

Response and revisions for the manuscript

“A probabilistic assessment of calcium carbonate export and dissolution in the modern ocean ” Biogeosciences Discuss., doi:10.5194/bgd-12-20223-2015

by G. Battaglia, M. Steinacher, and F. Joos, March 2016

We thank the anonymous reviewer and W. Koeve for their constructive reviews and comments. We appreciate the effort and time committed by the reviewers.

We follow reviewer #1's advice and change the evaluation of the Monte Carlo ensemble by applying global skill scores instead of regional, basin-specific skill scores. This adjustment required to re-run the Monte Carlo ensemble and implied changes in illustrations, tables and text. While the results of the new global approach are numerically not identical to that of the regional approach, conclusions remain unchanged from the previously submitted manuscript and numerical results are similar for the two approaches. This agreement further illustrates the robustness of our Monte Carlo approach.

At this occasion, we also corrected a recently found error in the physical core of the model. The evaluation section and the illustrative runs are updated; the corrections are minor and negligible.

We also follow both reviewers' suggestion to improve the structure of the paper and the figure numbering. The figures are now in order of appearance in the text.

We add a revised manuscript with track changes in addition to the answers presented here. The intention is to thoroughly document our changes in response to the review comments. We are aware that the new editorial procedure of BG does not require the submission of a revised manuscript at this stage and that the editor's decision will apply to the previously submitted manuscript.

Please find below our response (normal font) to the comments by the reviewers (*in italic*) and suggested text additions to the manuscript (**in blue**).

Anonymous Referee #1

General comment:

This is a very interesting article concerned with a probabilistic assessment of calcium carbonate export and dissolution. The probabilistic assessment chosen by the authors is very promising and approaches like this are urgently needed in order to arrive at a better model presentation of CaCO_3 . The article is therefore definitely worthy of publication.

Many thanks.

Specific comments:

** The regional skill assessment seems to be problematic. Figure 8 shows clear inconsistencies for*

the median CaCO_3 export, especially when considering the Southern Ocean. There is a visible mismatch between the Atlantic, Indian and Pacific part of the CaCO_3 export of the Southern Ocean, generated by the regional skill assessment. The credibility of the solution is therefore questionable. I advice to shift the focus of this paper from the regional skill assessment to the global skill assessment. This includes a significant re-write and different illustrations.

Done – switched to global skill assessment.

We do not share the reviewer's opinion on the credibility of the regional skill assessment. However, the perception by the reviewer and the general reader is important. The application of global skill scores is easier to explain and offers the advantage of a fully self-consistent solution. We therefore followed the reviewer's advice and present results from a global skill assessment in the revised manuscript. We emphasize that the results from the new global skill assessment are similar to those of the regional approach and that our conclusions remain unchanged. The results appear therefore robust within the model context and only weakly dependent on details of the setup.

A new Monte Carlo ensemble was set up, run, and evaluated. The results from the ensemble simulations presented in the previous manuscript version imply that the application of a uniform rain ratio for the entire Southern Ocean appears overly simplistic. Therefore, the Southern Ocean sectors of the Pacific, Indian, and Atlantic are treated individually in the new setup. In addition, the previous results also imply that the prior ranges for the rain ratio values in the Atlantic regions were very broad. Thus, narrower prior ranges are applied in the Atlantic in the new setup.

The visible step-like changes in CaCO_3 export between the Atlantic and the Indian and Pacific sector of the Southern Ocean with lower export (per unit area) in the Atlantic sector than in the Indian and Pacific sectors also emerge in the new setup. This is attributable to the choice of the regional boundaries and the assumption that the spatial pattern of export within a region is identical to the pattern simulated by the standard version of the model. The standard model yields relatively little zonal variation in the CaCO_3 export fluxes in the Southern Ocean in contrast to the data assimilation with lower than zonally-averaged export in the Atlantic. While our Monte Carlo approach is suitable to estimate export fluxes over larger regions, the detailed spatial patterns in CaCO_3 export remain unconstrained.

The following text is added after the first paragraph of section 4.2:

“Median CaCO_3 export production per unit area in the constrained ensemble is considerably lower in the Atlantic sector of the Southern Ocean ($135 \text{ mmol-C m}^{-2} \text{ yr}^{-1}$) as compared to the Pacific ($301 \text{ mmol-C m}^{-2} \text{ yr}^{-1}$) and Indian ($326 \text{ mmol-C m}^{-2} \text{ yr}^{-1}$) sectors (Fig. 7 top). This is attributable to the choice of the regional boundaries for the rain ratio regions and the assumption that the spatial pattern of export within a region is identical to the pattern simulated by the standard version of the model. The standard model yields relatively little zonal variation in the CaCO_3 export fluxes in the Southern Ocean in contrast to the data assimilation with lower than zonally-averaged export in the Atlantic. This reflects the much lower TA^* reconstructed in the deep Atlantic as compared to the deep Pacific and Indian (Fig. 1). A large export in the Atlantic sector of the Southern Ocean tends to yield high simulated TA^* concentrations in the Antarctic Bottom Water that fills the deep Atlantic. The Monte Carlo data assimilation therefore requires low CaCO_3 export in the Atlantic sector to minimize model-data mismatches in the deep Atlantic. It is difficult to correctly represent water mass formation and circulation in the Southern ocean and our model may be biased. A

known bias is that the Atlantic Bottom Water circulation is too sluggish, also evidenced by simulated low radiocarbon signatures (Figs A2. A4). The influence of a potential bias in South Atlantic export on global CaCO_3 export is estimated to be relatively small; assuming the same (median) CaCO_3 export per unit area in the Atlantic sector as estimated for the Indian sector would yield $0.06 \text{ Gt-C yr}^{-1}$ higher export than suggested by the ensemble median. While our Monte Carlo approach is suitable to estimate export fluxes over larger regions, the detailed spatial patterns in CaCO_3 export remain unconstrained.”

** Throughout the whole paper, there are barely any time periods mentioned. Yet, the knowledge of the considered time periods is of importance, especially when mentioning mean concentrations and when comparing the model results to observations. Please add the considered time periods whenever necessary.*

Done – time periods mentioned.

