Climate of the Past Discussions

Clim. Past Discuss., author comment AC1
https://doi.org/10.5194/cp-2021-76-AC1, 2021

Reply on RC1

Camille Godbillot et al.

Author comment on "Parallel between the isotopic composition of coccolith calcite and carbon levels across Termination II: developing a new paleo-CO_2 probe" by Camille Godbillot et al., Clim. Past Discuss., https://doi.org/10.5194/cp-2021-76-AC1, 2021

Thank you for accepting to review the manuscript, and for your positive assessment of our work and your constructive comments on it. We've listed the answers to your comments below:

I have two concerns / suggestions I would like the authors to consider and one request.

First, the framing of the paper, from the title and abstract, is about "developing a new paleo- CO_2 probe" (…). Although they do have some good proxy records to compare against their data, it leaves open a question of whether "mismatches" (assuming there should be a relationship) between $[CO_2]_{aq}$ and large-small vital effects are due to physiological or oceanographic confounding effects, or both. This is a problem if the paper is setting out to make a solid contribution towards a new pCO_2 proxy (no major step forward in quantifying a robust pCO_2 - coccolith isotope relationship), but is less so if just trying to gather good data and understand how these size-specific coccolith isotope records actually behave in practice and especially across a range of temporal scales and magnitudes of pCO_2 change (…). So, I would strongly recommend reorienting the paper towards the best interpretation of the data you have rather than trying to reach for a CO_2 proxy which isn't there (yet).

We understand this remark and fully acknowledge that taken alone, this manuscript does not offer a ready-to-go novel pCO_2 proxy. We have probably not made sufficiently clear in our introduction that this manuscript is part of a series of papers which collectively support that the coccolith vital effects convey a pCO_2 signal. As such, we (and other teams) have tried to develop this exciting research avenue for several years now. This is something that we could better introduce in a revised manuscript, should we be invited to submit one by the Handling Editor.

Concerning the paper's orientation, as we highlighted in the discussion, the direct comparison of fossil coccolith $\delta^{13}C_{small}$- $\delta^{13}C_{large}$ and a theoretical CO_2 concentration would greatly benefit from better constraints on both the productivity changes and on the evolution of the chemical disequilibrium at the air-sea interface in the mid-latitude North Atlantic during Termination II. Although our results indicate that $[CO_2]$ might exert a first order control on coccolith differential vital effects, we agree that better knowledge is needed before we can (collectively) come up with a more robust transfer function between these two parameters.

In this regard:

- Although we would be happy to leave the current title as is, as we feel it is rather objective, we would be also happy alter it for "Parallel between the isotopic composition of coccolith calcite and carbon levels across Termination II:

  Is there a CO_2 signal in the magnitude of the vital effects?"– this is dependent of Reviewer 1 and the Editor's advice.

- In the revised manuscript, we will modify the parts of the abstract and of the conclusion that probably oversold the transfer function and will focus instead on how this study fits in with previously obtained data on the tight link between the coccolith vital effect and CO_2 levels.

Second point, and related to the first, is that I'd like a more detailed consideration of the underlying driver of your change in the large-small coccolith isotope offsets, especially for carbon. The raw data - Figure 2 - shows a ~1‰ negative shift in small coccolith $\delta^{13}C$ across Termination II whereas the large fraction hardly changes. The *G. bulloides* record trends slightly positive. In this instance, I can't see how this can be explained other than that the vital effects in the small cell sizes are increasing across TII - i.e. going further from equilibrium calcite - as CO_2 rises. Whereas the large cell sizes change less. OK, this gives you a reduction in large-to-small vital effects, as you'd expect with increasing CO_2, BUT it's the small coccolithophores that are driving this change, not the large cell sizes that should be most limited and sensitive to changing $[CO_2]_{aq}$. With no major change in local $\delta^{13}C_{DIC}$ (based on *G. bulloides*) how do you account for this big shift in the small coccolith $\delta^{13}C$? This seems to me to be at the heart of understanding this record but is never really addressed.

