 2009) and salinity on the solubility of CO$_2$ is discussed elsewhere (Sigman and Boyle, 2000; Brovkin et al., 2007; Sigman et al., 2010; Menviel et al., 2012) and is part of the standard forcing.

**Oceanic carbonate:** The partial pressure of CO$_2$ in the ocean depends on alkalinity and processes that alter surface ocean alkalinity thus influencing atmospheric CO$_2$. The sedimentary burial of CaCO$_3$ removes twice as much alkalinity as carbon from the ocean leading to oceanic CO$_2$ outgassing. Increased CaCO$_3$ burial over the deglacial has been put forward as the coral reef hypothesis (Berger, 1982) to explain increases in atmospheric CO$_2$. Vecsei and Berger (2004) reconstructed coral reef growth history and provided a lower limit estimate of about 380 GtC for the amount of shallow water carbonate deposition over the deglacial period. Other estimates of CaCO$_3$ deposition amount to 1,200 GtC or even more (Milliman, 1993; Kleypas, 1997; Ridgwell et al., 2003). This yields a large range of possible scenarios with substantial impacts on atmospheric CO$_2$. Here, we scale the reconstructed deposition history based on Vecsei and Berger (2004) (Fig. 3D) by applying a constant scaling to vary CaCO$_3$ deposition within the published range.

Potential changes in the rain ratio (CaCO$_3$:$C_{org}$) affect both alkalinity and CO$_2$ (e.g. Berger and Keir, 2013; Archer and Maier-Reimer, 1994; Sigman et al., 1998; Archer et al., 2000; Matsumoto et al., 2002; Tschumi et al., 2011). An increase in the export of CaCO$_3$ relative to $C_{org}$ from surface waters has similar consequences for CO$_2$ as shallow water carbonate deposition (Sigman and Boyle, 2000). There is a lack of estimates how the rain ratio has varied in the past. Here, we vary the global rain ratio of 0.083 for LGM conditions (see also Appendix A; Fig –3F; Table 2) between 0.045 and 0.098, while the...
rain ratio is 0.083 in the standard setup and for PI conditions.

**Oceanic organic matter:** There is a broad range of mechanisms, in addition to circulation changes, that affect the cycling of organic matter within the ocean and the whole ocean nutrient inventories, and thereby surface ocean nutrient and DIC concentrations as well as atmospheric CO$_2$. Several studies have suggested changes in the remineralization length scale of organic carbon due to colder ocean temperatures (see e.g. Matsumoto, 2007; Menviel et al., 2012; Roth et al., 2014). A deeper remineralization of organic matter leads to the removal of nutrient and DIC from the shallow subsurface ocean and, via ocean–sediment interactions, to globally reduced nutrient and increased alkalinity inventories (Menviel et al., 2012; Roth et al., 2014), resulting in a decrease in atmospheric CO$_2$ (Matsumoto, 2007; Menviel et al., 2012) amplified through ocean–sediment interactions (Roth et al., 2014). Weathering-burial feedbacks also lead to an amplification of changes in atmospheric and oceanic $\delta^{13}$C as shown by Tschumi et al. (2011) and Roth et al. (2014). Here, the remineralization profile in the upper 2 km is varied between the standard Martin curve (Eq. A1) and a linear profile (75 m to 2000 m). The modification in the remineralization profile of particulate organic matter is different than those applied by Menviel et al. (2012) and Roth et al. (2014) in simulations with the Bern3D model. Here, only the remineralization profile in the upper 2000 m is changed and, in contrast to these earlier studies, it is assumed that the fraction of exported particles that reach the sediments below 2000 m is not altered. The implicit assumption is that cooler temperatures at the LGM only significantly affect the dissolution of relatively small or labile particles in the upper ocean, while large or refractory particles sink fast enough to reach ocean sediments both under LGM and PI conditions.

Oceanic organic carbon and nutrient inventories are further affected by changes in the weathering flux compensating for burial of organic matter (Cartapanis et al., 2016, 2018). A negative balance between the riverine input of PO$_4$, DIC, DI$^{13}$C, and Alk, originally buried as organic material, and sedimentary burial of particulate organic matter leads to a transient decrease in atmospheric CO$_2$ (Cartapanis et al., 2016) and also affects atmospheric and marine $\delta^{13}$C. Here the weathering flux to the ocean (Table 1) is kept constant in the standard setup of the Bern3D (see also Appendix A), but might have varied with changes in climate and CO$_2$. To account for this possibility, we varied the input of PO$_4$, DIC, DI$^{13}$C, and Alk that compensates for the burial of particulate organic material in the sediment in factorial experiments (Fig. 3F; Table 2). This variation in the input will be referred to as changes in the oceanic weathering flux.

Another widely discussed mechanism relates to iron fertilization in the Southern Ocean (Martin, 1990; Archer et al., 2000; Sigman and Boyle, 2000; Parekh et al., 2008; Sigman et al., 2010; Fischer et al., 2010; Martinez-Garcia et al., 2014). The effect of iron fertilization on oceanic carbon and $\delta^{13}$C further leads to changes in the cycling of marine organic material and to related changes in the whole ocean inventory of $\delta^{13}$C$_{DIC}$ and other variables. These changes are broadly similar to the mechanisms outlined above and is thus not explicitly considered further. Other changes caused by altering the organic matter remineralization depth or the weathering input flux and implicitly included in the Monte Carlo simulations. Similarly, other processes such as changes in the ratio of nutrient to carbon uptake by marine organisms (Heinze et al., 2016) or changes in the association of POC with ballast minerals (Armstrong et al., 2002) may have affected the efficiency of
Table 2. Overview of mechanisms and parameters considered in factorial sensitivity experiments. Bold entries mark the standard parameter values. Values represent the export rain ratio at LGM, the change in remineralization profile (see text) at LGM, the scaling factors applied to the standard shallow water carbonate deposition history, and the land carbon uptake over the deglacial in GtC. In the case of a standard parameter equal SO wind stress, SO gas transfer rate, and the organic weathering flux, values represent the ratio of LGM to unity PI forcing.

<table>
<thead>
<tr>
<th>Mechanisms</th>
<th>Parameter values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Southern Ocean wind stress</td>
<td>0.4, 0.5, 0.6, 0.7, 0.8, 0.9, <strong>1.0</strong></td>
</tr>
<tr>
<td>Southern Ocean gas exchange rate</td>
<td>0.4, 0.5, 0.6, 0.7, 0.8, 0.9, <strong>1.0, 1.1, 1.2</strong></td>
</tr>
<tr>
<td>Export rain ratio</td>
<td>0.045, 0.05, 0.055, 0.063, 0.068,</td>
</tr>
<tr>
<td></td>
<td><strong>0.073, 0.083, 0.088, 0.093, 0.098</strong></td>
</tr>
<tr>
<td>Shallow water carbonate deposition</td>
<td>0, 0.5, <strong>1</strong>, 1.5, 2, 3, 4</td>
</tr>
<tr>
<td>% change towards linear remineralization profile</td>
<td><strong>0, 10, 20, 40, 60, 80, 100</strong></td>
</tr>
<tr>
<td>Organic weathering flux</td>
<td>0.5, 0.6, 0.7, 0.8, 0.9, <strong>1.0, 1.2, 1.4, 1.6, 2.0</strong></td>
</tr>
<tr>
<td>Land biosphere uptake (GtC)</td>
<td>-500, -200, <strong>0, 445, 890, 1335</strong></td>
</tr>
</tbody>
</table>

the marine biological cycle in reducing carbon in surface water ocean and the atmosphere.

**Land biosphere carbon inventory:** Changes in land biosphere carbon stock are prescribed following the evolution illustrated in Fig. 3E. We assume that the change in land carbon stock over the Holocene is well constrained and amounts to an uptake of 290 GtC between 11–5 kyrBP and a release of 36 GtC thereafter (Elsig et al., 2009). Scenarios with different cumulative land carbon change imply a corresponding uptake or release of terrestrial carbon during the last glacial termination (18–11 kyrBP). The δ^{13}C signature of terrestrial carbon is set to -24 ‰. Additional sensitivity tests varying the δ^{13}C signature between -23 ‰ and -25 ‰ are conducted and will be discussed in Section 4.

2.2.3 Data constraints for deglacial carbon and carbon-isotope changes

We consider four globally relevant observational constraints (Table 3) to estimate the PI-LGM change in the land biosphere carbon inventory, Δland. These are the PI-LGM difference (Δ) in (i) atmospheric CO₂ (ΔCO₂), (ii) atmospheric δ^{13}C(CO₂) (Δδ^{13}C_{atm}), (iii) mean ocean δ^{13}C(DIC) (Δδ^{13}C_{DIC}), and (iv) deep (equatorial) Pacific carbonate ion concentration (ΔCO₃²⁻) (Table 3). The carbon isotope constraints are directly relevant for the isotopic mass balance used to infer Δland. The carbonate ion concentration difference constrains changes in the ocean’s alkalinity budget, and the change in atmospheric CO₂ constrains the integrated effect of deglacial carbon cycle changes. We allow for a relatively wide uncertainty range of 20 ppm for ΔCO₂, which is admittedly wider than the proxy uncertainty, to avoid an overfitting of model outputs. The Δδ^{13}C_{atm} range for the constraint is 0.1 ±0.05 ‰ and based on ice core measurements (Schmitt et al., 2012). Peterson et al. (2014) compiled 480 benthic foraminiferal records and report a whole-ocean PI minus LGM change in δ^{13}C_{DIC} of 0.34±0.19 ‰ and we use their range (0.15 to 0.53 ‰) as constraint. Proxy reconstructions of ΔCO₃²⁻ based on measured B/Ca ratios in foraminifera show
Table 3. Overview of the four proxy-based constraints representing change ($\Delta$) between PI minus LGM. LGM refers to the period 20 to 19 kyrBP for the ice core and to the period 23 to 19 kyrBP for the ocean sediment data. Similarly, PI refers to 500 to 200 yrBP for the ice core data and to 6,000 to 200 yrBP and to 8 to 6 kyrBP for marine $\delta^{13}$C and CO$_2^{-3}$, respectively. The assumed uncertainty range in $\Delta$CO$_2$ is considered to be wider than the uncertainty from the ice core data. This is to avoid overfitting and taking into account uncertainties in the emulator introduced in Section 2.2.4.