All ensemble runs are spun up to equilibrium under preindustrial boundary conditions, with atmospheric CO_2 set to 278 ppm and with wind-stress forcing prescribed from a monthly wind-stress climatology (NCEP/NCAR) as mentioned already in the manuscript.

The text at the beginning of section 2 is modified to read:

“Total alkalinity data are from the GLODAP carbon climatology (Key, 2004) and salinity (Antonov et al., 2010), temperature (Locarnini et al., 2010), oxygen and phosphate (Garcia et al., 2010a, 2010b) data from the World Ocean Atlas. These observational data serve to split the alkalinity signal into its different physical and biogeochemical components such as TA^* , our target variable in the data assimilation. The gridded data products from GLODAP and the World Ocean Atlas are derived from samples taken during the previous few decades. “

New text was added at the end of the 1. paragraph of section 3.1:

“We implicitly neglect potential changes in TA^* over the industrial period by comparing model results for preindustrial conditions with TA^* data reconstructed from recent measurements. Such changes are negligible in simulations with prescribed anthropogenic forcing in the Bern3D model.”

** Further, Figure A2 & A3 and page 20244 refer to the year 1994. Here, it would be preferential to at least compare to a ten-year average in order to analyse a more robust model result.*

Done. The model results for CFC11 shown in Figure A3 are now given for the period 1990-1999. This did hardly affect the evaluation as inter-annual variability is low in our model. The radiocarbon results shown in Figure A2 are for the preindustrial period.

In addition the following clarification is added in the appendix:

“The atmospheric history of CFC11 is prescribed according to Bullister 2011. Here we do not account for potential changes in ocean circulation and CFC11 solubility over the industrial period.”

Technical comments:

** The figures and tables seem to appear in random order. Please fix this, so that Figure 1 is the first figure referred to in the text, Figure 2 the second one and so forth.*

Done. Figure and table numbering is adjusted as requested

* page 20224, line 4: Correction: "... CaCO_3 export fluxes ..."

Corrected.

* page 20224, lines 11-17: Please mention at least once that these are simulated results.

Done. Sentence modified to read:

"The median (and 68% confidence interval) of the constrained model ensemble for global biogenic CaCO_3 export is 0.90 (0.72--1.049) Gt PIC/yr .."

* page 20224, line 11: Correction: "... that is within the lower half ... "

* page 20225, line 10: Correction: "... coccolithophorids, which ..."

* page 20226, line 7: Correction: "... range over several orders ..."

* page 20226, line 16: Correction: "... this method ..."

All sentences corrected.

* page 20227, line 27 + page 20228, line 3 & line 25 + page 20229, line 11: Which time- periods and products are these mean concentrations from? Please denote.

Done. Please see answer to the second specific comment given above.

* page 20229, line 19: Correction: "... in some places ..."

Inserted.

* page 20230, lines 21-22: Is this resolution valid for all components of the Bern3D model?

Inserted extra information to clarify:

"Here an **ocean** version with a~horizontal resolution of 41 by 40 grid cells and 32 logarithmically-scaled vertical layers is used (see also Roth and Joos, 2014). **The horizontal resolution is the same for the components atmosphere, ocean, sea ice, and sediments of the Bern3D model.**"

* page 20233, lines 9: Please explain the variable k_{eff} earlier in the text, i.e. directly after equation (4).

* page 20235, line 18: Correction: "... (calculated from GLODAP (Key et al, 2004) and World Ocean Atlas 2009 (WOA09) (Locarnini et al., 2010; Antonov et al.,2010)) ..."

* page 20237, line 19: Correction: "... to the part of ..."

* page 20240, line 26: Is that the export flux at 75m depth? Please specify.

* page 20241, line 27: Correction: "... are associated ..."

* page 20242, line 7: Correction: "... are dissolving ..."

Text corrected and modified as suggested

* page 20245, line 5: What about changes in the TA^* -profiles?

The figure below shows basin-average profiles in TA^* for low, standard and high diapycnal diffusivities and for the three idealized dissolution profiles (fast, slow, constant) discussed in the manuscript.