The main difficulty in dealing with an offset is indeed to determine which of the large $\delta^{13}C$ or the small $\delta^{13}C$ (or both) changed throughout the interval. We agree that a clear understanding of what drives the isotopic evolution of the size fractions taken separately would massively contribute to our understanding of the evolution of the small-large coccolith offset across the interval of study. In culture experiments, increased photosynthetic activity under increasing $[CO_2]$ concentrations tends to build a ^{13}C-enriched internal carbon pool of large cells, which leads to higher $\delta^{13}C$ in large coccoliths. This biogeochemical phenomenon reduces the isotopic offset of $\delta^{13}C_{large}$ *both* with inorganic calcite (the absolute vital effect) *and* with smaller and less carbon limited cells (the differential vital effect).

The reviewer's observations on the conflicting behavior of the small fraction's absolute vital effect and differential vital effect are correct. Although our work originally attempted to use the foraminiferal record (*G. bulloides*) to constrain the absolute vital effect, we concluded that *G. bulloides* cannot be used to derive a reliable inorganic reference against which it would be possible to compare the isotopic changes of the individual size fractions. Indeed, we detail in paragraph 3.2. the uncertainties surrounding the calcification depth and the biogeochemistry of this particular foraminifera (between 70-100m depth according to Rebotim et al., Biogeosciences, 14, 827–859, 2017), which might record changes in seawater chemistry at a different depth from where the coccoliths are biomineralised (*i.e.* predominantly in the uppermost meters of the water column, O'Brien et al. Earth Syst. Sci. Data 5, 259–276, 2013). Therefore, without a reliable inorganic reference for the surface ocean, it is difficult to say which of the 2-3 mm or 5-8 mm fraction is responsible for the observed changes in the $\Delta^{13}C_{small\text{-}large}$ and $\Delta^{18}O_{small\text{-}large}$, a caveat that also captured our frustration. But we hope that our data are sufficiently convincing in that we can work with an offset that does not require an inorganic reference.

Finally, a request. Although there are lovely SEMs of the size splits in the SI, and I'm sure that all the splits look equally lovely, it would help enormously if you could provide some assemblage composition data for some selected representative samples through your record - both species composition and coccolith size distribution. I know this is some extra work, but at the moment it's impossible to properly compare data from coccolith separates like these from different studies (using different methods) unless there's reporting of what is actually being measured. This kind of quantitative assemblage data would also allow better comparisons between studies and across timescales - e.g. knowing that we're comparing small (of defined size range) retics to small retics. Your samples look quite confined in their taxonomic composition - and I'd want to document that (and get others to do the same) - so that we can spot issues if a (90%) *Calcidiscus*-rich assemblage is compared to a (50%) *Calcidiscus*-rich assemblage.

We fully acknowledge the point made by the Reviewer, as it would indeed be ideal to compare the isotopic signals from different sites/studies with strictly comparable coccolith assemblages. We also agree that the assemblage itself, and changes thereof, is a valuable source of information. Unfortunately, we do not have these micropalaeontological data. The rationale of comparing our data with other published datasets relies on the fact that a number of studies (from culture, sediment and numerical experiments) have linked the magnitude of individual coccolithophore vital effects to the degree of carbon limitation experienced by the cell, which to first order depends on the cell's size (and ultimately coccolith size). Thus, it was important that we carefully check our coccolith fractions for signs of (foram/coccolith) fragments that could eventually pollute (as they convey a distinct isotopic signal) the size-restricted fractions that we obtained.

- **Line 29 – would recommend rephrasing, especially the use of "overtakes"**

We will make the recommended changes.

- **Line 63 – lower case "a" after the colon.**

Corrected.

- **Line 64 – late Miocene not Late Miocene – informal division.**

Corrected, thanks for spotting this!