<table>
<thead>
<tr>
<th>Constraint</th>
<th>uncertainty range</th>
<th>references</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta$CO$_2$</td>
<td>80 to 100 ppm</td>
<td>see text</td>
</tr>
<tr>
<td>$\Delta\delta^{13}$C$_{atm}$</td>
<td>0.1±0.05‰</td>
<td>Schmitt et al. (2012)</td>
</tr>
<tr>
<td>$\Delta\delta^{13}$C$_{DIC}$</td>
<td>0.34±0.19‰</td>
<td>Peterson el al. (2014)</td>
</tr>
<tr>
<td>$\Delta$CO$_2^{-3}$</td>
<td>-6 to 5 µmol kg$^{-1}$</td>
<td>see text</td>
</tr>
</tbody>
</table>

little change from LGM to PI in the equatorial deep Pacific (Yu et al., 2013). Based on a different approach, Qin et al. (2017) also conclude on negligible change in $\Delta$CO$_2^{-3}$ in the western tropical Pacific across the last glacial termination. In a recent study, Luo et al. (2018) report, based on CaCO$_3$ reconstructions in the South China Sea, that carbonate ion concentrations in the mid-depth Pacific hardly changed from LGM to PI. All these studies point to a small change in $\Delta$CO$_2^{-3}$, providing a further constraint on deglacial simulations. However, the uncertainty in these reconstructions is of the same order as the change itself. The target range for $\Delta$CO$_2^{-3}$ is chosen here from -6 to 5 µmol kg$^{-1}$.

2.2.4 Emulator and Monte Carlo setup

Sensitivities of the four target proxies ($\Delta$CO$_2$, $\Delta\delta^{13}$C$_{atm}$, $\Delta\delta^{13}$C$_{DIC}$, and $\Delta$CO$_2^{-3}$) to changes in the selected seven forcings are computed based on 50 factorial experiments. For each forcing, $i$, and related parameter change, $\Delta p_i$, (Table 2) the corresponding sensitivity, i.e., the change in target per unit change in parameter, is computed. These discrete sensitivities are fitted linearly or quadratically for each target, $T$, and forcing to get a continuous function for the sensitivity, $S^T_i(\Delta p_i)$. These sensitivity functions are used to build a simple representation to compute changes in target variables for a combination of changes in parameters:

$$\Delta T(\Delta p_1, ..., \Delta p_7) = \sum_{i=1}^7 \Delta p_i \times S^T_i(\Delta p_i)$$ (1)

In order to evaluate the assumed linear additivity of the seven mechanisms, a 60 member ensemble of parameter combinations is calculated both with the Bern3D model and Eq. 1 (Fig. 1). The seven parameters are sampled uniformly using Latin Hypercube Sampling (LHS; McKay et al., 1979). The results from the Bern3D model and Eq. 1 are regressed against each other for each target variable (Fig. C1). The corresponding linear fit is used to account for non-additive behavior when combining forcings. This then yields the following emulator:

$$\Delta T(\Delta p_1, ..., \Delta p_7) = a^T + b^T \times \left( \sum_{i=1}^7 \Delta p_i \times S^T_i(\Delta p_i) \right)$$, (2)
Figure 4. Temporal evolution of the perturbation in (A) atmospheric CO$_2$ and $\delta^{13}$C$_{atm}$ (inverted y-axis), and (B) the calculated perturbation in terrestrial carbon storage following Eq. A2 and A3 and $\delta^{13}$C$_{DIC}$ anomalies for a pulse uptake of 100 GtC by the land in a closed (green), open (orange), and open system with enabled weathering feedback (purple). Solid lines refer to left and dashed lines to right y-axis in both panels.

where $\alpha$, $\alpha^T$ is the offset and $b$, $b^T$ the slope of the respective linear fit as given in Fig. C1. The root mean square deviation between the emulator (Eq. 2) and the Bern3D model is 8.8 ppm for $\Delta$CO$_2$, 0.06 ‰ for $\Delta \delta^{13}$C$_{atm}$, 0.02 ‰ for $\Delta \delta^{13}$C$_{DIC}$, and 4 µmol kg$^{-1}$ for $\Delta$CO$_3$$^2$- . The seven parameters of the LGM to PI sensitivity experiments are sampled uniformly, by drawing random samples over their employed ranges (Table 2) to generate a 500,000 member Monte Carlo ensemble. The range for $\Delta$land was extended from 1,335 to 1,500 in order to cover the range of estimates. This is justified by the very linear relationship of the four targets to changes in $\Delta$land (e.g. Fig. 7). The above emulator is used for each of these members to estimate the corresponding changes in the four targets. Members which yield changes in the targets that lie outside the defined ranges (Table 3) are excluded from the analysis; all remaining members are used to calculate median and confidence ranges in $\Delta$land and other variables of interest.
3 Results

3.1 Pulse experiments

We start by investigating how atmospheric CO₂ and δ¹³C signatures evolve in response to a negative pulse emission carbon uptake by the land biosphere when all other forcings remain unchanged. To this end, 100 GtC of light carbon (δ¹³C=-24 ‰) are instantaneously (in the first time step of the year) removed from the model atmosphere to test the Bern3D response for different model setups (see Appendix A). This yields the Green’s functions (or impulse response functions) for carbon and ¹³C shown in Fig. 4.

First, we apply the pulse to a closed system, which is including only the atmosphere, ocean, and land biosphere components. The removal of 100 Gt of light carbon from the atmosphere results in an initial decline in atmospheric CO₂ followed by a typical impulse–response recovery (e.g. Maier-Reimer and Hasselmann, 1987; Siegenthaler and Joos, 1992). Atmospheric CO₂ recovers as the ocean starts to outgass CO₂ (Fig. 4). In a closed system, atmospheric CO₂ reaches a new equilibrium after about 2 kyr, at 8 ppm below the initial steady state. As light carbon is removed, the remaining carbon in the ocean and atmosphere becomes relatively enriched in ¹³C. Initially, the δ¹³C atm perturbation is removed much faster than the atmospheric CO₂ perturbation as exchange with the ocean and land biosphere dilutes the imposed isotopic perturbation (Fig. 4A) (Fig. 4A: Joos and Bruno, 1996). The oceanic δ¹³C increases and equilibrates after about 2 kyr. The difference in the response of CO₂ and δ¹³C is related to their different properties. CO₂ represents a concentration, whereas δ¹³C stands for the ¹³C to ¹²C isotopic ratio. The capacity of the ocean to remove an atmospheric perturbation in CO₂, and equally in ¹²CO₂ and ¹³CO₂, is limited by the acid-base carbonate chemistry described by Revelle and Suess (1957). In contrast, the removal of a perturbation in the isotopic ratio ¹³CO₂/¹²CO₂ is hardly affected by this chemical buffering; the buffering affects ¹³CO₂ in the nominator and ¹²CO₂ in the denominator about equally leading to a near cancellation of its effect on the ratio. The resulting change for the new equilibrium in δ¹³C amounts to 0.066 ‰ for the atmosphere, 0.060 ‰ for the ocean, and 0.065 ‰ for the 4-box land biosphere. The mass balance between ocean, land and atmosphere is maintained such that the inferred land perturbation (calculated using Eq. A2 and A3) corresponds to the prescribed removal of 100 GtC (Fig. 4B).

In the second, open system experiment, marine sediments are included. The evolution of atmospheric CO₂ (see Archer et al., 1998) is similar to the closed system except that CaCO₃ compensation leads to a further recovery of atmospheric CO₂ until equilibrium is reached after ~600 years. CO₂ equilibrates at around 3.5 ppm below the initial steady state. For the first kyr, the evolution of δ¹³C in the atmosphere and ocean is comparable to the closed system experiment but differs thereafter. In both the atmosphere and ocean, the novel and most important finding from the pulse experiment is that the δ¹³C perturbation is decreasing compared to the open system experiment on a multi-millenial timescale (see dashed lines in Fig. 4B).

In the ocean–atmosphere–land biosphere system is increasingly and eventually completely removed by open system processes. Fig. 4B shows the δ¹³C_DIC perturbation to decrease on a multi-millenial timescale in contrast to the closed system assumption (dashed lines), related to imbalances in the sedimentation (deposition minus redissolution/oxidation at the ocean–sediment interface) and weathering fluxes of POC and CaCO₃. In other words, benthic foraminifera record a much smaller perturbation in seawater δ¹³C than expected from
closed system modelling. The resulting difference in the $\delta^{13}C_{DIC}$ perturbation between the open and closed system causes an erroneous mass balance inference for the terrestrial biosphere, when applying conventional closed system equations (Eq.

A2 and A3). On a timescale of 10 kyr, underestimation of the terrestrial carbon inventory change amounts to 30 %. The error increases as times evolves.

In detail, the difference between the open and closed system pulse responses is explained as follows. The $^{13}$C budget in the model may change as a result of open system changes due to imbalances between input and burial fluxes of POC and CaCO$_3$ , or from and due to changes in the $\delta^{13}$C signatures of the respective fluxes. Carbon–climate feedbacks over the course of The uptake of isotopically light land carbon from the atmosphere leads to an increase in the experiment cause changes in temperature and ocean circulation, which affect the export of $\delta^{13}$C signature of surface waters and by that in the $\delta^{13}$C signatures of exported and eventually buried POC and CaCO$_3$. The removal of light terrestrial carbon increases the mean $\delta^{13}$C signature of surface waters and by that the burial flux amounts to -8.7 ‰ ($\delta^{13}C_{CaCO_3}$: 2.9 ‰) and $C_{org}$ ($\delta^{13}C_{POC}$: -20.2 ‰) for the first 10 kyr compared to -9.1 ‰ for the weathering input. This difference tends to mitigate the $\delta^{13}$C signatures of exported $DIC$ perturbation. Further, carbon–climate feedbacks cause changes in temperature and ocean circulation, which affect the export of POC and CaCO$_3$. Burial fluxes of POC further depend on O$_2$ concentrations and burial fluxes of CaCO$_3$ on CO$_2$ concentrations which evolve over the course of the simulation. As a result, the cumulative sedimentation-weathering imbalance for the open system setting for the first 10 after the pulse uptake sedimentation–weathering imbalance amounts to a removal of 48.5 GtC from the system. The mean $\delta^{13}$C signature of the burial flux, combining mean CaCO$_3$ ($\delta^{13}C_{CaCO_3}$=2.9 ) and $C_{org}$ ($\delta^{13}C_{POC}$= -20.2 ) signatures, over these during the first 10 kyr amounts to -8.7 and is thus larger than the mean $\delta^{13}$C signature of the weathering input. The contribution to differences in the carbon isotopic budget relative to the closed system is dominated by changes in POC cycling and its associated ($\delta^{13}C$ = -9.1 , determined after the spin-up). Hence, while changes in the CaCO$_3$ cycle play a smaller, but non-negligible role. Overall, $^{13}$C is lost from the atmosphere–ocean system and $\delta^{13}$C$_{DIC}$ is decreasing. The resulting difference in the $\delta^{13}$C$_{DIC}$ perturbation between open and closed system causes an erroneous mass balance inference for the terrestrial biosphere, when applying conventional closed system equations (Eq. A2 and A3). On a timescale of 10 , underestimation of the terrestrial carbon inventory change amounts to 30 . The error increases as times evolves. The contributions to this decrease in $\delta^{13}$C are 65 and 35 for changes in the cycling of POC and CaCO$_3$, respectively (as calculated 10 after the pulse).