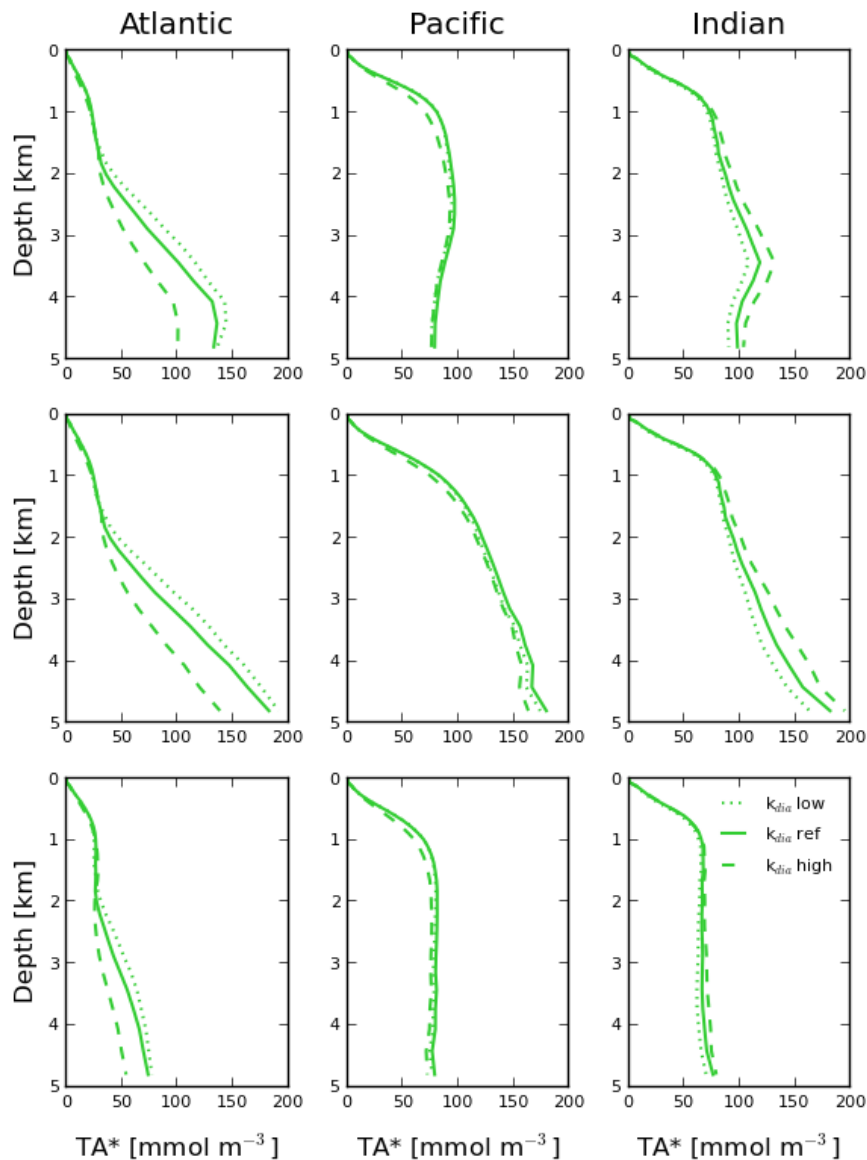


Figure: Simulated TA^* profiles for the Atlantic, Indian, and Pacific and for different diapycnal mixing coefficients and dissolution schemes. For these illustrative simulations, calcite and aragonite particles were assigned equal parameter values and 10% of export is assumed to be in the form of aragonite. k_{dia} is set to 0.1 (low), 0.2 (standard), and 0.5 (high) $10^{-4} \text{ m}^2/\text{s}$. Top row (“fast”): $k_0=10/\text{day}$, $n=1$, $k_{bg}=0$; middle row (“slow”): $k_0=0.16/\text{day}$, $n=2$, $k_{bg}=0$; bottom row (“constant”) : $k_0/v = 1/(2900 \text{ m})$.

The following text is added in the revised manuscript to the second paragraph of section 4.3.2

“Basin-average profiles in TA^* vary little in the upper ocean and the deep Pacific ($< 14 \text{ mmol/m}^3$) and modestly in the deep Atlantic (< 44) and deep Indian (< 18) when varying diapycnal diffusivity between 0.1 and $0.5 \text{ m}^2 \text{ s}^{-1}$... In conclusion, simulated TA^* is only weakly affected by uncertainties in the diapycnal mixing coefficient. “

* page 20246, line 15: Correction: "... observation-based TA* ..."

OK.

* page 20246, lines 15-20: Correlation is just one measure which is not very informative in itself. What about standard deviation and the bias?

We do not agree with the reviewer. Correlation between TA* fields simulated for different dissolution schemes is the metric of choice to illustrate that different dissolution schemes are difficult to distinguish or to constrain by the TA* data. The highly correlated patterns in TA* for the three dissolution profiles indeed imply that the spatial pattern in TA* can hardly be used to discriminate between these schemes given existing uncertainties in the magnitude of CaCO₃ export. The absolute concentration of TA*, and thus mean data-model bias and standard deviation, can be varied, completely independently from the dissolution scheme, by varying CaCO₃ export.

Nevertheless, we computed the standard deviation between normalized model and observational fields. The normalization for an individual TA* field is achieved by dividing the TA* data at each grid cell by the ocean average TA* concentration:

$$TA^*_{norm} = TA^* / TA^*_{average}$$

The RMSE between normalized observed and modeled fields are 58%, 47%, and 42% for the three dissolution cases (fast, slow, constant).

To make this point clear, the text was changed to read:

“Corresponding to the high correlation, the RMSE between normalized observed and normalized modelled fields vary within a limited range (42 to 58%) for the three dissolution cases. This high spatial correlation in simulated TA* **and uncertainties in CaCO₃ export** make it difficult to distinguish different dissolution parameterizations. **The magnitude of CaCO₃ exports modulates absolute TA* concentrations and thus model-data bias and root mean square errors.** Given these uncertainties, we cannot objectively determine the preferred dissolution scheme from TA* data.”

* page 20247, line 23: Correction: "... a global CaCO₃ export ..."

Corrected.

* page 20247, lines 7-11: What are the error-estimates of Sarmiento and Gruber (2006) and Lee (2001)?

Sarmiento and Gruber (2006) do not provide a quantitative error assessment. The uncertainty estimate by Lee 2000 of **+/- 0.3** Gt-C yr⁻¹ is now given in the text.

* Table 2: Please refer to the depth of the export flux.

* Table 5: Correction: "... are estimated ...".

Done.

* Figure 1: The bottom row should rather be displayed in a separate figure.

Agreed. The color bar of the lower panel is now adjusted to better visualize the influence of ocean-sediment fluxes on simulated TA*.

* Figure 2 & 4: Please enlarge the font - the labelling of the axes is not readable.

Done.

** Figure 3: What time period is depicted? Further, the western/eastern boundaries of the arctic part of North Pacific and North Atlantic are missing.*

Figure 3 is removed from the revised manuscript. It also showed results for a pre-industrial steady state.

** Figure 7, 8, A1, A2: What time period is depicted?*

Intro of Appendix now extended to:

„Results are for a **pre-industrial** steady state of the Bern3D ocean model configuration **The atmospheric history of CFC11 is prescribed according to Bullister, 2011. Here we do not account for potential changes in ocean circulation and CFC11 solubility over the industrial period.**”

** Figure A1: The references for the World Ocean Atlas should be bracketed.*

Done.

Review by W. Koeve (Referee)

The CaCO_3 cycle is an important component of the oceanic carbon cycle. Modelling and model evaluation of this component, however, appear to be less well developed, compared to that of the so called organic tissue pump. The manuscript of Battaglia and co-authors applies a probabilistic approach using the Bern3D ocean model to assess export and dissolution of calcium carbonate on global and basin scales and in the modern ocean. It is one of the most detailed studies on this subject, that the reviewer has seen. I regard it an important contribution to our quantitative understanding of the oceanic carbon cycle, its modelling and hence forecast.

The study provides new estimates of CaCO_3 export and the large scale distribution of its dissolution, both with respect to the three major oceans, as well as to the three principal domains where dissolution might occur (in/on top of the sediment, below the saturation horizon, and above it).

A major result of this study is that the kind of observations (alkalinity, CaCO_3 deep ocean flux data) and tracers constructed from them (TA^) that are currently available do not allow to constrain at the same time CaCO_3 export and the functional form of its dissolution in the ocean. In particular, significant dissolution above the saturation horizon can not be ruled out based on the specific combination of data and the approach and model chosen. "Dissolution schemes, with and without dissolution above the saturation horizon, achieve realistic (i.e. observed) TA^* distributions". Ocean circulation, including mixing, affect concentration gradients such that different dissolution schemes can't be distinguished statistically.*

Thank you.

I suggest publication of this work in BG, however, after moderate revisions of the ms. Important issues:

Structure: In particular, the organisation of the paper and of the arguments put forward lacks clarity. In fact, I got the strong impression that the authors have reorganised their paper very recently, but without making sure that the order of arguments supports reading and understanding of the presented material as good as it could. This reorganisation is very obvious from the order of references to several figures which are misplaced to a degree I have not seen before. For example, Fig 2 is referred to three text pages after Fig. 3, Fig. 4 is referred to 2 pages after first referral to Fig. 5. Finally Fig. 6 is referred to on p20245 for the first time, long after Figs 7, 8, and 9 have been discussed on the four pages before. This is clearly not a good practice and very confusing for the reader. Please improve this.

Done. Figure numbering is adjusted to reflect first appearance of figures in the text.

TA^ -CFC method: In the abstract, introduction and discussion you make reference to 'the TA^* -CFC-age' method, which has been used by others (e.g. Feely et al.) in support of shallow (above saturation horizon) dissolution of CaCO_3 . The way you present this material is not really appropriate. Since the paper of Friis et al. 2006 it is well acknowledged that TA^* as a tracer being produced in the interior of the ocean and being destroyed only at the very surface (either explicitly as in Friis et al. or Koeve et al. or implicitly when TA^* is diagnosed or 'constructed'). Such a tracer has to show gradients towards the surface, and significant concentrations far away from the site of TA^* generation. These gradients, if not properly corrected for the effect of mixing will give rise to artificial rate estimates when combined with age tracers. The Friis paper is well received by the*

community, hence the TA-CFC-age based estimates are more a historical note, and not a state-of-the-art rate estimate. I would hence not consider the difference between rates estimates from this method and yours to be a surprising/important result worth to be mentioned in the abstract or the introduction. This avoids the problem of introducing this method in more detail there (you can't assume that everybody knows the TA*-CFC-method!). I therefore suggest to discuss the older TA*-CFC-age based estimates briefly in the discussion only. By the way, TA* tracer mixing is only one of several issues with this approach.*

We followed the advice by the reviewer and de-emphasized the text on the TA*-CFC method. Nevertheless, this method appears still to play some role in the general discussion. The widely cited Berelson et al, 2007 review relies on results obtained with the TA*-CFC method and even more recently Carter et al., 2014 appear to argue for this method and Barrett et al., 2014, included estimates based on this method for comparison, though with a cautionary note.

We removed text referring to TA*-CFC method from the abstract. In the introduction, the reference to this method is now extremely brief to establish the general context and current uncertainties. The following text was removed from the introduction: “In particular, this adds to the discussion initiated in Friis et al., 2006 on the difficulty to uniquely relate upper ocean TA* concentrations to either dissolution or mixing processes.”

CO₃-ions: *In the introduction you speak about carbonate ions in a sometimes strange, awkward or wrong way. Eg. 20225, 11 you say that CaCO₃ is formed from Ca²⁺ and CO₃²⁻. To the extent that the actual carbon species used in the formation of CaCO₃ is known it appears to be different for different CaCO₃ producers. Even if 1 mol of CO₃²⁻ would be taken up per mol of CaCO₃ formed (and exported), the net effect would not be a decrease of 1 mol of CO₃²⁻ in the surface ocean. This is simply due to rapid re-adjustments within the carbonate system. The actual rate of change per mol of CaCO₃ depending on conditions. Also, 20225, 14-15, the 'uptake and release of CO₃²⁻ introduces vertical gradients in alkalinity and DIC' is misleading, at least. 20226, 27: 'mixing of CO₃²⁻' is awkward. CO₃²⁻ is NOT a tracer, hence it is not mixed conservatively. Please consult e.g. Wolf-Gladrow et al., 2007, Mar. Chem., 106:287ff for details. Or use a tool like co2sys to take a look at the effect of mixing of e.g. two water masses with distinct TA and DIC for the effect on CO₃²⁻ concentrations along the mixing line. Please carefully revise the text accordingly.*

Thank you for pointing out this misleading wording. Text adjusted to:

“The CaCO₃ cycle is driven by calcifying organisms such as coccolithophorids, foraminifera or pteropods, which remove calcium Ca²⁺, **alkalinity and dissolved inorganic carbon** from the pelagic surface ocean waters to form shells and structures of CaCO₃.”

“The **formation and dissolution of** CaCO₃ introduces a vertical gradient in alkalinity and dissolved inorganic carbon.”

“... physical transport and mixing of the **released alkalinity and dissolved inorganic carbon** and ocean-sediment fluxes.”

Methodology: *Latin-hypercube, the specific way in which skill scores are assigned and used when computing e.g. medians could at times be more specifically presented and/or supported by references. Having not done this myself (like most of the readers of BG), sometimes had to guess what you actually did. From the list of possible journals within the special issue you chose BG instead of GMD. Hence I think you should consider that the audience of BG consists of many non-modellers. Also your work should be understandable to people that address the CaCO₃ cycle from the*

observational side in order to make your work most influential. Following to my argument about the organization of the paper, I sometimes had the impression that some details of your approach become clear late in the paper while the respective Fig. numbers indicate that that section may have been moved towards the back of the paper recently. In practice, section 3.3.2 could be extended providing more details on how eg. medians shown in Fig. 5 are computed. It is particularly in section 4.1 where I sometimes missed details of the approach, e.g. when you speak about 'the optimisation procedure with the application of regional skill scores' (20239, 19) or the 'median fields' (20240, 3) shown in Fig. 4. How are the latter actually computed. Potentially a reader (me) would better understand all this if s(he) reads Steinacher et al. carefully first, but then s(he) may not return to continue reading your paper. Which I would regard a pity.