In the third pulse experiment, changes in global mean air temperature and atmospheric CO$_2$ concentration affect weathering fluxes of CaCO$_3$ and CaSiO$_3$ following Colbourn et al. (2013). The implemented feedbacks cause a reduction in modeled weathering fluxes in response to decreased atmospheric CO$_2$ and temperature over the course of the experiment. Decreased weathering fluxes of CaCO$_3$ and CaSiO$_3$ lead to a decline in alkalinity supplied to the ocean resulting in a net transfer of carbon to the atmosphere. This further reduces the initial CO$_2$ perturbation (Fig. 4A). The mean $\delta^{13}$C value related to weathering is -9.1 ‰ thereby leading to a slightly higher $\delta^{13}$C$_{DIC}$ perturbation at 10 kyr compared to the open system experiment with fixed weathering fluxes. The error related to the calculated land perturbation is comparable on a 10 kyr time horizon, and increases less as time progresses further compared to the open system setup with fixed weathering.
Figure 5. Histogram (blue bars, normalized) and kernel density estimate (blue line) of $\Delta$land where parameter combinations fulfill all four data-based constraints. Vertical thick black line indicates the median $\Delta$land of $\sim$850 GtC and thin black lines show the $\pm 1\sigma$ range. Remaining four lines show kernel density estimates of $\Delta$land when considering one data-based constraint at a time.

From these pulse experiments it becomes clear that sediment and weathering interactions affect the carbon and carbon isotope $\delta^{13}C$ budgets on deglacial timescales. The contribution to differences in the carbon isotopic budget between the closed and open system is dominated by changes in POC cycling and its associated time scales. Taken at face values, the results suggest that the classical $\delta^{13}C$ signature, while changes in the $\text{CaCO}_3$ cycle play a smaller, but non-negligible, role on the isotopic budget. Overall we find that calculations of C-based estimates of $\Delta$land are substantially biased low, because of the neglect of open system processes. However, uptake by land carbon is the only forcing in these idealized pulse simulations, whereas climate and biogeochemical cycles underwent a massive reorganization over the glacial–interglacial transition. This reorganization affected open system processes and the $^{13}C$ budget in addition to carbon uptake by the land biosphere. In the next section, we estimate $\Delta$land that rely on the closed system isotopic mass balance approach are underestimated by taking into account deglacial processes.

3.2 Ensemble approach to constrain $\Delta$land

We now turn to estimate $\Delta$land. We recognize represent the deglacial reorganization of the ocean, as represented in the standard model setup and complemented by the seven deglacial carbon cycle mechanisms and constrained by the four observational
targets within an open system model framework. Applying the Bern3D emulator (Eq. 2) to our 500,000 member parameter ensemble, we find that the prior range in $\Delta$land, ranging from -500 to 1,500 GtC, is constrained to $\sim$450 to $\sim$1,250 GtC ($\pm 1\sigma$ range, see Fig. 5) in the light of the four observational targets. The constrained median amounts to $\sim$850 GtC, and almost no ensemble members which fulfill all four observational constraints are found for negative $\Delta$land values. The constrained ensemble thus clearly suggests a positive $\Delta$land. **When considering single constraints only.**

Next, we show that it is not sufficient to use $\Delta^{13}C_{DIC}$, or any other target, in isolation to constrain $\Delta$land values cover. The probability distribution of $\Delta$land covers the whole range of sampled values almost uniformly (Fig. 5) when applying only one out of the four targets. Considering either $\Delta^{13}C_{DIC}$ or $\Delta^{13}C_{atm}$ together with $\Delta CO_{2}$, $CO_{2}$ as constraints moves the $\Delta$land distribution towards the **constrained multi proxy-based** distribution (blue line in Fig. 5; not shown). The We conclude that the incorporation of multiple constraints is thus essential in narrowing down the essential to narrow uncertainties in $\Delta$land estimate.

With the chosen ranges in the seven parameters, our unconstrained ensemble yields a very large spread in the four target variables. $\Delta^{13}C_{DIC}$ ranges from -0.77 to 1.25 %e; $\Delta^{13}C_{atm}$ from -0.9 to 1.32 %e; $\Delta CO_{2}$ from -107 to 181 ppm; and $\Delta CO_{3}^-$ from -49 to 42 μmol kg$^{-1}$. Within the optimization procedure, we specify that only defined, observation-based ranges of these target variables are allowed (Section 2.2.3), and that all four of these constraints have to be met at the same time. This yields specific distributions of the target variables within our constrained ensemble (Fig. 6, magenta black lines). The distribution of $\Delta^{13}C_{atm}$ values, for instance, is almost uniform within the defined observational range (Fig. 6B), while $\Delta CO_{2}$, $\Delta CO_{3}^-$, and $\Delta^{13}C_{DIC}$ values cluster towards the lower end of the specified target ranges (Fig. 6C,D,A). This is a result of the underlying carbon cycle–climate processes as tested within Bern3D. For example, it remains difficult to model a PI-LGM $\Delta CO_{2}$ of $\sim$90 ppm under the constraint of meeting the other three observational targets. Most ensemble members represent a $\Delta CO_{2}$ of $\sim$82 ppm (Fig. 6C). Higher $\Delta CO_{2}$ are present in the unconstrained ensemble, but are generally associated with even more negative $\Delta CO_{3}^-$ (not shown), which is outside the defined target range for $\Delta CO_{3}^-$. As a further test of our optimization scheme and the resulting representation of our target variables within the constrained ensemble, we consider different subsets of our ensemble where the range of the input parameter $\Delta$land is modified. We define four such and define four sets where $\Delta$land is allowed to be maximally 1,000, 750, 450, and smaller than 0 GtC (Fig. 6, blue, green, orange, and grey-magenta lines). Only one target variable, $\Delta^{13}C_{DIC}$, is sensitive to this restriction within the constrained ensemble and positive $\Delta$land. The distributions of $\Delta^{13}C_{atm}$, $\Delta CO_{2}$, and $\Delta CO_{3}^-$ are almost identical for all subsets of $\Delta$land with the exception of $\Delta$land smaller than 0 GtC. Hence, these three targets do not per se discriminate between smaller and larger estimates of positive $\Delta$land. In contrast, restricting $\Delta$land to smaller values shifts the realized $\Delta^{13}C_{DIC}$ towards lower values and the mean of the constrained distribution shifts away from the mean observational estimate of 0.34 %e (Fig. 6A). For example, $\Delta^{13}C_{DIC}$ is always below 0.35 %e for $\Delta$land below 450 GtC and always below 0.23 %e for negative $\Delta$land estimates. Thus, not only very few process combinations yield a negative $\Delta$land in our 500,000 member ensemble (Fig. 5), these solutions are in addition biased low in $\Delta^{13}C_{DIC}$. Also, it is visible that for Further, solutions with negative $\Delta$land not only $\Delta^{13}C_{DIC}$ values are also yield $\Delta CO_{2}$ and $\Delta CO_{3}^-$ values at the very low end of the observational range, but the same holds true for $\Delta CO_{2}$ and $\Delta CO_{3}^-$ constraints (Fig. 6B,C). Visual inspection of
Figure 6. Histogram (thin lines, normalized) and kernel density estimate (thick lines) of (A) $\Delta \delta^{13}C_{DIC}$, (B) $\Delta \delta^{13}C_{atm}$, (C) $\Delta CO_2$, and (D) $\Delta CO_3^{2-}$ values for parameter combinations calculated with the emulator that fulfill the observational constraints. Different colors in all four panels correspond to sub-samples divided along $\Delta land$ values that are smaller than 1,500, 1,000, 750, 450, and 0 GtC. Darker grey shading indicates the range of the respective observational constraint.

Fig. 6A shows a number of samples cluster around the best guess estimate of $\Delta \delta^{13}C_{DIC}$, except when $\Delta land$ is restricted to below 450 GtC. Taken together, our framework strongly suggests that the land biosphere sequestered carbon over the deglaciation and that the sequestered amount is likely larger than 450 GtC.

3.3 Carbon and $\delta^{13}C$ changes in LGM to PI sensitivity simulations

We now discuss the results obtained with the Bern3D model for the transient LGM to PI simulations. This section 3.3 is intended for readers interested in a more detailed quantification of deglacial carbon cycle mechanisms. We recall that the model is initialized with a spin up for PI conditions and simulations started 40 kyrBP under LGM forcing (Fig. 3). Forcing the model from PI into an LGM state by adjusting GHG radiative forcing, ice sheet extent and albedo, orbital forcing, and coral reef...
regrowth (standard simulation) applying standard forcings leads to a shoaling and slight slowdown of the Atlantic meridional overturning circulation of about 4.4 Sv (PI steady state: 18 Sv) and a global cooling of the ocean and the atmosphere of about 1.4 °C and 3 °C over the glacial, respectively. New production of organic matter decreases from 11.98 Gt C yr⁻¹ to a mean value over the glacial of 11.25 Gt C yr⁻¹. The oceanic carbon inventory increases at the expense of the atmosphere, yielding a higher CO₃²⁻ concentration during the glacial and a PI-LGM ∆. Turning to the deglaciation, CO₂ of increases by 27.8 ppm (Fig. 7). ∆δ¹³C from LGM to PI, while δ¹³C atm and ∆δ¹³C DIC show little change between LGM and PI (Fig. 7). By applying the standard transient forcings alone, neither the ∆δ¹³C constraints in the atmosphere and ocean nor the ∆CO₂ constraint is met, calling for the consideration of additional processes.

3.3.1 Sensitivity of carbon cycle processes to changes in additional deglacial processes

The mean δ¹³C signature of the ocean–atmosphere system changes in response to potential deglacial processes (Fig. 7A-B). For simplicity, we focus on the change in mean ocean δ¹³C DIC (Fig. 7A) as the ocean holds about 20 times more carbon than the atmosphere and interactive land biosphere together. The factorial simulations reveal a ∆δ¹³CDIC (PI-LGM) between -0.3 ‰ to more than 0.6 ‰ when halving or doubling the weathering flux of organic material, respectively. Prescribed changes in Southern Ocean (SO) wind stress, inducing changes in ocean circulation, the organic matter remineralization profile, and in the rain ratio lead to a change in the ocean signature of about -0.2 ‰, while changes in the SO air–sea gas transfer rate and in coral reef growth have a small influence on ∆δ¹³CDIC. In summary, not only changes in terrestrial carbon storage but several processes, that were potentially important for the reorganisation of the deglacial carbon cycle, influence the mean ocean. The influence of these mechanisms on δ¹³C signature in comparison to the observed deglacial change of 0.34 ± 0.19. Their influence δDIC cannot be ignored when estimating ∆land based on reconstructed changes in δ¹³C.

Reconstructions of the change in δ¹³C in the atmosphere and ocean differ in magnitude. The change in the atmosphere (0.1 ± 0.05 ‰) is about one third of the oceanic change (0.34 ± 0.19 ‰). Changes in the organic weathering flux and terrestrial carbon storage affect ∆δ¹³CDIC and ∆δ¹³Catm in an identical way. Changes in SO wind stress, rain ratio, coral reef growth, and the remineralization of organic material have a similar, though not completely identical, influence on the two isotope variables. In contrast, changes in the SO air–sea gas transfer rate alter ∆δ¹³Catm, while hardly changing ∆δ¹³CDIC. The strong sensitivity of δ¹³Catm to changes in SO gas transfer rate is due to the large disequilibrium between the atmosphere and surface ocean. This makes this process, in addition to temperature driven changes in fractionation, potentially important in setting atmospheric δ¹³C independently from mean ocean δ¹³CDIC change (see also Fig. C2B).