The revised text is hopefully easier to understand as we omit the regional skill score approach and present a global skill assessment following the request of reviewer 1. The text on skill scores in section 3.3.2 is expanded and the computation of median and similar measures is explained. The new text reads:

“The skill scores S_m of the individual ensemble members are likelihood-type functions corresponding to a Gaussian distribution of the data-model discrepancy ($TA^*_{\text{model}} - TA^*_{\text{obs}} - TA^*_{\text{sedcorr}}$) with zero mean and variance σ^2 . S_m is an indication of the relative performance/credibility of each individual model configuration. Configurations which have relatively small deviations from the data are judged more probable than configurations which differ greatly from the observations.

S_m are used as weight to compute probability density functions (PDFs) and related measures such as the median (50th percentile) and the 16th and 84th percentiles defining the one standard deviation confidence interval (1σ) of the ensemble results. PDFs represent weighted and normalized histograms of the variables of interest. The normalization is such that the integral over a PDF equals 1. A cubic spline interpolation is used to arrive at a continuous PDF from the discrete, normalized histogram. For the computation of median and confidence ranges the histograms are converted to cumulative distribution functions (CDFs). We interpolate linearly within the discrete CDFs to arrive at the chosen percentiles (i.e. $\text{CDF}=(0.16, 0.5, 0.68)$).

The above explanations apply for any simulated quantity of interest. In the following we will present PDFs, median values, and 1σ confidence ranges for aragonite and calcite export and dissolution as well as for tracer concentrations at individual grid cells or integrated over regions or the whole ocean. Spatial integrations are done for each ensemble member individually and before computing the PDFs and associated measures from the full ensemble.”

Text on the Monte Carlo Ensemble in section 3.3.1 is also extended as explained under specific comments.

Prescribing the saturation state from observations: This is potentially a (severe?) limitation of this study. The actual saturation state is due to the pressure effect, the pre-formed DIC and TA, the remineralised DIC and TA and the imprint of CaCO_3 dissolution (in terms of DIC and TA). To the extend that CaCO_3 dissolution is saturation dependent, the former feeds back on its conditions in the real ocean, but not in your modelling. I suggest to discuss, and potentially quantify in sensitivity runs, how significant that feed-back is for TA^ profiles and inventories in the non-constant cases.*

We do not agree with the reviewer that the application of observation-derived fields of the saturation of water with respect to CaCO_3 , Ω , is a limitation of our study. To the contrary, the observation-derived Ω -field provides the best possible boundary condition for the dissolution model in our approach. It is a strength rather than a weakness of the assessment.

As explained by the reviewer, the saturation of water with respect to CaCO_3 , Ω , is affected both by the dissolution of organic matter and CaCO_3 . The simulated saturation horizon and Ω -field is thus not a direct and simple reflection of the CaCO_3 feedback on these quantities. The magnitude of the mentioned CaCO_3 dissolution feedback and its influence on Ω and the saturation horizon is fully constrained by TA^* . TA^* is the quantity that reflects the dissolution of CaCO_3 below the euphotic zone and defines its imprint on the fields of DIC and Alk, variables which in turn define (together with pressure, temperature, salinity and nutrient concentrations) Ω . This is why we use TA^* as our target variable and not Ω and the saturation horizon. If TA^* is modelled in agreement with observations, then the imprint of CaCO_3 dissolution on DIC, Alk and thus on Ω and on the saturation horizon is correctly modelled and the mentioned feedback is correctly represented.

Constraining the saturation horizon and the Ω -field would require not only to constrain CaCO_3 and dissolution, but also to constrain export and remineralization of organic matter in the form of particulate and dissolved organic matter. This would add another level of complexity and many free parameters in the assimilation. This is beyond the scope of this study.

In addition, the results imply that the probabilistic estimates of CaCO_3 export and dissolution do not depend sensitively on the exact location of the saturation horizon. As discussed in the text, very similar patterns of the target variable TA^* emerge for dissolution parameterizations that do/do not depend on Ω and on the saturation horizon. We note that the saturation horizon is reasonably well represented by the standard setup of the Bern3D model in the Atlantic, Indian, and South Pacific, and is too deep in the North Pacific.

The following text is added at the end of 3.3.1:

“... Mismatches in modelled and observed saturation states are particularly large in the **North Pacific**, where the modelled **calcite** saturation horizon is up to 1.5 km too deep. **The calcite saturation horizon is well represented in the South Pacific, Indian and Atlantic by the model. The results presented in section 4 suggest that estimated CaCO_3 export production fields and dissolution rates are insensitive to the choice of the saturation field, because saturation-dependent and saturation independent parameterizations of dissolution yield similar TA^* fields.**”

Units: In 20229, 22-25 you say that you will present TA^ in TA-units, but later you give TA^* inventories in Pmol C . I suggest that, for the purpose of your paper, it would be least confusing for the reader if you always use mol C based units (for local TA^* , its inventories, and ALSO the CaCO_3 flux values, which are currently in Gt PIC ; Gt PIC-C , or PIC-CaCO_3 is not specified). Currently you use three different sets of units, which is not helpful for the reader.*

Done. We specify that TA^* concentrations are always in alk-equivalents and add that TA^* inventories are in Pmol C , in line with Koeve et al. 2014.

For CaCO_3 fluxes we used Gt PIC yr^{-1} for comparison with Berelson et al. 2007. We change the notation to **Gt-C yr^{-1}** , **$\text{mmol-C m}^{-3} \text{yr}^{-1}$** , **$\text{mmol-C m}^{-2} \text{yr}^{-1}$** to avoid any ambiguities.

Added: “**TA* inventories are in P mol- C and CaCO₃ fluxes are given in carbon units (Gt-C yr⁻¹ , mmol-C m⁻³ yr⁻¹ , mmol-C m⁻² yr⁻¹).**”

Below I provide some more specific comments and suggestions, ordered largely by appearance in the ms;

20225, 10: name other CaCO₃ producers, please

Added: “calcifying organisms such as coccolithophorids, **foraminifera or pteropods,**”

20225, 17: *'This redistribution' refers to mixing and transport in the sentence before, right? But isn't the CaCO₃ counter pump, which makes up the gradients (l 15) what in particular affects the ocean-atmosphere partitioning of CO₂? The redistribution tends to reduce that effect. Rephrase please.*

Sentence modified to read:

“This redistribution of alkalinity and carbon **by biogenic and physical transport** affects the partitioning of carbon between the ocean and atmosphere ...”