Considering ∆CO₂ and ∆CO₃²⁻, the changes in investigated mechanisms significantly influence atmospheric CO₂ and deep Pacific carbonate ion concentration (with the exception of air–sea gas transfer rate). A general relationship becomes apparent. A positive change in ∆CO₂ is always accompanied by a negative change in ∆CO₃²⁻ and vice versa (Fig. 7C-D). Thus, the explanation of the observed deglacial increase in CO₂ of ∼90 ppm requires a deglacial decrease in deep Pacific carbonate ion concentration (Fig. 6D), at least in our model setting and considering the mechanisms implemented in the standard and the factorial simulations. Changes in alkalinity and DIC (PI-LGM) for the standard forcing amount to about 500 Gt eq and 365 Gt C, respectively. In the case of rain ratio, coral reef growth, and changes in the remineralization profile, changes in
Figure 7. Sensitivity of (A) $\Delta^{\delta^{13}}C_{DIC}$, (B) $\Delta^{\delta^{13}}C_{atm}$, (C) $\Delta CO_2$, and (D) $\Delta CO_2^{-3}$ to changes in Southern Ocean wind stress (SO wind), Southern Ocean gas exchange-transfer rate (SO kgas), export rain ratio, shallow water carbonate deposition (coral), remineralization profile (rem. profile), organic weathering flux (org. wea.), and land carbon uptake (land). Sensitivities are shown relative to the std. standard forcings (std. forc., white dots). Values in panel (C) correspond to the entries in Table 2. Black crosses with error bars show estimates based on measurements ($\Delta CO_2$: Petit et al. (1999); Monnin et al. (2001); Siegenthaler et al. (2005); Lüthi et al. (2008); Köhler et al. (2017); $\Delta^{\delta^{13}}C_{atm}$: Schmitt et al. (2012); $\Delta CO_2^{-3}$: Yu et al. (2013); Qin et al. (2017); Luo et al. (2018); $\Delta^{\delta^{13}}C_{DIC}$: Peterson et al. (2014)). Dots in dark grey shading show absolute and in light grey shading relative (to std. forcings) values. $\Delta$ indicates the PI-LGM difference.
Figure 8. $\Delta$CO$_2$ for defined sensitivity experiments plotted against (A) $\Delta$δ$^{13}$C$_{atm}$, (B) $\Delta$δ$^{13}$C$_{DIC}$, (C) ΔCO$_3^-$, and (D) $\Delta$δ$^{13}$C$_{atm}$ vs. $\Delta$δ$^{13}$C$_{DIC}$. Black crosses indicate results with std. forcings alone and dark grey shadings show ranges for data based constraints ($\Delta$CO$_2$: Petit et al. (1999); Monnin et al. (2001); Siegenthaler et al. (2005); Lüthi et al. (2008); Köhler et al. (2017); $\Delta$δ$^{13}$C$_{atm}$: Schmitt et al. (2012); ΔCO$_3^-$: Yu et al. (2013); Qin et al. (2017); Luo et al. (2018); $\Delta$δ$^{13}$C$_{DIC}$: Peterson et al. (2014)). For the direction of change in the forcings see Fig. 7. Note that in (A) changes in Δland that yield a positive ΔCO$_2$ are overlain by the organic weathering flux line.

alkalinity and DIC are largest and occur close to a 2:1 ratio. Changes in land carbon uptake remove carbon from the system, leading to changes in alkalinity via CaCO$_3$ compensation whereas in the case of the organic weathering flux, not only carbon is added/removed from the system but also nutrients and alkalinity, following Redfield ratios (see appendix A). Changes in alkalinity and DIC resulting from changes in Southern Ocean wind stress and gas transfer rate are small.
3.3.2 Response relationships between different carbon cycle properties

The results for the target variables are plotted against each other to gain further insight on the relative importance of the different mechanisms to meet the observational constraints (Fig. 8). This yields a characteristic slope for each target pair and mechanism. For example, this slope is negative and equates to about -90 ppm per ‰ for the pair $\Delta CO_2$ and $\Delta \delta^{13}C_{DIC}$ and the mechanism of land carbon storage (grey line in Fig. 8A). Roughly similar slopes are found for variations of the organic weathering flux, SO wind stress, and SO air–sea gas transfer rate. Though, the overall impact of SO air–sea gas transfer rate changes on these targets is small. Visual inspection of Fig. 8A reveals that variations in these four processes alone cannot prompt the model results to agree with these two observational constraints. The slopes are too small in magnitude and either $\Delta CO_2$ or $\Delta \delta^{13}C_{DIC}$ remains outside the observational range when varying these four processes in combination.

The four processes are effective in varying $\Delta \delta^{13}C_{DIC}$ but relatively ineffective in varying $\Delta CO_2$. Processes that have a much larger impact on $\Delta CO_2$ than on $\Delta \delta^{13}C_{DIC}$ are related to the CaCO$_3$ cycle (coral reef growth and rain ratio) and changes in the upper ocean remineralization of particulate organic matter. This implies that variations in $\Delta$land, organic weathering, and ocean circulation (SO wind) are required to meet the $\Delta \delta^{13}C_{DIC}$ target, while variations in coral reef regrowth, rain ratio, and upper ocean remineralization depth are necessary to meet the $\Delta CO_2$ target. Indeed, the median changes for the parameters in the observationally constrained ensemble (Fig. C2) imply a deeper remineralization of organic matter and a reduced rain ratio at LGM than PI and a three to four times higher amount of CaCO$_3$ deposition during coral reef regrowth than applied in the standard run. These, together with a reduced SO wind stress at the LGM, all contribute to a deglacial increase in CO$_2$. The associated decrease in $\Delta \delta^{13}C_{DIC}$ from these changes is in most Monte Carlo realizations more than compensated by a deglacial increase in land carbon inventory, and in many realizations by an increase in the glacial organic weathering flux (Fig. C2, 8F-G). In general, the smaller the change in $\Delta$land, the larger the increase required in the glacial flux of organic weathering.

Slopes for the target pair $\Delta \delta^{13}C_{atm} – \Delta CO_2$ resemble the $\Delta \delta^{13}C_{DIC} – \Delta CO_2$ pair (Fig. 8A-B). Variations in SO air–sea gas transfer rate, SO wind stress, $\Delta$land, and organic weathering flux show relatively high sensitivity for $\Delta \delta^{13}C_{atm}$ and low sensitivity for $\Delta CO_2$, whereas coral reef regrowth, rain ratio, and upper ocean remineralization changes exert a large impact on $\Delta CO_2$ and affect $\Delta \delta^{13}C_{atm}$ little (Fig. 8B). In contrast to the $\Delta \delta^{13}C_{DIC} – \Delta CO_2$ target pair (Fig. 8A), the slopes for the $\Delta \delta^{13}C_{atm} – \Delta CO_2$ pair do not fall on roughly two characteristic slopes but vary across processes (Fig. 8B).

The clear relationship between oceanic alkalinity and oceanic carbon uptake (Fig. 7C-D) is also visible in Fig. 8C. Higher alkalinity during the glacial (negative $\Delta CO_3^{2–}$) generally results in a larger drawdown of atmospheric CO$_2$ (positive $\Delta CO_2$). An interesting exception is that a modest deepening of the remineralization of organic matter in the upper ocean leads to large changes in atmospheric CO$_2$, while at the same time hardly changing deep Pacific $CO_3^{2–}$, making it a potentially important process to fulfill the $\Delta CO_2$ target without shifting $\Delta CO_3^{2–}$ away from the observationally constrained range (Fig. 8C). Accordingly, the remineralization depth of organic matter is deeper at the LGM than PI in all Monte Carlo realizations (Fig. C2E).
Next, we consider the $\Delta \delta^{13}C_{DIC} - \Delta \delta^{13}C_{atm}$ target pair (Fig. 8D). The difficulty in simultaneously fulfilling the atmospheric and oceanic $\delta^{13}$C constraints with a negative $\Delta$land is becoming obvious. With negative $\Delta$land, $\Delta \delta^{13}C_{atm}$ and $\Delta \delta^{13}C_{DIC}$ both are moved towards negative values relative to std. forcings. Therefore, the isotopic constraints could only be fulfilled with increased glacial organic weathering flux such that $\Delta \delta^{13}C_{atm}$ and $\Delta \delta^{13}C_{DIC}$ both are moved towards positive values relative to the std. forcings (Fig. 8D). Considering the $\Delta$CO$_2$ and $\Delta$CO$_3^-$ constraints (Fig. 8C) implies that negative $\Delta$land can only be offset by increased glacial organic weathering or rain ratio. However, reaching a $\Delta$CO$_2$ of 80–100 ppm remains difficult for negative $\Delta$land when combining information from all panels of Fig. 8. This explains why only very few solutions with negative $\Delta$land and no solutions with $\Delta$land smaller than -220 GtC are found in the Monte Carlo simulations (Section 3.2).

### 3.3.3 Sensitivity of the spatial distribution of $\Delta \delta^{13}C_{DIC}$ to changes in deglacial processes

In this section, we describe the spatial distribution of changes in $\Delta \delta^{13}C_{DIC}$. Despite little change on global average, the reference run with standard LGM forcings (Fig. 9A) shows substantial spatial changes in $\Delta \delta^{13}C_{DIC}$ from internal reorganization linked to ocean circulation changes. Reduced glacial ocean ventilation as evidenced in changes in ideal age leads to the accumulation of light carbon from marine export productivity in the ocean interior. Circulation changes hence explain the variability in the spatial pattern of $\Delta \delta^{13}C_{DIC}$ in Fig. 9A. In particular, the deepening and strengthening of the AMOC during the deglaciation lead to positive $\Delta \delta^{13}C_{DIC}$ changes in the deep Atlantic and to negative $\Delta \delta^{13}C_{DIC}$ changes in the upper Atlantic, consistent with proxy reconstructions. The small mean change in $\Delta \delta^{13}C_{DIC}$ results from a complex interplay of processes affecting the export of POC and CaCO$_3$ as well as sedimentation-weathering imbalances.

The sensitivity simulations may be grouped by processes with small and large changes in the spatial pattern of $\Delta \delta^{13}C_{DIC}$. $\Delta$land, coral reef regrowth, and organic weathering flux affect the input of carbon and carbon isotopes into the atmosphere–ocean system, leading to relatively uniform patterns of change (Fig. 9B, F, and H). Changes in SO wind stress, SO gas transfer rate, rain ratio, and upper ocean remineralization profile on the other hand mainly lead to a redistribution of carbon and carbon isotopes in the atmosphere–ocean system amplified by sediment interactions (Fig. 9C,D,E,G).