20225, 20: *50ppm on century times scales appears to be a lot. I think, that that is not in agreement with e.g. Hofman and Schellnhuber, 2009 (PNAS, I think).*

Gangsto et al., BG, 2011 find a decrease of 30 ppm by year 2100 and of 50 ppm by year 2300 in response to a hypothetical stop of CaCO₃ export after 2000 CE (their Figure 15b). Text changed to read: “.. tend to decrease atmospheric CO₂ by **a few ppm, and in the extreme case by up to ~ 50 ppm,** on century time scales“

20226, 15: *'the TA*-CFC age method'; you can't assume everybody to know what that is; either you introduce it here, or (better) leave it out; much of the issues raised by the TA*-CFC-age approach disappears when understand the general nature of the TA* tracer; however, that one (TA*) has not even been introduced at this point to the reader; please do so at least in a generic sense (e.g. TA* to reflect the imprint of CaCO₃ dissolution on TA) and rewrite the paragraph accordingly; perhaps point to section 2 for details)*

OK. Rephrased to:

“Friis et al. 2006 nevertheless, demonstrated that **the method** which is often employed to derive these upper ocean dissolution rates (**see Discussion section on TA*-CFC age method**) (Berelson et al., 2007), might not be applicable, ..”

20226, 20-21: *perhaps add a reference here*

The following references are added at the end of line 21: **Archer et al.,1996**

20227, 27 & 20228, 3 & 20228, 25: *at this point it is not clear what the mean values are referring to, i.e. which dataset (GLODAP, WOA)?*

True. Added introductory sentence. See reviewer #1.

20228, 12: *'observations of TA', it is not clear whether you did this with observed TA (bottle data e.g. from WAVES) or gridded GLODAPv1 or regridded GLODAP to your Bern3D grid. Please*

specify.

Added to first paragraph:

“... We first regridded all required, gridded datasets to the Bern3D model grid (40x41x32 grid boxes) using the area-weighted regridding method of Ferret before deriving the other properties.”

20229, 12: *refer to Fig. 1a, not just Fig. 1*

Yes. Figure 1 a, b are now two separate figures (see request of reviewer 1).

20229, 20: *Is the fraction of TA* above the saturation horizon similar to that reported in Koeve et al., 2014?, Compare, please.*

Slightly higher there, at 44.7%. Added:

“..similar to Koeve et al., 2014 who found 44.7% above the calcite saturation horizon (Koeve et al., 2014.”

20229, 22-25: *TA-units. Here, like in much of section 2 your description follows that of Koeve et al. 2014 to a certain extend. However, later in the text you don't follow your own advise. TA* inventories are given in Pmol C, not Pmol TA. See my general comment above for consolidating the units.*

We follow the units used in Koeve et al. 2014.

20230, 6: *Latin-Hypercube. Since you don't introduce this method in too much detail, perhaps give at least some references pointing to papers that present more details, if possible from ocean studies, helpful to readers from that domain.*

Inserted:

“varied using a Monte Carlo sampling method (McKay, 1979, Steinacher et al., 2013)”

Added to section 3.3.1

“Following Steinacher et al., 2013 and 2016, we run a 1000 member Latin Hypercube ensemble to constrain the export flux out of the surface ocean, and the dissolution of aragonite and calcite within the water column. Latin Hypercube sampling (McKay et al. 1979) is a statistical, Monte Carlo method to generate controlled random samples from a multidimensional distribution (15 dimensions in our case). The defined parameter ranges are divided into equally probable intervals (1000 in our case). Random samples are then generated in each interval. This method ensures that the sampled values are representative of the real variability while minimizing the number of required samples and thus the computational costs. We sample 15 parameters ...”

Changed Latin-hypercube to **Monte Carlo** in many instances: Perhaps easier to associate with.

20231, 2 : *'scaling-factor', as used here is lab slang. Please be more specific.*

Changed to:

“The global mean air-sea transfer rate is reduced by 19% compared to OCMIP-2 to match observation-based estimates of natural and bomb-produced radiocarbon (Müller et al., 2008).”

20231, 3: 'describes'? 'represents'

Ok. Changed.

20231, 16: 'six regions (Fig. 3).,': *The regions are not really visible from that figure. This confusion is amplified by the fact that the text speaks about different values of the rain ratio while the caption of Fig. 3 talks about a constant value for the explicit experiment shown in the figure. I suggest to add a map in which the six regions are colour coded. This figure can be presented in the appendix. Fig. 3 should not be referred to here.*

The original Fig. 3 has become obsolete as we no longer discuss regional skill scores

20231, 23-29: *please give Friis et al. and Koeve et al. as references for the explicit TA* tracer*

Added reference to **Koeve et al. 2014**. Friis et al. 2006 did not explicitly include TA*, but TA0.

20232, 5-10: *perhaps mention explicitly that TA* of the deepest wet box is affected/modified accordingly (from sediment fluxes)*

True. Added:

“Simulated TA* concentrations tend to be lower with the sediment module enabled than without sediment module, because a fraction of the CaCO₃ export flux is removed from the ocean and buried in the geosphere.”

20232, 16, 'thereafter' ? *is there any thereafter? I though all your runs are spin-up runs. Delete the phrase 'and kept constant thereafter'.*

True. Deleted.

20233, 3: *(here and elsewhere) 'Gangstoeet' al misses a blank before 'et'*

Thanks.

20234, 22: *Ref to Tab. 4. Looking at the table I don't understand k_{dia} since that is introduced much later in the text. I suggest to improve the table caption accordingly.*

Inserted:

“CaCO₃ export and TA* inventories for different physical mixing (**diapycnal mixing coefficient, k_{dia}** and ...”

20234, 23 *(and elsewhere) a ratio is not to be given in % (but here in mol/mol) Also: I suggest to always be explicit 'molar CaCO₃/POC export rain ratio' instead of just 'rain ratio'. Rain ratio may be easily mixed up with rain rate, which is POC flux to the sediment. It could also be the POC/PON rain ratio, or whatever. Avoid ambiguities.*

Inserted:

“We define eight such regions each assigned an independent value for the **export** rain ratio parameter (**mol inorganic carbon / mol organic carbon exported, later given in % inorganic to**

organic carbon exported). “

20234, 24-25: *Units, see general remark above*

We follow the units used in Koeve et al. 2014.

20235, 11: *f_{calc} is a fraction, but is given in %, correct please (see above)*

Corrected.

20237, 15: *change title to 'A first order correction to CaCO₃ burial' since that is the important aspect*

Here, we intend to stress that it is about TA* fluxes. Modified to read:

“A~first order correction for ocean-sediment **TA*** fluxes ..”

20237, 20-21: *isn't this in conflict with 20238, 12-13? '25%' is not equivalent to 'largely unchanged'; also the phrase on 20237 is presenting some results in the mid of the methods presentation. Improve please.*

No. Largely unchanged on 20237, 20-21 refers to the fluxes of CaCO₃ export and of CaCO₃ dissolution within the water column. Text on 20238, 12-13 refers to the TA* inventory. This is affected as CaCO₃ burial removes the corresponding alk-equivalents.

The results discussed in this section refer to the application of a sediment correction and do not represent a main result such as estimated export and dissolution fields and we prefer to keep the text as is.

20238, 8: *'sediment BURIAL correction'*

Inserted.

20239, 8, *'by definition' this is explicitly true for your model TA* tracer, but not 'by definition' for the reconstructed (or diagnosed) TA*; you mentioned some non-zero (negative) values yourself, likely in waters close to outcrops or at the surface*

Adjusted text to: “**close to** zero”

20239, 11, *'age of water' this is very implicitly known here only; you could refer to A2, though that is $\delta^{14}C$ which is not age (see e.g. Koeve et al., 2015, GMD, and refs therein)*

The sentence is intended to provide a qualitative explanation for the increase in TA* from the deep Atlantic to the deep Pacific.

Added:

“... increasing with the age of water masses (**see Fig. A2 showing the distribution of $\Delta^{14}C$, a proxy for water mass age**).

20241, 3: *Fig. 7: I don't understand how the black line in Fig. 7 reflects the sums of the coloured lines ('the sums of regional PDFs', line 4).*

In the previous regional skill assessment, the regional probability density functions (the coloured lines) are added to obtain the PDF for the global ocean (black line) by convolution

(<https://en.wikipedia.org/wiki/Convolution>). In the revised manuscript, we present a global skill-score approach and the convolution of regional results is not needed anymore. The computation of PDFs is now explained in section 3.3.2.

20141, Fig. 8: *SO export fluxes show strong discontinuities when moving from PAC to ATL and from ATL to IND. That points to some limitation of your realisation of a regional approach (e.g. not considering the SO as an explicit region). This should be discussed.*

This concerns the main issue raised by reviewer # 1. Please see our response to reviewer 1.

20241, 18-20: *treatment of deepest wet box in no-sediment runs needs to be presented earlier, i.e. section 3*

Done. Section 3.1 extended to read:

“In sensitivity simulations with the sediment module enabled, the flux of CaCO_3 , and of other particles, reaching the sea floor is passed to the sediment module from where a α -fraction potentially re-dissolves back into the water column. **In simulations without the sediment module, the entire flux reaching the ocean floor re-dissolves back into the water column.**”

20242, 6-14: *I wonder how sensitive that is to the aspect of having no feedback from CaCO_3 dissolution on the imposed omega field*

Please see the response above.

20244, 3, k_{dia} . *This is one of the details I missed earlier in the paper.*

It is now explicitly mentioned in the Introduction that uncertainties in k_{dia} are discussed in Section 4.

20244: *are global mean $\delta\text{ell}14\text{C}$ values meaningful? Koeve et al., 2015 GMD showed that different ocean models which applied the same 'scaling factor' may have quite different preformed $\delta\text{ell}14\text{C}$; hence the global mean $\delta\text{ell}14\text{C}$ is not a good measure of the mean age; this is likely the case also for one model with different k_{dia} realisations, I suspect; please discuss*

The simulated global mean preindustrial $\Delta^{14}\text{C}$ signature of DIC is a meaningful first-order measure to indicate model performance with respect to surface-to-deep transport and water mass age in our model. On basin average, simulated surface $\Delta^{14}\text{C}$ signatures are close to observation-inferred signatures. In addition, surface values vary relatively little with the applied changes in diapycnal diffusivity. Thus, $\Delta^{14}\text{C}$ difference in the thermocline and the deep ocean and mean ocean $\Delta^{14}\text{C}$ reflect changes in the surface-to-deep exchange time scales. In other words, and as illustrated in Figure A4, surface-to-deep $\Delta^{14}\text{C}$ and thus the surface-to-deep age difference is low for high diapycnal mixing (strong overturning) and low for low diapycnal mixing.

The following text is added in the first paragraph of section 4.3.3 (now section 4.3.2):

“Simulated surface-to-deep $\Delta^{14}\text{C}$ gradients are too low (high) relative to the observed gradients for the high (low) diapycnal diffusivity parameter, thereby indicating too fast (slow) surface-to-deep water exchange (Fig. A4).”

20245, 26-28: *This is implicit only in the comparison of upper and lower panels of Fig. 6. Could you prepare a figure to make that explicit, please? Also, I think Fig. 6 should be shown much earlier in the paper. It could help to understand Figs. 7-9 which you discuss before Fig. 6.*

We prefer not to add additional figures as the number of figures is already large. Text added such that it is clear that this comparison is necessary.

20246, 5-9: *One of the CaCO₃ related papers by Andy Ridgwell should be discussed here. Didn't that show also that parameter values could not be constrained well/independently from TA data, if I recall correctly.*

The following text was added at the end of the second paragraph of section 5.1:

“In addition, Ridgwell et al., 2007, used an Ensemble Kalman Filter approach to assimilate total alkalinity and phosphate data into their model and determined a global CaCO₃ export flux of 1.2 Gt-C y⁻¹. “

And at the end of section 5.4:

“This approach is used in previous studies (Archer et al., 1994, Ridgwell et al., 2007).”