The removal of light land carbon uptake of carbon by the land biosphere leaves the atmosphere and ocean relatively enriched in $\delta^{13}$C (Fig. 9B). As a result of the carbon removal from the coupled system, changes in bottom water concentrations of CO$_3^{2-}$ and O$_2$ may influence sedimentation fluxes of POC and CaCO$_3$ locally, and lead to small local patterns (see for instance negative anomalies in $\Delta \delta^{13}C_{DIC}$ in the NADW region of Fig. 9B). Generally though, a smooth pattern emerges for changes in $\Delta$land. Similarly, the doubling of the organic weathering flux during the glacial results in a strong uniform change in $\Delta \delta^{13}C_{DIC}$ (Fig. 9H and Fig. 7A) as more isotopically light carbon is added to the system at the LGM than PI. Increased rates of coral reef regrowth have little impact on $\Delta \delta^{13}C_{DIC}$ as the isotopic signature is similar for DIC and deposited CaCO$_3$ (Fig. 9F). The slight increase in $\Delta \delta^{13}C_{DIC}$ in response to enhanced coral reef regrowth has been attributed to changes in the fractionation during marine photosynthesis (Freeman and Hayes, 1992; Menviel and Joos, 2012).

Changes in the other four mechanisms not only affect the mean $\Delta \delta^{13}$ affect the spatial pattern of $\Delta \delta^{13}C_{DIC}$ (see Fig. 7A), but also change the spatial pattern significantly (Fig. 9C, D, E, and G). As described we note that these patterns result from
Figure 9. Simulated deglacial change in the isotopic signature of DIC, $\Delta \delta^{13}C_{DIC}$ (PI minus LGM). Values are displayed along sections across the Atlantic, Southern Ocean, and Pacific and in response to standard deglacial forcings and (B) to (H) due to changes in individual mechanisms as inferred from factorial simulations. $\Delta \delta^{13}C_{DIC}$ are is shown for a deglacial carbon uptake of 445 GtC by the land biosphere (B), Southern Ocean wind stress (C) and gas transfer rate (D) reduced scaled by 0.6 at LGM a factor of 0.4 during the glacial relative to PI, rain ratio as reduced by 0.45 at LGM relative to PI 0.045 during the glacial (E), an additional deglacial shallow water carbonate deposition scaled by a factor of $\sim$1.500 GtC 4 (F), a change in the organic matter remineralization between a linear profile during the glacial and the standard depth-scaling (G), and for a 400-towfold increase in the organic weathering flux during the glacial compared to PI (H). The absolute change in $\Delta \delta^{13}C_{DIC}$ is shown in panel (A) for the standard forcings (bright colors), while panel (B) to (H) show the difference relative to the standard run (pastel-colored).
adjustment processes acting on different timescales. We recall the forcing history for changes in Southern Ocean wind stress, Southern Ocean gas transfer rate, the rain ratio, the remineralization profile, and the organic weathering rate; PI values are set to LGM values at 40 kyr BP, kept constant from 40 kyr BP to 18 kyr BP, scaled back to the PI value over the termination (18 kyr BP to 11 kyr BP) and kept constant thereafter (see also Fig. 3 and Table 2). Results are therefore not directly comparable to the step-change experiments described by Tschumi et al. (2011). As discussed in detail by Tschumi et al. (2011) and Menviel et al. (2015), a more poorly ventilated ocean due to reduced wind stress leads to an increase in $\delta^{13}C_{atm}$ and a subtle initial decrease in $\delta^{13}C_{DIC}$. Changes in temperature and circulation in response to decreased Southern Ocean wind stress at the beginning of our simulation (Fig. 3F) lead to lower marine oxygen concentrations and thus an increase in POC sedimentation. The following Subsequent removal of $^{13}$C depleted carbon from the ocean increases $\delta^{13}C_{DIC}$. Restoring wind stress to its initial value over the deglacial reverses the changes described above. As the sediment feedbacks act on longer timescales, propagation of the signal to the whole ocean is not yet achieved (Fig. 9C). A complex interplay of processes such as changes in circulation and subsequent changes in export fluxes, oxygen concentrations, and remineralization of POC as well as changes in the lysocline and CaCO$_3$ cycling spatially overlay each other, yielding the $\Delta \delta^{13}C_{DIC}$ distribution seen in Fig. 9C.

Ocean–sediment interactions also play an important role for $\delta^{13}C_{DIC}$ in the case of changes in the Southern Ocean gas transfer rate. Decreasing the gas transfer rate at the beginning of the 40 kyr simulation (Fig. 3F) leads to a strong increase in $\delta^{13}C_{atm}$ and an initial subtle decrease in $\delta^{13}C_{DIC}$. The ocean–sediment interactions are very similar to as described for SO wind stress changes above. The sedimentation fluxes of POC and CaCO$_3$ increase in response to reduced oxygen concentrations from a lower gas transfer rate and changes in the CO$_3^{2−}$ concentration, as does the $\delta^{13}C$ signature of the flux, leading to a removal of $^{13}$C from the ocean. From LGM to PI the opposite process takes places (increasing gas transfer back to its initial value), however, due to the long timescales of ocean–sediment interactions, the signal has not propagated to the whole ocean and in the Atlantic and Pacific negative $\Delta \delta^{13}C_{DIC}$ from the glacial still prevail (Fig. 9D).

As discussed in detail in Tschumi et al. (2011), responses in $\delta^{13}C$ from reducing the export rain ratio are small in both the ocean and atmosphere (Fig. 7A-B) and arise from weathering-sedimentation imbalances and changes in the isotopic value of CaCO$_3$ and POC. This negative perturbation in $\delta^{13}C$ is not removed entirely over the deglacial and Holocene due to the long ocean–sediment response timescale, leaving the generally negative $\Delta \delta^{13}C_{DIC}$ as seen in Fig. 9E.

Finally, a change to a linear remineralization profile in the upper 2 km during the glacial leads to a dipole pattern in $\delta^{13}C_{DIC}$ with enriched values in the upper 1 km of the ocean and depleted values underneath (not shown). Over Across the deglacial, the process is reversed, yielding the pattern in $\Delta \delta^{13}C_{DIC}$ seen in Fig. 9G from the overlap of different response timescales. Overall, applied changes in the remineralization profile lead to loss of carbon from the atmosphere–ocean system from LGM to PI.

In summary, the carbon and carbon isotope balance in our model simulations can change as a result of altered input fluxes, from changes in the surface ocean $\delta^{13}C$ signatures, which are then reflected in POC and CaCO$_3$ export fluxes and coral reef regrowth, and from altered sedimentation fluxes as a result of altered bottom water CO$_3^{2−}$ and O$_2$ concentrations. For all seven mechanisms, weathering-sedimentation imbalances and feedbacks exert an impact on the carbon and carbon isotope budget.
and need to be taken into account on glacial–interglacial timescales. Also, the overlap of different response timescales for atmosphere–ocean and ocean–sediment interactions shape the patterns in $\Delta \delta^{13}C_{DIC}$ shown in Fig. 9.

4 Discussion

We show that carbon exchange with ocean sediments and the lithosphere biases earlier $\delta^{13}C$-based estimates of deglacial change in land carbon storage significantly towards lower values. This finding appears very robust considering modern estimates and reconstructions of ocean–sediment and weathering fluxes, and our understanding of the marine carbonate chemistry. These geological fluxes are an integral part of the Earth system. Many earlier estimates of land carbon storage change rely on the budget of the stable carbon isotope, but neglected the isotopic exchange fluxes with marine sediments and the lithosphere. This approach became popular, perhaps as it is simple and transparent to solve two global budget equations.

However, results from idealized pulse-release experiments as well as from transient deglacial simulations show that this neglect is not justified. Several processes that were potentially important for the reorganisation of the deglacial carbon cycle influence the mean ocean $\delta^{13}C$ signature significantly. Their influence cannot be ignored when analyzing the budget of $\delta^{13}C$ on deglacial timescales. The importance of ocean–sediment and lithosphere fluxes in regulating past atmospheric CO$_2$ and carbon, isotopes, nutrient, and alkalinity inventories in the ocean is also emphasized in previous modelling studies (e.g. Huybers and Langmuir, 2009; Tschumi et al., 2011; Roth and Joos, 2012; Roth et al., 2014; Wallmann et al., 2015; Heinze et al., 2016). Data-based reconstructions of burial fluxes of calcium carbonate and organic matter (Cartapanis et al., 2016, 2018) reveal that these fluxes are more dynamic than often previously assumed.

For cost efficiency, we build a reduced-form emulator of the Bern3D model explicitly simulating the carbon exchange with ocean sediments and the lithosphere. Orbital changes and radiative forcing by greenhouse gases and ice albedo as well as freshwater input fluxes are taken into account. The strength of seven generic, archetypical deglacial carbon cycle processes was varied in a Bayesian Monte Carlo approach using 500,000 realizations. The model outcomes are used to constrain the deglacial change in the land biosphere inventory using four observational constraints. These constraints are the deglacial changes in marine ($\Delta \delta^{13}C_{DIC}$) and atmospheric ($\Delta \delta^{13}C_{atm}$) carbon isotopic composition, in atmospheric CO$_2$ ($\Delta$CO$_2$), and in deep Pacific carbonate ion concentration ($\Delta$CO$_3^{2−}$). This yields an observation-constrained probability distribution for the change in land carbon inventory, $\Delta$land, between the preindustrial (PI) and Last Glacial Maximum (LGM).

4.1 Uncertainties of this study

There are a number of uncertainties that are not explicitly addressed by in our approach. These may affect the median and distribution of our reconstruction of the deglacial change in land carbon inventory ($\Delta$land). We rely on characteristic forcing–response relationship as obtained with the Bern3D model and these may be different than in reality. Shortcomings related to coarse model resolution, simplified ocean dynamics, global parameterizations of ocean–sediment interactions or highly parameterized representation of biological processes may bias model outcome. Further, the current crop of models, including the Bern3D model, is not able to freely simulate reconstructed variations in atmospheric CO$_2$ and other biogeochemical
variables over the deglacial period. Nevertheless, the Bern3D model simulates the modern distribution of a range of water mass, ventilation, and biogeochemical tracers in good agreement with observational data (Roth et al., 2014). Global inventories and spatial distribution of biogenic CaCO$_3$, organic carbon, and opal in the sediments as well as their fluxes between the ocean, marine sediments and the lithosphere are in agreement with pre-industrial data-based estimates (see Appendix B and Table 1).

The Bern3D model simulates a weaker and shallower Atlantic Meridional Overturning Circulation at the LGM compared to the PI and the simulated anomalies in $\delta^{13}$C of DIC agree with corresponding reconstructions (see Fig. 9 in Menviel et al., 2012). Regarding dissolved oxygen, qualitative reconstructions show a general decrease in oxygenation at intermediate depths and increase in the deep ocean (Jaccard and Galbraith, 2012; Jaccard et al., 2014; Galbraith and Jaccard, 2015) across the deglaciation. This pattern is also reproduced (not shown) in Bern3D model simulations that match/come close to matching the four data-based constraints.

We selected a set of seven generic, archetypical processes that are varied in deglacial simulations in addition to the processes explicitly implemented in the Bern3D model. These archetypical processes are intended to represent the space of multi-proxy response relationships (Fig. 8) for all the different processes which plausibly influenced deglacial carbon cycle changes. This is a simplification. Though, we argue that the seven selected processes roughly cover the plausible range of multi-proxy responses (Fig. 8). In addition, uncertainties in the importance of these processes are explicitly considered in our Monte Carlo approach and contribute to the uncertainty range in our estimate of $\Delta$land.

For example, there are various carbon reservoirs with an isotopic light signature very similar to that of plant and soil carbon. These include organic carbon stored in ocean sediments and shelves (Broecker, 1982; Ushie and Matsumoto, 2012; Cartapanis et al., 2016), dissolved organic carbon (DOC) in the ocean (Hansell, 2013), or gaseous CO$_2$ stored in the unsaturated zones of aquifers from the decomposition of organic material (Baldini et al., 2018). Any carbon release or uptake from these reservoirs might be wrongly attributed to a change in the carbon inventory of the land biosphere given their very similar $\delta^{13}$C signature and similar multi-proxy response relationships (e.g., compare pink and black line in Fig. 8A-D for changes in organic weathering and land carbon). Indeed, changes in these reservoirs have been invoked as an alternative explanation for the reconstructed deglacial change in marine $\delta^{13}$C (Broecker, 1982; Zimov et al., 2006, 2009; Zech et al., 2011; Kemppinen et al., 2018). Here, we represent variations in these carbon stocks by varying weathering-burial imbalances of organic material in addition to adding variable amounts of isotopically light land carbon to the atmosphere in our 500,000 member ensemble. Thereby, we consider uncertainties related to inventory changes in these other isotopically light reservoirs as further elaborated in the next paragraph.

Information in Baldini et al. (2018) (their Table 1) implies that the deglacial change in CO$_2$ stored in the unsaturated zone of aquifers, neglected in the Bern3D model, is in the range of 12 to 86 GtC between the Last Glacial Maximum and the preindustrial period. This range is (Baldini et al., 2018) and small compared to the estimated change in land carbon of around 850 GtC. Perhaps more important, the modern inventory of refractory DOC is about 650 GtC (Hansell, 2013) and assumed constant in Bern3D, while changes in the small inventory of labile DOC are explicitly simulated. It is unclear how the DOC pool varied over the past. Lower temperature might have favored the preservation of DOC, while a smaller vegetation cover on land might have reduced the input of DOC into the ocean. Further, Cartapanis et al. (2016) suggest an
approximately linear decrease in the flux of organic carbon transferred to deep ocean sediments from around 28 GtC kyr\(^{-1}\) at the LGM to around 17 GtC kyr\(^{-1}\) during the Holocene, corresponding to a cumulative imbalance of around 55 GtC over the deglaciation. Here, we varied changes in organic carbon sources to the atmosphere–ocean system over a very wide range in comparison to the inventory changes discussed above. Organic-like organic-like carbon input by weathering at the LGM is varied in the unconstrained ensemble within 50\% to 200\% of its modern value. This corresponds over a range corresponding to a cumulative PI-LGM anomaly in this input flux of 780 to -1,560 GtC. These anomalies in input, though not directly comparable to inventory changes, are large in comparison to the inventory changes discussed here. In summary, a potential misattribution of changes in other organic carbon reservoirs to changes in \(\Delta\)land is included in our uncertainty estimate of \(\Delta\)land.

Uncertainties related to changes in the \(\delta^{13}C\) signature of land carbon, also leading to a potential misattribution when estimating \(\Delta\)land, are thought to be small on the global scale (Joos et al., 2004; Ciais et al., 2012). The \(\delta^{13}C\) signature of the terrestrial carbon uptake/release in this study was set to 24. In past studies, also slightly different \(\delta^{13}C\) signatures for the terrestrial biosphere have been used, ranging from about 25 to 23. To test the robustness of our \(\Delta\)land estimate to the inferred \(\delta^{13}C\) signature of the terrestrial carbon, two series of experiments were conducted with \(\delta^{13}C\) signatures of 25 and 23. The new sensitivities of the data-based target constraints to changes in \(\Delta\)land were determined and 100,000 parameter combinations were calculated. Resulting estimates of \(\Delta\)land vary only by a few tens of for both \(\delta^{13}C\) signatures. Considering the spread of possible \(\Delta\)land estimates in this study, the exact value of the \(\delta^{13}C\) signature of the terrestrial uptake within the considered range thus seems to play a minor role in determining \(\Delta\)land.

Our approach is limited in that we only consider the change between LGM and PI for four relevant proxy variables. This restriction allowed us to build a cost-efficient and non-linear emulator to explore a very large range of parameter combinations. Future efforts to refine estimates of \(\Delta\)land may explicitly consider the temporal evolution of the different proxies (Schmitt et al., 2012; Yu et al., 2014; Eggleston et al., 2016; Bauska et al., 2016; Peterson and Lisiecki, 2018) to study biogeochemical changes over the past glacial–interglacial cycles (Menviel et al., 2012; Heinze et al., 2016; Ganopolski and Brovkin, 2017). Heinze et al. (2016) applied a linear inverse modelling approach to approximate the evolution of about eighty proxy records for atmospheric CO\(_2\) and sedimentary CaCO\(_3\), opal, and \(\delta^{13}C\) of benthic and planktonic foraminifera over the last 130 kyr. This was done by scaling the magnitude of seven processes with the ice core deuterium signal in their annual mean circulation–biogeochemical model. Here, we explicitly consider seasonality and non-linearities in forcing–response relationships and restrict the focus to the deglaciation period. Additional proxy data may be included in future work. These include, for example, changes in oxygen (e.g. Jaccard and Galbraith, 2012; Galbraith and Jaccard, 2015; Hoogakker et al., 2015; Gottschalk et al., 2016), in export productivity and burial (e.g. Kohfeld et al., 2005; Anderson and Burckle, 2009; Cartapanis et al., 2016), in radiocarbon (e.g. Skinner et al., 2017), or in isotopes of protactinium, thorium, and neodymium (e.g. Böhm et al., 2015). Another avenue for improvement is to consider spatial patterns such as the documented changes in the patterns of \(\delta^{13}C\) of DIC between the upper and deep, and the Atlantic and Indo-Pacific ocean (e.g. Oliver et al., 2010; Peterson and Lisiecki, 2018) in the pattern of CO\(_2\)\(^{2-}\) (Fig 6 in Menviel et al., 2018).
4.2 Contribution of deglacial mechanisms to the CO\textsubscript{2} rise

Turning to the role of different mechanisms for the deglacial increase in atmospheric CO\textsubscript{2}, we find that ocean circulation changes only partly explain the deglacial CO\textsubscript{2} rise. Ocean circulation changes are not able to simultaneously explain glacial–interglacial changes in $\delta^{13}$C\textsubscript{DIC} and CO\textsubscript{2} in our model (see Fig. 8A), in agreement with earlier studies (e.g., Tschumi et al. (2011); Heinze et al. (2016)).

A modest, and plausible (Bendtsen et al., 2002; Matsumoto, 2007), deepening of the remineralization of organic matter in the upper ocean (see Fig. C2 panel E) leads to large changes in atmospheric CO\textsubscript{2}, while at the same time hardly changing deep Pacific CO\textsubscript{2}\textsuperscript{−}. This holds also for other biological mechanisms such as an increased nutrient utilization in response to iron fertilization or an elevated phosphorus input to the ocean (Meniel et al., 2012). This renders this class of processes to be potentially important to explain part of the deglacial CO\textsubscript{2} increase, as reconstructions suggest small LGM to PI changes in deep Pacific CO\textsubscript{2}\textsuperscript{−} concentrations (Yu et al., 2014). Accordingly, in all Monte Carlo realizations the remineralization depth of organic matter is deeper at the LGM than PI in our approach. We note that the modification in the remineralization profile of particulate organic matter is different than those applied by Meniel et al. (2012) and Roth et al. (2014) in simulations with the Bern3D model. Here, only the remineralization profile in the upper 2,000 is changed and, in contrast to these earlier studies, it is assumed that the fraction of exported particles that reach the sediments below 2,000 is not altered. The implicit assumption is that cooler temperatures at the LGM than PI only affect the dissolution of relatively small or labile particles in the upper ocean in a significant way, while large or refractory particles sink fast enough to reach ocean sediments both under LGM and PI conditions.

Our solutions also point to a significant role of alkalinity based mechanisms for the deglacial CO\textsubscript{2} increase. A decrease in the surface ocean alkalinity tends to increase the CO\textsubscript{2} partial pressure and in turn atmospheric CO\textsubscript{2}. Surface ocean alkalinity is governed by the cycling of CaCO\textsubscript{3} and, to a smaller extent, of organic carbon within the ocean as well as by the balance of alkalinity input by weathering and alkalinity loss by burial of CaCO\textsubscript{3}, and, to a smaller extent, by organic material (see Fig. C2D). The constrained ensemble yields a large extra burial of CaCO\textsubscript{3} as well as an increase in the rain ratio of CaCO\textsubscript{3} to organic matter by particle export over the deglaciation. This is qualitatively consistent with the reconstruction of Cartapanis et al. (2018) who suggest that the CaCO\textsubscript{3} burial flux (below 200 m) increased by about 0.2 GtC yr\textsuperscript{−1} from the LGM to the Holocene. This suggests that changes in alkalinity based mechanisms contributed to the deglacial CO\textsubscript{2} increase. In summary, our multi-proxy approach suggests, in agreement with earlier studies (e.g., Tschumi et al., 2011; Meniel et al., 2012; Heinze et al., 2016; Ganopolski and Brovkin, 2017), a combination of processes is responsible for glacial–interglacial CO\textsubscript{2} variations, while the attribution of change to individual mechanisms remains difficult. In addition, several processes that were potentially important for the reorganisation of the deglacial carbon cycle influence the mean ocean $\delta^{13}$C signature significantly. Their influence cannot be ignored when analyzing the budget of $\delta^{13}$C on deglacial timescales.
4.3 Comparison to other estimates of $\Delta$land

$\Delta$land estimates range from -400 GtC, e.g. a larger terrestrial carbon inventory during the glacial (Zimov et al., 2006, 2009; Zech et al., 2011; Kemppinen et al., 2018), to 1,500 GtC (Adams and Faure, 1998), based on a variety of methods each associated with its own uncertainties and limitations. Studies suggesting a negative $\Delta$land (Zimov et al., 2006, 2009; Zech et al., 2011; Kemppinen et al., 2018) point that the reconstructed deglacial $\delta^{13}$C$_{DIC}$ change is due to changes in other organic carbon reservoirs. These postulated changes would not only need to explain the observed deglacial change in $\delta^{13}$C$_{DIC}$, but in addition also explain the imprint of... The low end is based on reconstructions of the carbon content in paleosols (Zimov et al., 2006, 2009; Zech et al., 2011). These reconstructions are scarce, of local nature, and restricted to cold high-altitude or high-latitude regions, rendering extrapolations to the global scale speculative. In a recent data synthesis, Lindgren et al. (2018) suggest that the loss of carbon from northern hemisphere permafrost from the LGM to PI was lower than initially suggested by Zimov et al. (2009) and that total land carbon storage in northern mid- and high-latitudes increased by about 400 GtC over the deglaciation. Kemppinen et al. (2018) suggest a negative $\Delta$land on the isotopic budget. Considering additional data-based constraints, as proposed in this study, suggests based on results from an Earth System Model of Intermediate Complexity. However, these authors did not consider any isotopic constraints. Our results suggest negative $\Delta$land values to be highly unlikely. In addition, our results show that and $\Delta$land is likely larger than 450 GtC. Otherwise, solutions for $\delta^{13}$C$_{DIC}$ are significantly biased low compared to the data-based reconstruction.