20246, Section 4.5. *You use flux data instead of TA* as a constraint. Is it possible to combine both? Would that be a better constraint?*

Ideally, multiple constraints would have a narrow and overlapping confidence interval, such that only a small interval fulfills both constraints and is therefore more probable. Here, the confidence interval arising from the TA* data constraint is much smaller and well within the confidence interval obtained with the flux measurements. In this sense, flux measurements do not point to a more unique optimal solution.

20247: *You discuss the TA*-CFC-age method before introducing it. I suggest to reorganise the text accordingly.*

A reference to the TA*-CFC age method is needed here to explain the high export values reported by Berelson et al., 2007. The shortcomings of the method are, however, better explained in the context of upper ocean CaCO₃ dissolution.

The text has been modified to read:

“This estimate is based on sediment trap data and other information constraining the flux to the deep ocean (> 2000 m to 0.6+/-0.4 Gt-C yr⁻¹) **and results obtained with the so-called TA*-CFC age method** suggesting an upper ocean dissolution of 1 Gt-C yr⁻¹. **The TA*-CFC age method is heavily criticized by Friis et al., 2006 and tends to bias estimates systematically towards high values (Friis et al., 2006). This method and its shortcomings are further discussed in the next section on upper ocean dissolution.** While our estimated flux to the deep ocean of 0.52 (0.43-0.61) Gt-C yr⁻¹ is roughly consistent with the budget of Berelson et al., 2007, their export estimate, and upper ocean dissolution, is in clear conflict with our results and those of other studies (Lee, 2001, Sarmiento et al., 2006, Jin et al., 2006) that apply a range of different methodologies. We attribute this mismatch to deficiencies in the TA*-CFC age method, **implying that the export estimate by Berelson et al., 2007 is biased high.”**

20249, Fig. 10 *very nicely reflects the current status of our quantitative understanding of CaCO₃ export. This is very helpful.*

Thank you.

20249, 26, lab slang again: *'the TA*-CFC age method' needs to be introduced before being*

referred to that often; 20249-20250 (Section 5.2) Please rewrite this paragraph. First explain how the TA-CFC method is supposed to work, thereafter its by now well know caveats. Overall there is too much talking about estimates that have been disqualified 10 years ago by Friis et al.. But what about the work of Barrett, you cite that paper earlier, but do not refer much to the non-TA* related details presented there. Do the same arguments apply?*

The caveat and the study by Friis et al. may not be known to all readers as suggested by the reviewer. As the method is still applied in a recent study (Barrett et al., 2014) and used to derive the widely cited estimates by Berelson et al., 2007 it seems appropriate to discuss this method in the context of our findings.

The references to the TA*-CFC method are now deleted in the first paragraph of section 5.2, except in the last sentence where it is used to introduce the following paragraph on the method. The text of the first two paragraphs of section 5.2 is modified and the description of the TA*-CFC age method is now extended to read:

“.. As mentioned above, we link **the differences between the estimates of Berelson et al. 2007 and this study** to methodological problems (Friis et al., 2006) associated with the TA*-CFC age method **that very likely introduce a bias in the results of Berelson et al., 2007.**

The TA*-CFC age method relies on deduced, observation-derived TA* concentrations and estimates of water mass age, typically derived from measurements of chlorofluorocarbons (CFCs) and their known atmospheric history. **TA* concentrations are plotted against their CFC-age and a line is fitted to this data.** The higher the TA* concentration for a given water mass, the more TA* must have been added by dissolution to this particular water parcel **according to this method.** The slope of the relationship between TA* and age **is in this sense** the CaCO₃ dissolution rate (**mol/(volume x time)**).”

Barrett et al. 2014 present estimates of CaCO₃ dissolution in the the top 1000 m of the North Atlantic along the A16N transect. These authors suggest upper ocean dissolution of CaCO₃ by biologically-mediated mechanisms based on the measured decrease of CaCO₃ particles with depth and an application of the TA*-CFC age method. The following text is added in the first paragraph of section 5.2:

“High dissolution rates in the range of ~ 0.1 to 0.4 mmol m⁻³ yr⁻¹ are estimated by Barrett et al., 2014 for a transect in the upper tropical and northern North Atlantic. These values are much larger than our estimates of order 0.01 mmol m⁻³ yr⁻¹ for the upper tropical and northern Atlantic. These high estimates are based on the measured decrease of suspended CaCO₃ particles with depth multiplied with a CaCO₃ particle settling velocity of 80 m day⁻¹. These estimates may be affected by uncertainties in the assumed particle settling velocity. CaCO₃ particle settling velocities are reported by Jansen et al., 2002, to vary greatly (0.15 to 3440 m day⁻¹) and to be typically order one m day⁻¹ for coccolitophorides and several 100 m day⁻¹ for foraminifera and pteropods. Our dissolution rates would be consistent with the measured depth gradient in suspended biogenic CaCO₃ particles for an average settling velocity of a few m day⁻¹.”

Section 5.3: For the relative importance of shelf vs. open ocean CaCO₃ production, perhaps refer to Milliman or some other global CaCO₃ production review that considers non-open ocean realms.

The following references are added at the end of section 5.3:

Milliman, GBC, 1993, Vecsei and Berger, GBC, 2004

Section 5.4: I see the advantage applying the the 'constant' dissolution formulation to e.g. climate models, but are there potential downsides of your suggestion? Discuss please.

Text added at the end of the paragraph:

“A shortcoming of the application of an exponential particle flux profile for CaCO_3 is that it is not easy to account for the potential influence on dissolution of changes in environmental variables, including a decrease in saturation state as expected under ongoing ocean acidification, or in the the quality, form and size distribution of exported CaCO_3 particles.”

Section 6: Perhaps you should at least mention the limitation of your study, which I argued about above: i.e. that you prescribed the omega distribution from observations and CaCO_3 dissolution can not feed back on its conditions.

Please see our response above. First paragraph extended with:

“The saturation state of water with respect to calcite and aragonite is prescribed using observational estimates to provide realistic boundary conditions for the CaCO_3 dissolution parameterization.”

Overall, a very interesting work, which I was very happy to read and review - though it was not always easy. Looking forward to see an improved version being published.

Many thanks for your helpful comments.