On the other hand, The high end of the $\Delta$land estimates at the very high end are also in conflict with some data-based constraints and Hoogakker et al. (2016) point to the difficulty of deriving range, implying much smaller land carbon storage during the LGM than today, is based on estimates relying on pollen records (Adams et al., 1990; Van Campo et al., 1993; Crowley, 1995; Adams et al., 2009). Pollen and macrofossils recorded in various archives hold information on past vegetation distribution, but do not allow constraining carbon inventories neither in soils nor in vegetation, rendering the derivation of $\Delta$land estimates based on pollen data. We will illustrate some of these issues with the example of the study by Adams and Faure (1998) that provides reconstructions of $\Delta$land for three time slices, 18, 8, and 5 and assumes that about 1,400 of the regrowth of the terrestrial biosphere occurred between 18 and 8. This is in conflict with the evolution of the terrestrial biosphere uptake as proposed by Elsig et al. (2009). In their best guess scenario Elsig et al. (2009) assume a growth of the terrestrial biosphere of about 150 between 11 and 8 and 700 between 18 and 11 which is only 60% of the amount given by Adams and Faure (1998). While the growth of the terrestrial biosphere from 18 to 11 is a scenario estimate, the growth over the Holocene is constrained based on-difficult (Hoogakker et al., 2016). The ice core CO$_2$ and $\delta^{13}$C$_{atm}$ measurements to... records constrain the preindustrial terrestrial carbon uptake over the Holocene to about 250 GtC (Elsig et al., 2009). Assuming the total estimate of 1,500 GtC by Adams and Faure (1998) was right, this would leave more than 1,200 GtC to be transferred to the terrestrial biosphere between the LGM and the beginning of the Holocene. Considering possible processes that could achieve such a large transfer seems difficult. Generally this might pose an upper limit to $\Delta$land.

Our estimate of $\Delta$land appears consistent with a recent biome-based reconstruction of soil carbon storage (Lindgren et al., 2018). Lindgren et al. (2018) report an increase in soil carbon inventory of about 400 GtC for the region north of about 30°N
and considering changes in permafrost, mineral and peatland as well as in loess and subglacial soil. This is in agreement with our $\Delta$land estimate, when assuming an increase in carbon stored in vegetation and in tropical and southern hemisphere ecosystems and soils. Furthermore, modeling studies have generally yielded positive $\Delta$land estimates of about 400–900 GtC (Joos et al., 2004; Prentice et al., 2011; Brovkin et al., 2012; O’Ishi and Abe-Ouchi, 2013; Ganopolski and Brovkin, 2017; Davies-Barnard et al., 2017).

In summary, many lines of evidence point to a positive $\Delta$land and we suggest a likely range of about 450 to 1,250 GtC. Yet, the uncertainty in this estimate remains large, owing to both uncertainties in processes and in observational constraints. Unfortunately, the necessity to include exchange with sediments and the lithosphere and other deglacial processes in the isotopic budget adds complexity. This tends to increase the uncertainty range compared to the simplified and biased assessments that consider the isotopic budget in the closed ocean–land–atmosphere system. Further research is necessary to better understand the temporal evolution of the glacial/interglacial carbon cycle.

5 Summary and conclusions

We used a Bayesian approach to constrain the change in the land biosphere carbon inventory between the Last Glacial Maximum and the preindustrial period. Four data-based constraints are applied: the deglacial change in the $\delta^{13}$C signature of atmospheric CO$_2$ and of dissolved inorganic carbon in the ocean, the deglacial change in atmospheric CO$_2$ and in deep Pacific carbonate ion concentrations. The strength of generic, archetypical mechanisms for deglacial biogeochemical changes was varied in 500,000 simulations in an emulator, built from a large suite of Bern3D model simulations. Carbon, nutrient, alkalinity, and carbon isotope exchange with the lithosphere and ocean sediments is explicitly taken into account, in contrast to earlier studies which applied the isotopic budget of $^{13}$C to estimate deglacial change in land biosphere carbon.

We demonstrate in idealized and transient deglacial simulations that isotopic exchange with ocean sediments and the lithosphere is important for the budget of $^{13}$C on timescales of the deglaciation. We find that the carbon stocks in the land biosphere were likely around 450 to 1,250 GtC ($\pm 1 \sigma$ range of standard deviation) larger at preindustrial times than at the last glacial maximum. The median estimate is $\sim$850 GtC. This is much larger than earlier estimates based on the budget of the stable carbon isotope $^{13}$C. These earlier studies neglected interactions with the lithosphere and ocean sediments and the influence of other deglacial carbon cycle processes on the isotopic budget. This neglect biases their estimate significantly low. Our multi-proxy approach suggests that a combination of different mechanisms contributed to the deglacial increase in CO$_2$. These include, aside from ocean circulation, temperature, and salinity changes, a net removal of alkalinity from the surface ocean by increased burial of calcium carbonate in coral reefs and ocean sediments and potentially increasing export of calcium carbonate from the surface to the deep ocean. Further, changes in biological mechanisms such as a deglacial shoaling of the remineralization depth for organic matter may have been instrumental to increase atmospheric CO$_2$. The results demonstrate that ocean sediments and the weathering–burial cycle are an integral part of the Earth system playing a fundamental role on glacial–interglacial timescales.


Appendix A: The Bern3D model and pulse experiments

A1 The Bern3D model

The Bern3D EMIC features a three dimensional geostrophic ocean (Müller et al., 2006; Edwards et al., 1998) with an isopycnal diffusion scheme and Gent–Mc Williams parameterization for eddy-induced transport (Griffies, 1998). The model includes a thermodynamic sea-ice component coupled to a single layer energy–moisture balance atmosphere (Ritz et al., 2011). Further, a sediment module (Tschumi et al., 2011; Heinze et al., 1999), and a 4-box terrestrial biosphere (Siegenthaler and Oeschger, 1987) are coupled to the model.

The horizontal resolution is 41x40 grid cells (Roth et al., 2014; Battaglia and Joos, 2018a, b) and is the same for the ocean, atmosphere, and sea-ice components. In the vertical, the ocean has 32 logarithmically spaced layers. Wind stress at the surface is prescribed following the NCEP/NCAR monthly wind stress climatology (Kalnay et al., 1996). Carbonate chemistry, air–sea gas exchange for CO$_2$ and $^{14}$CO$_2$ is implemented according to OCMIP-2 protocols (Najjar and Orr, 1999; Orr et al., 1999) with updates for the $^{14}$C standard ratio and half-life (Orr et al., 2017), the calculation of the Schmidt number (Wanninkhof, 2014) and the carbonate chemistry (Orr and Epitalon, 2015). In order to match observational estimates of natural and bomb-produced $^{14}$C, the global mean air–sea transfer is reduced by 19 % compared to OCMIP-2 in order to match observation-based radiocarbon estimates (Müller et al., 2008). Another update with respect to the OCMIP-2 protocols concerns the gas transfer velocity which scales linearly with wind speed following Krakauer et al. (2006). Analog and consistent formulations are used for air–sea exchange of oxygen and $^{13}$CO$_2$.

Biogeochemical cycling in the model is detailed in Tschumi et al. (2011) and Parekh et al. (2008) with further documentation of results in follow-up studies (e.g. Menviel et al., 2012; Menviel and Joos, 2012; Roth and Joos, 2012; Roth et al., 2014; Menviel et al., 2015; Battaglia et al., 2016; Battaglia and Joos, 2018b, a). Dissolved inorganic carbon and semi-labile organic carbon (DIC, DOC), the corresponding isotopic forms (DI$^{13}$C, DO$^{13}$C, DI$^{14}$C, DO$^{14}$C), as well as alkalinity (Alk), phosphate (PO$_4$), oxygen (O$_2$), iron (Fe), silica (Si), and an ideal age tracer are explicitly transported by advection, diffusion, and convection. New production of organic matter is limited to the euphotic zone in the uppermost 75 m and calculated as a function of light availability, temperature, and phosphate and iron availability (Doney et al., 2006). Bacteria and plankton are not explicitly represented. Two thirds of the new production are transferred to the pool of dissolved organic carbon (DOC) and the remainder is exported as particulate organic carbon (POC) (Tschumi et al., 2008). The biological fluxes of C, PO$_4$, Alk, and O$_2$ and the corresponding elemental ratios in DOC and POC are linked by fixed Redfield ratios (P:Alk:C:O$_2$:Alk=1:17:117:170; Sarmiento and Gruber, 2006). DOC decays with a life time of 0.5 yr. POC is remineralized following a Martin’s Curve (Martin et al., 1987) given by

$$
F_{POC}(z) = F_{POC}(z_0) \times \left( \frac{z}{z_0} \right)^{-\alpha} \text{ for } z > z_0
$$

(A1)
where $z_0$ refers to the reference depth of 75 m (see also Roth et al., 2014). Export of calcium carbonate ($\text{CaCO}_3$) and of opal is computed from new production and availability of dissolved silica in the euphotic zone. The export rain ratio of $\text{CaCO}_3$ to POC is set to 0.083 in the absence of dissolved silica, while the production ratio of $\text{CaCO}_3$ to POC is reduced at the favor of opal production in regions with abundant availability of silicic acid. Particulate $\text{CaCO}_3$ and opal is remineralized below the euphotic zone with an e-folding depth scale of 5,066 m and 10,000 m, respectively.

A 10-layer sediment diagenesis module (Tschumi et al., 2011; Heinze et al., 1999) is used to explicitly calculate transfer fluxes to and redissolution fluxes from reactive sediment to the water column as well as loss fluxes to the lithosphere for nutrients, carbon and carbon isotopes. In equilibrium, loss fluxes to the lithosphere are equal to input fluxes by weathering. The sediment module dynamically calculates the transport, redissolution/remineralization, and bioturbation of solid material, the porewater chemistry, and diffusion in the top 10 cm of the sediment. Four solid tracers ($\text{CaCO}_3$, opal, POC, clay) and seven tracers in the porewater (DIC, $\text{DI}^{13}$C, $\text{DI}^{14}$C, alkalinity, phosphate, oxygen, and silicic acid) are modelled. The dissolution of $\text{CaCO}_3$ depends on the pore-water $\text{CO}_3^{2-}$ concentration. The oxidation rate of POC, on the other hand, depends on oxygen concentrations in the porewater and the weight fraction of POC in the solid phase of the sediment. Denitrification is not considered in this model version, but $\text{O}_2$ is not consumed below a threshold, somewhat reflecting the process of denitrification without modeling $\text{NO}_3^-$. The respective reaction rate parameters for $\text{CaCO}_3$ dissolution and POC oxidation are global constants (see Roth et al., 2014). The model assumes conservation of volume, i.e. the entire column of the sediments is pushed downwards if deposition exceeds redissolution into pore waters. Any solid material that is pushed out of the diagenetic zone (top 10 cm) disappears into the subjacent diagnostically consolidated zone (burial or loss flux) (see Tschumi et al., 2011, for more details). Carbonate chemistry within sediment pore waters is calculated as in the ocean by using the MOCSY routine of Orr and Epitalon (2015).

Weathering fluxes of $\text{PO}_4$, Alk, DIC, $\text{DI}^{13}$C, and silicic acid are added uniformly to the coastal surface ocean. The global weathering fluxes are set equal to the burial fluxes diagnosed at the end of the model spin-up. In the standard model setup these diagnosed input fluxes are kept constant. The pulse experiments include a setting where the input fluxes vary as a function of global mean surface air temperature and $\text{CO}_2$ following Colbourn et al. (2013). One of the factorial sensitivity runs assesses the impact of altered input fluxes of organic material across the deglacial. This weathering–burial cycle and the associated burial–nutrient feedbacks (Tschumi et al., 2011) are important for the mass balance of $^{13}$C and for the removal of marine perturbations in nutrients, carbon, and isotopes on deglacial time-scales. In other words, these processes directly affect the estimate of the change in land carbon from the $^{13}$C mass balance.

The atmosphere exchanges carbon and carbon isotopes with a four box land biosphere model (Siegenthaler and Oeschger, 1987). It includes a mass of 2,220 GtC and represents the carbon that is actively exchanged with the atmosphere. This 4-box reservoir is only used to simulate the dilution of an atmospheric isotopic perturbation by the land biosphere, but not to address changes in land carbon stocks. The carbon inventory of the land biosphere, and thus the dilution of an isotopic perturbation, is about 20 times smaller than that of the ocean–atmosphere–sediment system.

$^{13}$C stocks and flows are modeled in the atmosphere–ocean–sediment–landbiosphere system. $^{13}$C fluxes to the lithosphere associated with the burial of POC and $\text{CaCO}_3$ and weathering input are explicitly simulated. $^{13}$C fractionation is considered for
air–sea gas transfer, for carbonate chemistry, for the formation of CaCO$_3$, POC, DOC, and for photosynthesis on land, while no fractionation is assumed during remineralization on land and in the ocean. Gross air–to–sea and gross sea–to–air fluxes of $^{13}$CO$_2$ are implemented considering kinetic fractionation (Siegenthaler and Oeschger, 1987) and strongly temperature–dependent equilibrium fractionation between the various carbonate species (Mook, 1986). Isotopic fractionation during CaCO$_3$ formation is small and computed following Mook (1986). This results in an isotopically heavy signature of around 2.9 ‰ for CaCO$_3$. Fractionation during photosynthesis in the ocean and thus between dissolved CO$_2$ ([CO$_2$]) and POC and DOC is calculated according to Freeman and Hayes (1992). The fractionation increases logarithmically with [CO$_2$] in surface water and results in an isotopically light signature for POC and DOC of around -20 ‰. A lowering of [CO$_2$], or correspondingly of pCO$_2$, by about 10 % yields a change in the isotopic signature of POC by +0.55 ‰. The burial of POC and CaCO$_3$ results in an isotopic signature of the burial flux of around -9 ‰, intermediate between the isotopically light POC and the isotopically heavy CaCO$_3$ signature. In equilibrium, the burial flux is compensated by a weathering input flux of equal amount and signature. On land, a constant fractionation of 18.1 ‰ is applied for simplicity.

A2 Initialisation of Bern3D simulations

The model is spun up over 60,000 yrs to a preindustrial equilibrium corresponding to 1765 CE boundary conditions similar to the approach outlined in Roth et al. (2014). CO$_2$ is set to 278 ppm and $\delta^{13}$C of CO$_2$ to -6.305 ‰. The loss of tracers due to sedimentary burial is compensated during the spin-up by variable weathering fluxes in order to conserve oceanic inventories of tracers. After the system has equilibrated, the weathering fluxes are kept constant at the values diagnosed at the end of the spin-up in the standard setup.

A3 Bern3D pulse experiments

Idealized experiments with a pulse-like carbon removal from the atmosphere are conducted in i) a closed system, ii) an open system, and iii) an open system model configuration allowing for weathering feedbacks. In i), only the atmosphere–ocean–land biosphere model components are used and all tracers are conserved within these three reservoirs. In ii), the sediment module is added to the configuration outlined above and the carbon and tracer inventories within the atmosphere–ocean–land biosphere system are allowed to vary due to weathering-burial imbalances. In iii), weathering fluxes vary in response to changes in atmospheric temperature and CO$_2$ concentrations following the parameterizations given in Colbourn et al. (2013), accounting for changes in CaCO$_3$ and CaSiO$_3$ weathering on land.

Simulations are started from the end of the PI spin-up and atmospheric CO$_2$ and $\delta^{13}$C are computed in a prognostic way based on air–sea and air–land fluxes. In year 100 of the simulation, 100 GtC of carbon with a $\delta^{13}$C signature of -24 ‰ are instantaneously removed from the atmosphere, mimicking an immediate regrowth of the terrestrial biosphere. After the pulse, the model is run for 100 kyr and anomalies are expressed relative to a control run. Control simulations show negligible changes in CO$_2$ and $\delta^{13}$C as model drift is small.
Table B1. Old and new parameter values in the Bern3D model that were varied in the tuning of the sediment component.

<table>
<thead>
<tr>
<th>Variable</th>
<th>old value</th>
<th>new value</th>
</tr>
</thead>
<tbody>
<tr>
<td>ratio of Ca:P in calcifiers</td>
<td>0.3</td>
<td>0.333</td>
</tr>
<tr>
<td>redissolution length scale of calcite</td>
<td>2,900 m</td>
<td>5,066 m</td>
</tr>
<tr>
<td>diffusion coefficient in the sediment pore-water</td>
<td>$8 \times 10^{-6}$ cm$^2$ s$^{-1}$</td>
<td>$5.5 \times 10^{-6}$ cm$^2$ s$^{-1}$</td>
</tr>
<tr>
<td>reactivity for calcite in the sediment</td>
<td>1,000 l mol$^{-1}$ yr$^{-1}$</td>
<td>800 l mol$^{-1}$ yr$^{-1}$</td>
</tr>
</tbody>
</table>

The implied change in the land biosphere carbon stock based on a closed system assumption is calculated for all three model settings by solving the following equations for $\Delta M_{TER}$, i.e. $\Delta$land:

\[
\Delta(M_A + M_B + M_O) = 0
\]

\[
\Delta(\delta_A M_A + \delta_B M_B + \delta_O M_O) = 0
\]

with $M$ representing the inventory, $\delta$ the isotopic signature, $\Delta$ the respective time interval, and the subscripts $A$, $B$, and $O$ referring to the atmosphere, terrestrial biosphere, and ocean, respectively. The calculated $\Delta$land is then compared to the prescribed carbon uptake. This allows us to investigate the validity of the closed system assumption on different timescales.

**Appendix B: Sediment tuning**

The aim of the sediment tuning was to improve the representation of export, deposition, and dissolution fluxes of POC, CaCO$_3$, and opal, as well as the distribution of weight fractions of these tracers in the sediment module of the Bern3D model after the change of the horizontal model resolution introduced in Roth et al. (2014). One concern was the total amount of CaCO$_3$ in the reactive uppermost 10 cm of the sediment represented in the sediment module. Prior to tuning, the global CaCO$_3$ content in the model was $\sim$400 GtC. Using observational constraints on modern global bulk mass accumulation rates (MAR) (Cartapanis et al., 2016) and global carbonate MAR (Dunne et al., 2012; Jahnke, 1996) and the density, the bulk sediment, and the porosity in the uppermost 10 cm as used in the model, we estimate a target CaCO$_3$ stock of $\sim$1,000 GtC. The second concern was the opal burial that used to be $\sim$3 Tmol Si yr$^{-1}$ but with new observational data suggesting a burial flux of $\sim$6.5 Tmol Si yr$^{-1}$ (Tréguer and De La Rocha, 2013). Further, all other sediment related fluxes of POC, CaCO$_3$, and opal were tuned with regard to observational constraints (see Table 1).

The tuning was carried out in two steps. In the first step, three parameters were varied: the ratio of Ca:P in calcifiers, the redissolution length scale for the calcite profile, and the reactivity for calcite in the sediment. In the second step, the reactivity of calcite and the diffusion coefficient in the sediment pore-water were varied. For both steps, a 100 member LHS ensemble was run and skill scores calculated as outlined in Steinacher et al. (2013). For the calculation of the skill scores, ocean data from GLODAP v.2 (Lauvset et al., 2016) and the World Ocean Atlas 2013 version 2 (Locarnini et al., 2013; Zweng et al., 2013; Garcia et al., 2013a, b) datasets, for export, deposition, and burial of POC, CaCO$_3$, and opal values from Battaglia et al. (2016), Tréguer and De La Rocha (2013), Sarmiento and Gruber (2006), Milliman and Droxler (1996), Feely et al. (2004), and for the
Figure B1. (A) Distribution of CaCO$_3$ in the Bern3D model after tuning of the sediment component under pre-industrial boundary conditions compared to (B) sediment core-top CaCO$_3$ data (Cartapanis et al., 2018). Core-top data were gridded onto the Bern3D grid and are a mean of the top 10 cm over the past 6 kyr. Grey indicates land (within coastlines) or missing data.

sediment data compilations by Cartapanis et al. (2016, 2018) were used to compare the model performance to. The distribution of CaCO$_3$ in the sediments after the tuning as compared to observational data is shown in Fig. B1. Export, deposition, and burial fluxes for POC, CaCO$_3$, and opal and the global sediment inventories in the model with respective observational ranges from the literature are given in Table 1 and the old and new parameter values in Table B1. The distribution of other tracers is comparable to before and as outlined in Roth et al. (2014).

Appendix C: Additional Figures
Figure C1. Regression of emulator results for the four data-based constraints against results from a Bern3D LHS ensemble with the same parameter combinations (as outlined in Section 2.2). The regression given in the figure is used to correct emulator results for non-linearities (Eq. 2).
Figure C2. Histograms (colored thin line, normalized) and kernel density estimates (colored thick line) of applied changes in the seven mechanisms (A to G) for all emulator results that fulfill the four observational constraints. Colors of the lines correspond to sub-samples of these results according maximum changes in $\Delta$land of 1,500, 1,000, 750, 450, and 0 GtC. Thin red line gives the histogram of the prior distribution and vertical thick red line the standard parameter value. For details on the mechanisms see Section 2.2.
Competing interests. The authors declare that they have no conflict of interest.

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References


